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Quantum Theory of Light and Dispersion Forces in Non-Reciprocal and Bianisotropic Media

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Declaration

I, David Butcher, declare that I am the original author of the work contained in this thesis unless otherwise stated.

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M.C., N.R., G.B., T.D.

Abstract

In this thesis the quantum theory of light in absorbing media, macroscopic quantum electrodynamics, is extended to describe the quantised electromagnetic field in general linear absorbing media. The Casimir-Polder potential between a particle and examples of bianisotropic media are presented.

The electromagnetic properties of a general linear medium that may possess a spatially non-local and even non-reciprocal response are fully described by a complex conductivity tensor. The quantisation of the electromagnetic field in general media was achieved through enforcing a commutation relation between Langevin noise current operators whose presence also ensure compliance with the fluctuation dissipation theorem. The quantisation of the electromagnetic field in media characterised by bianisotropic response functions is shown. The duality invariance of an electromagnetic system was found to be a continuous symmetry in general and a discrete symmetry when the medium has a reciprocal response.

The chiral component of the Casimir-Polder potential was derived in the weak coupling regime. It was found to be either attractive or to repel chiral molecules, depending on the chiral identity of the objects. The molecule and the medium must both possess chiral features otherwise the chiral contribution to the Casimir-Polder potential does not exist. By constructing a cavity between media of opposite chirality, enantiomer separation can occur when particles are initially in an excited state and the resonant frequency of the media is equal to the relevant transition frequency of the molecule.

The quantisation of the electromagnetic field in moving media was achieved on the observation that a locally responding isotropic dielectric in motion is equivalent to a non-reciprocal linear bianisotropic medium from the perspective of a moving frame. A generalised reciprocity condition that constrains the Green's function of the electromagnetic field in moving media was derived.

Thesis Publications

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Chapter 1

Introduction

The behaviour of the electromagnetic field is altered by the presence of material bodies. In the classical formulation of electrodynamics, the effect of the electromagnetic fields interaction with media is to generate additional polarisation and magnetisation fields which become part of the medium assisted electromagnetic fields. However, the classical interpretation could not adequately describe the electrodynamics of quantum objects as it does not take into account phenomena such as the discrete nature of energy at the quantum scale. Although theories of the quantised electromagnetic field in media have existed since the conception of quantum field theories, it was only in the 1990's that the influence of absorption in real materials was fully accounted for. The theories successfully described the electromagnetic fields interaction with inhomogeneous absorbing linear magnetodielectric materials in terms of macroscopic parameters and became known as macroscopic quantum electrodynamics. The dynamics of atoms and molecules are also affected by the presence of media, this can be due to changes in the electromagnetic field which subsequently alter the behaviour of the particle or because of dispersion forces. These are generated by the correlation of quantum fluctuations of the electromagnetic field in the presence of polarisable and magnetisable objects. For example, an atom near a dielectric medium will experience an attractive force that is dependent on the properties of the atom and the properties of the medium.

Modern experimental methods have the capacity to enact a high level of control on atoms at the quantum scale, this means that it is necessary for theory to describe the electromagnetic response of surrounding materials in order to fully account for the interaction between particles and media. Despite the existence of natural materials with unusual optical properties, the recent progress in nanotechnology and metamaterials has opened up the possibility that novel and exotic media can be designed and created to particular specifications. Metamaterials are media that are constructed out of nanoscale constituent parts, generally ordered in specific patterns, to achieve a desired response when an electromagnetic field is applied. For example, a bianisotropic material will generate additional components of the medium assisted fields due to the mixing of electric and magnetic effects when an electromagnetic field is applied. Materials constructed on the nanoscale can exhibit spatially non-local responses to the electromagnetic field, which makes the interaction of media with the electromagnetic field non-trivial. Although the interaction of moving dielectrics with the electromagnetic field has been known to violate time reversal symmetry, the versatility of metamaterials means that materials that lack time reversal symmetry when an electromagnetic field is applied, sometimes referred to as non-reciprocal media, can be constructed.

The potential broad range of effects that media can now exhibit in the presence of electromagnetic fields means that materials can be chosen or tailored for their specific affect on nearby particles. This can lead to better control of particles or the generation of additional components of dispersion forces. In order to describe this novel behaviour, the theory of the electromagnetic field in linear absorbing media

needs the capacity to account for all linear absorbing materials, including non-reciprocal, bianisotropic and spatially non-local media. The initial formulation of macroscopic quantum electrodynamics was limited to simple linear media that could be described by an electric permittivity or a magnetic permeability, this was subsequently extended to include linear reciprocal spatially non-local materials through a conductivity tensor description, therefore the theory needs to be extended to include all linear media within its framework.

This thesis discusses the extension of macroscopic quantum electrodynamics to general media in the linear response regime and the dispersion forces that can be generated from novel materials. Chapter 2 will provide a brief introduction to macroscopic quantum electrodynamics and to a dispersion force between a medium and a particle, the Casimir-Polder force. In chapter 3 macroscopic quantum electrodynamics will be extended to include the quantised electromagnetic field in general linear absorbing media, including non-reciprocal materials. Chapter 4 will introduce magnetoelectric coupling terms to describe general linear bianisotropic media and will explore the duality properties of bianisotropic media. Chapter 5 will present the quantisation of the electromagnetic field in chiral media and will derive the chiral component of the Casimir-Polder potential which will be applied to a chiral molecule in a cavity geometry. In chapter 6 the effect of motion on absorbing linear materials and the extra contribution this provides to the Casimir-Polder potential will be discussed.

Chapter 2

Quantisation of the Electromagnetic Field in Linear Media

James Clerk Maxwell showed in his seminal work [1] that the electromagnetic field could be described by four equations, now known as Maxwell's equations. The development of quantum theory in the 1920's proved that physical quantities were constrained by additional laws governing their behaviour, for example, commutation relations between observables such as position and momentum. In contrast all classical observables commute, although it should be noted that there is a link between classical Poisson brackets and quantum commutation relations. This means that a description of the electromagnetic field that was consistent with quantum theory and with Maxwell's equations for classical fields was required. The successful quantisation, sometimes referred to as second quantisation, of the free space electromagnetic field was achieved and is known as quantum electrodynamics (QED). This quantum field theory has proven very successful and has accurately predicted the behaviour of many quantum systems.

As the understanding and subsequent applications of QED became more sophisticated, the next step was to consider the behaviour of quantised electromagnetic fields in media. To accurately model materials in a field theory, behaviour such as dispersion, spatial inhomogeneity and dissipation need to be included in the description of the media. Although the introduction of dispersion and spatial inhomogeneity are not trivial, it was accounting for the absorption, or dissipation, from the electromagnetic field into media that proved problematic until recently. The issues arose when introducing dissipation after the electromagnetic field had been quantised as this lead to incorrect commutation relations between fields or a thoery without the correct photonic annihilation and creation operators.

To overcome this problem, a quantum theory for the electromagnetic field in homogeneous dielectrics that accounted for the dissipation into the medium was developed [2]. The dielectric medium was modelled by a matter field coupled to a reservoir of harmonic oscillators that governed the absorption. Since then, other models have been developed [3, 4] which successfully quantise the electromagnetic field in absorbing media, where the theory has now been extended to apply to linear inhomogeneous magnetodielectrics. However, in general the links between the fundamental parameters in microscopic models and the macroscopic functions that usually describe a material are not always straightforward. A macroscopic perspective was first explored in Ref. [5] and subsequently in Ref. [6], which presented a quantised theory of the electromagnetic field in absorbing media in terms of macroscopic variables and as such has been called macroscopic QED. This theory complies with Maxwell's equations and so is consistent with classical electrodynamics. The commutation relations between the electromagnetic fields agree with the expected values from quantum theory and compliance with the fluctuation dissipation theorem ensures that the dissipation into the medium obeys the rules of statistical physics. Macroscopic

QED has been successfully applied to dispersion forces, such as the Casimir-Polder force for thermal [7] and non-thermal situations [8], for spontaneous decay [9] and for novel media and geometries [10, 11]. For an overview, see the review article [12].

This chapter will describe the quantisation of the electromagnetic field in linear absorbing media from a macroscopic perspective. Starting from a classical viewpoint with causal macroscopic response functions, the quantisation of the electromagnetic field in linear media will be achieved by introducing bosonic operators of the combined system of the medium and electromagnetic field. Section 2.1.4 will provide a brief discussion of alternative methods to the quantisation of absorbing media. The chapter will finish with an introduction to the dispersion force between a particle and a macroscopic body, the Casimir-Polder force.

2.1 Macroscopic Electrodynamics

The quantisation of the electromagnetic field in absorbing media can be approached from a microscopic or a macroscopic viewpoint. In this section a quantisation procedure in terms of the macroscopic properties of the medium is presented [6]. Starting from the classical Maxwell's equations, the components of a Langevin noise current are added to the polarisation and magnetisation fields [13] to account for the dissipative behaviour of a medium.

2.1.1 Classical Fields

To introduce the quantisation of the electromagnetic field in absorbing media, the starting point are the classical macroscopic Maxwell's equations in frequency space. In the absence of free charges these are

$$\nabla\cdot\mathbf{B}(\mathbf{r},\omega) = 0, \tag{2.1a}$$

$$\nabla\times\mathbf{E}(\mathbf{r},\omega) = i\omega\mathbf{B}(\mathbf{r},\omega), \tag{2.1b}$$

$$\nabla\cdot\mathbf{D}(\mathbf{r},\omega) = 0, \tag{2.1c}$$

$$\nabla\times\mathbf{H}(\mathbf{r},\omega) = -i\omega\mathbf{D}(\mathbf{r},\omega). \tag{2.1d}$$

It has been assumed that the fields behave in a time harmonic manner and so a time derivative acts on the fields as

$$\frac{\partial}{\partial t}\mathbf{F}(\mathbf{r},\omega) = -i\omega\mathbf{F}(\mathbf{r},\omega). \tag{2.2}$$

Taking this into consideration, the time harmonic term, $e^{-i\omega t}$, will be assumed but the notation suppressed from now on. The matter induced fields are the displacement field, $\mathbf{D}(\mathbf{r},\omega)$, and the magnetic field $\mathbf{H}(\mathbf{r},\omega)$. These are related to the applied electric field, $\mathbf{E}(\mathbf{r},\omega)$, and applied magnetic induction field, $\mathbf{B}(\mathbf{r},\omega)$, by the constitutive relations

$$\mathbf{D}(\mathbf{r},\omega) = \varepsilon_0\mathbf{E}(\mathbf{r},\omega) + \mathbf{P}(\mathbf{r},\omega), \tag{2.3a}$$

$$\mathbf{H}(\mathbf{r},\omega) = \frac{1}{\mu_0}\mathbf{B}(\mathbf{r},\omega) - \mathbf{M}(\mathbf{r},\omega), \tag{2.3b}$$

where $\mathbf{P}(\mathbf{r},\omega)$ and $\mathbf{M}(\mathbf{r},\omega)$ are the polarisation and magnetisation fields, respectively. These are defined as

$$\mathbf{P}(\mathbf{r},t) = \varepsilon_0\int_0^\infty \mathrm{d}\tau\,\boldsymbol{\chi}_{ee}(\mathbf{r},\tau)\cdot\mathbf{E}(\mathbf{r},t-\tau) + \mathbf{P}_\mathrm{N}(\mathbf{r},t), \tag{2.4a}$$

$$\mathbf{M}(\mathbf{r},t) = \frac{1}{\mu_0}\int_0^\infty \mathrm{d}\tau\, \boldsymbol{\chi}_{mm}(\mathbf{r},\tau)\cdot\mathbf{B}(\mathbf{r},t-\tau) - \mathbf{M}_\mathrm{N}(\mathbf{r},t), \tag{2.4b}$$

where $\boldsymbol{\chi}_{ee}(\mathbf{r},\tau)$ and $\boldsymbol{\chi}_{mm}(\mathbf{r},\tau)$ are the electric and magnetic susceptibilities for an inhomogeneous medium with a local spatial response, which is discussed in chapter 3. The noise polarisation field, $\mathbf{P}_\mathrm{N}(\mathbf{r},\omega)$, and the noise magnetisation field, $\mathbf{M}_\mathrm{N}(\mathbf{r},\omega)$ are Langevin noise fields and they are associated with the absorption into the medium. They have been introduced to ensure that the fluctuation dissipation theorem is always fulfilled and to ensure that the commutation relations between the quantised fields are consistent with the values obtained from free space electromagnetic field quantisation, as discussed in section 2.1.2.

Although a medium can be described in terms of the susceptibilities, the electric permittivity, $\boldsymbol{\varepsilon}(\mathbf{r},\omega)$, and magnetic permeability, $\boldsymbol{\mu}(\mathbf{r},\omega)$, are commonly used in their place. These are defined, in Fourier space, as

$$\boldsymbol{\varepsilon}(\mathbf{r},\omega) = \boldsymbol{I} + \boldsymbol{\chi}_{ee}(\mathbf{r},\omega), \tag{2.5a}$$

$$\boldsymbol{\mu}^{-1}(\mathbf{r},\omega) = \boldsymbol{I} - \boldsymbol{\chi}_{mm}(\mathbf{r},\omega). \tag{2.5b}$$

This means that the polarisation and magnetisation can be rewritten in the form

$$\mathbf{P}(\mathbf{r},\omega) = \varepsilon_0[\boldsymbol{\varepsilon}(\mathbf{r},\omega) - \boldsymbol{I}]\cdot\mathbf{E}(\mathbf{r},\omega) + \mathbf{P}_\mathrm{N}(\mathbf{r},\omega), \tag{2.6a}$$

$$\mathbf{M}(\mathbf{r},\omega) = \frac{1}{\mu_0}[\boldsymbol{I} - \boldsymbol{\mu}^{-1}(\mathbf{r},\omega)]\cdot\mathbf{B}(\mathbf{r},\omega) - \mathbf{M}_\mathrm{N}(\mathbf{r},\omega). \tag{2.6b}$$

The term $\boldsymbol{I}$ refers to the identity matrix. The response functions, $\boldsymbol{\varepsilon}(\mathbf{r},\omega)$ and $\boldsymbol{\mu}(\mathbf{r},\omega)$, are complex functions of frequency,

$$\boldsymbol{\varepsilon}(\mathbf{r},\omega) = \mathrm{Re}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)] + i\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)], \tag{2.7a}$$

$$\boldsymbol{\mu}(\mathbf{r},\omega) = \mathrm{Re}[\boldsymbol{\mu}(\mathbf{r},\omega)] + i\mathrm{Im}[\boldsymbol{\mu}(\mathbf{r},\omega)]. \tag{2.7b}$$

The real and imaginary parts of these response functions are related to the reactive and dissipative properties of the medium, respectively, when external electromagnetic fields are applied. It shall be assumed that all the media considered throughout this thesis will exhibit loss, this means that the imaginary parts of the response functions are positive, i.e., $\mathrm{Im}[\boldsymbol{\varepsilon}] > 0$ and $\mathrm{Im}[\boldsymbol{\mu}] > 0$. The response functions must comply with the causality requirement that for $t < 0$, $\boldsymbol{\varepsilon}(t), \boldsymbol{\mu}(t) = \mathbf{0}$. This is simply a mathematical statement that the polarisation and magnetisation at time t can not depend on an electric field at later times. An interesting implication of compliance with the causality condition is that the real and imaginary parts of a causal response function are connected by Kramers - Kronig relations, which are derived in Appendix A. These show that a non-zero real part of a response function will necessarily result in a non-zero imaginary part. Although generally the Kramers - Kronig relations do not guarantee that the imaginary parts are positive, for the media in this thesis it can be said that absorption is required for the medium to be causal.

By combining Maxwell's equations, Eqs. (2.1), with the constitutive relations, Eqs. (2.3), and the polarisation and magnetisation, Eqs. (2.6), it can be seen that the electric field satisfies the inhomogeneous Helmholtz equation

$$\left[\nabla\times\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)\cdot\nabla\times\boldsymbol{I} - \frac{\omega^2}{c^2}\boldsymbol{\varepsilon}(\mathbf{r},\omega)\right]\cdot\mathbf{E}(\mathbf{r},\omega) = i\omega\mu_0\mathbf{j}_\mathrm{N}(\mathbf{r},\omega), \tag{2.8}$$

where $\mathbf{j}_\mathrm{N}(\mathbf{r},\omega)$ is the noise current density, defined as

$$\mathbf{j}_\mathrm{N}(\mathbf{r},\omega) = -i\omega\mathbf{P}_\mathrm{N}(\mathbf{r},\omega) + \nabla\times\mathbf{M}_\mathrm{N}(\mathbf{r},\omega). \tag{2.9}$$

The noise current obeys the continuity equation

$$\frac{\partial \rho_{\mathrm{N}}}{\partial t} + \nabla \cdot \mathbf{j}_{\mathrm{N}} = 0, \tag{2.10}$$

where the noise charge density, ρ_{N}, is defined as the divergence of the noise polarisation field

$$\rho_{\mathrm{N}}(\mathbf{r}, \omega) = -\nabla \cdot \mathbf{P}_{\mathrm{N}}(\mathbf{r}, \omega). \tag{2.11}$$

It is important to note that the separation of the noise current into $\mathbf{P}_{\mathrm{N}}(\mathbf{r}, \omega)$ and $\mathbf{M}_{\mathrm{N}}(\mathbf{r}, \omega)$ is not necessarily well defined for all classes of media, as magnetic effects can always be absorbed into the transverse part of the noise polarisation field. This will be expanded upon in later chapters. The inhomogeneous wave equation for the electric field, Eq. (2.8), has a similar form to that which arises when there is an external electromagnetic source current. However, in this instance it is the noise current that takes the place of a source term. This means that the noise current field is acting as a source term for the electromagnetic fields due to the absorption into the medium. To solve Eq. (2.8), the Green's function, $\boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega)$, is used to write

$$\mathbf{E}(\mathbf{r}, \omega) = i\omega\mu_0 \int \mathrm{d}^3 r' \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{j}_{\mathrm{N}}(\mathbf{r}', \omega). \tag{2.12}$$

The Green's function is the fundamental solution to the vector Helmholtz equation,

$$\left[\nabla \times \boldsymbol{\mu}^{-1}(\mathbf{r}, \omega) \cdot \nabla \times \boldsymbol{I} - \frac{\omega^2}{c^2} \boldsymbol{\varepsilon}(\mathbf{r}, \omega)\right] \cdot \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{\delta}(\mathbf{r} - \mathbf{r}'), \tag{2.13}$$

where

$$\boldsymbol{\delta}(\mathbf{r} - \mathbf{r}') = \boldsymbol{I} \delta^{(3)}(\mathbf{r} - \mathbf{r}'). \tag{2.14}$$

To ensure that the electric field is physical, the Green's function is constrained by the boundary condition that as $|\mathbf{r} - \mathbf{r}'| \to \infty$, $\boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{0}$, which guarantees a unique Green's function. As a function of ω, the Green's function is an analytic function in the upper half plane. In the temporal domain, $\boldsymbol{G}(\mathbf{r}, \mathbf{r}', t)$ is a real function and therefore the Schwarz reflection principle

$$\boldsymbol{G}^*(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{G}(\mathbf{r}, \mathbf{r}', -\omega^*), \tag{2.15}$$

always holds, see Appendix B for a derivation. The Green's function must also be causal, so $\boldsymbol{G}(\mathbf{r}, \mathbf{r}', t) = \mathbf{0}$ for $t < 0$. An important property of the Green's function is the reciprocity condition,

$$\boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{G}^{\mathrm{T}}(\mathbf{r}', \mathbf{r}, \omega). \tag{2.16}$$

which is derived in Appendix C. This is true for all $\mathbf{r}$ and $\mathbf{r}'$ and it means that the propagation of the electromagnetic field in the medium obeys time symmetry. The implication of relaxing the reciprocity condition is the subject of chapter 3 where it will be discussed in more detail.

2.1.2 Langevin Noise Operators

In order to give a clearer picture of the formalism underpinning macroscopic QED, a brief discussion on the origin of the Langevin noise terms is necessary, which is based on material in Ref. [14].

It is initially assumed that a system under investigation is surrounded by an environment to which it is weakly coupled. The environment has a large number of degrees of freedom and therefore it can be said to have a continuum of potential states. A consequence of the weak coupling between the system

and the environment is that a finite disturbance of the system is caused by a large number of infinitesimal disturbances in the environment. The system can be considered as a dynamic system and the environment as a dissipative system or reservoir. As an example, a single mode optical field is considered and can be described by

$$\hat{a}(t) = \hat{a}(t')e^{[-(i\omega+\frac{1}{2}\Gamma)(t-t')]}, \tag{2.17}$$

where Γ is a damping term and $t - t' \geq 0$. The commutation relation is

$$[\hat{a}(t), \hat{a}^\dagger(t)] \propto e^{-\Gamma(t-t')}, \tag{2.18}$$

which approaches zero as t and t' diverge and therefore violates the uncertainty principle, this can be seen from the Robertson uncertainty relation

$$\Delta\hat{a}(t)\Delta\hat{a}^\dagger(t) \geq \frac{1}{2}|\langle[\hat{a}(t), \hat{a}^\dagger(t)]\rangle| \tag{2.19}$$

where $\Delta\hat{a}(t) = \sqrt{\langle\hat{a}^2(t)\rangle - \langle\hat{a}(t)\rangle}$. This erroneous result is because the fluctuations of the environment, which affects the noise of the system, have been ignored. To correct the result, the quantum Langevin equation is introduced

$$\dot{\hat{a}} = -(i\omega + \frac{1}{2}\Gamma)\hat{a} + \hat{f}(t), \tag{2.20}$$

where $\hat{f}(t)$ is an operator valued Langevin noise source. The solution to Eq. (2.20) is

$$\hat{a}(t) = \hat{a}(t')e^{[-(i\omega+\frac{1}{2}\Gamma)(t-t')]} + \int_{t'}^{t} \mathrm{d}\tau \hat{f}(\tau)e^{[-(i\omega+\frac{1}{2}\Gamma)(t-\tau)]}, \tag{2.21}$$

where t' is an arbitrary intial time and $t - t' \geq 0$ still holds. By assuming that the correct commutation relation holds at an initial time, t', the commutation relation in general is now

$$[\hat{a}(t), \hat{a}^\dagger(t)] = 1, \tag{2.22}$$

as required. This is found by the assumptions that

$$[\hat{a}(t_1), \hat{f}^\dagger(t_2)] = 0 \quad t_2 > t_1, \tag{2.23a}$$

$$[\hat{f}(t_1), \hat{f}^\dagger(t_2)]e^{i\omega(t_1-t_2)} = \Gamma\delta(t_1 - t_2). \tag{2.23b}$$

The term $\hat{f}(t)$ can be considered as a noise term if $\langle\hat{f}(t)\rangle = 0$ as this shows it has no coherence. In this section, the Langevin noise term was simply included and was assumed to behave in a way that ensured the consistency of the commutation relations. However, Langevin noise terms can be derived from open systems quantum theory where they are represented by the initial conditions of the operators of the reservoir [15]. In the formalism of macroscopic QED, the introduction of Langevin noise terms has the effect of coupling the system to a large dissipative environment. The associated fluctuations are then described by the Langevin noise operators, which are dependent on the initial conditions of the environment.

2.1.3 Field Quantisation

In section 2.1.1 the system was described in terms of the classical electromagnetic fields. To quantise the electromagnetic field in linear dissipative media, bosonic quantum operators of a combined matter-field system need to be introduced. It should be noted that these operators are not excitations of the matter

field or the electromagnetic fields separately, they are only excitations of the combined matter-field system. Although not necessary, here it is assumed that there are two types of quantum operator, one associated with electric excitations, $\hat{\mathbf{f}}_e(\mathbf{r},\omega)$, and another associated with magnetic excitations, $\hat{\mathbf{f}}_m(\mathbf{r},\omega)$. The electromagnetic field in the medium is then quantised by proposing that the noise polarisation and noise magnetisation fields are related to these matter-field system excitations through the imaginary parts of the response functions,

$$\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar\varepsilon_0}{\pi}\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)]}\hat{\mathbf{f}}_e(\mathbf{r},\omega), \tag{2.24a}$$

$$\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega) = \sqrt{-\frac{\hbar}{\mu_0\pi}\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)]}\hat{\mathbf{f}}_m(\mathbf{r},\omega). \tag{2.24b}$$

The square roots of the tensors, $\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)]$ and $\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)]$, can be found by considering the square root of a matrix. There exists matrices, $\boldsymbol{U}$, such that a diagonal matrix of the eigenvalues of the tensors, $\boldsymbol{D}_\varepsilon$ and $\boldsymbol{D}_\mu$, can be defined as

$$\mathrm{Im}[\boldsymbol{\varepsilon}] = \boldsymbol{U}\boldsymbol{D}_\varepsilon\boldsymbol{U}^{-1}, \tag{2.25a}$$

$$\mathrm{Im}[\boldsymbol{\mu}] = \boldsymbol{U}\boldsymbol{D}_\mu\boldsymbol{U}^{-1}. \tag{2.25b}$$

The unique square root of the diagonal matrices are $(\boldsymbol{D}_\varepsilon)^{\frac{1}{2}}$ and $(\boldsymbol{D}_\mu)^{\frac{1}{2}}$, which means that the square root of the tensors are

$$\sqrt{\mathrm{Im}[\boldsymbol{\varepsilon}]} = \boldsymbol{U}(\boldsymbol{D}_\varepsilon)^{\frac{1}{2}}\boldsymbol{U}^{-1}, \tag{2.26a}$$

$$\sqrt{\mathrm{Im}[\boldsymbol{\mu}^{-1}]} = \boldsymbol{U}(\boldsymbol{D}_\mu)^{\frac{1}{2}}\boldsymbol{U}^{-1}. \tag{2.26b}$$

To complete the quantisation procedure, a commutation relation between the system excitations is imposed

$$\big[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega), \hat{\mathbf{f}}^\dagger_{\lambda'}(\mathbf{r}',\omega')\big] = \delta_{\lambda\lambda'}\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'), \tag{2.27}$$

where $\lambda,\lambda' = e,m$. The ground state of the matter-field system, $|\{0\}\rangle$, is defined through

$$\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)|\{0\}\rangle = \mathbf{0} \quad \text{for all } \lambda,\mathbf{r},\omega. \tag{2.28}$$

To ensure the correct time evolution the constraint

$$[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega), \hat{H}_F] = \hbar\omega\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega), \tag{2.29}$$

is enforced on a Hamiltonian of the matter-field system, which is constructed from the system excitations,

$$\hat{H}_F = \sum_{\lambda=e,m}\int \mathrm{d}^3r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega). \tag{2.30}$$

The definitions given in Eqs. (2.24a) and (2.24b) can be used to rewrite the electric and magnetic induction fields in terms of the matter-field excitations

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = \sum_{\lambda=e,m}\int \mathrm{d}^3r'\boldsymbol{G}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r}',\omega), \tag{2.31a}$$

$$\hat{\mathbf{B}}(\mathbf{r},\omega) = \frac{1}{i\omega}\sum_{\lambda=e,m}\int \mathrm{d}^3r'\nabla\times\boldsymbol{G}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r}',\omega). \tag{2.31b}$$

The $\boldsymbol{G}_\lambda(\mathbf{r},\mathbf{r}',\omega)$ expressions have been introduced as notational shorthands for

$$\boldsymbol{G}_e(\mathbf{r},\mathbf{r}',\omega) = i\frac{\omega^2}{c^2}\sqrt{\frac{\hbar}{\pi\varepsilon_0}\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r}',\omega)]}\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega), \tag{2.32a}$$

$$\boldsymbol{G}_m(\mathbf{r},\mathbf{r}',\omega) = -i\frac{\omega}{c}\sqrt{-\frac{\hbar}{\varepsilon_0\pi}\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r}',\omega)]}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\nabla}']. \tag{2.32b}$$

The operation $\times\overleftarrow{\nabla}'$ is the derivative acting towards the left of the tensor, onto the second spatial variable. In tensor notation this is shown as

$$[\boldsymbol{T}\times\overleftarrow{\nabla}']_{ij}(\mathbf{r},\mathbf{r}') = \epsilon_{jkl}\partial'_l T_{ik}(\mathbf{r},\mathbf{r}'). \tag{2.33}$$

In Appendix D the derivation of an integral relation, which connects the fluctuations of the system with the dissipation into the medium, is shown. In terms of the notational shorthands, Eqs. (2.32a) and (2.32b), this can be expressed as

$$\sum_{\lambda=e,m}\int \mathrm{d}^3 s\boldsymbol{G}_\lambda(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}^\dagger_\lambda(\mathbf{r}',\mathbf{s},\omega) = \frac{\hbar\mu_0\omega^2}{\pi}\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)]. \tag{2.34}$$

The electric and magnetic induction field operators can be written in the Schrodinger picture as,

$$\hat{\mathbf{E}}(\mathbf{r}) = \int_0^\infty \mathrm{d}\omega\hat{\mathbf{E}}(\mathbf{r},\omega) + \mathrm{H.c.}, \tag{2.35a}$$

$$\hat{\mathbf{B}}(\mathbf{r}) = \int_0^\infty \mathrm{d}\omega\hat{\mathbf{B}}(\mathbf{r},\omega) + \mathrm{H.c.}, \tag{2.35b}$$

by integrating over their frequency components. To be compliant with the fluctuation dissipation theorem, which links the strength of the field fluctuations to the medium's dissipative response to applied electromagnetic fields, the vacuum field fluctuations must be non-zero. The average vacuum fluctuations of the electric field are zero,

$$\langle\{0\}|\hat{\mathbf{E}}(\mathbf{r})|\{0\}\rangle = \langle\hat{\mathbf{E}}(\mathbf{r})\rangle = 0, \tag{2.36}$$

but the average of the electric field correlations,

$$\langle\hat{\mathbf{E}}^2(\mathbf{r})\rangle = \langle\hat{\mathbf{E}}(\mathbf{r})\hat{\mathbf{E}}(\mathbf{r}')\rangle = \frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\omega\ \omega^2\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)], \tag{2.37}$$

is non-zero. This means the vacuum field fluctuations are

$$\langle|\Delta\hat{\mathbf{E}}(\mathbf{r})|^2\rangle = \langle\hat{\mathbf{E}}^2(\mathbf{r})\rangle - \Big(\langle\hat{\mathbf{E}}(\mathbf{r})\rangle\Big)^2 = \frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\omega\ \omega^2\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)], \tag{2.38}$$

as is required by the fluctuation dissipation theorem. Furthermore, the equal time field commutators return the correct free space values. To obtain these commutation relations, the forms of the electric field, Eq. (2.31a), and magnetic induction field, Eq. (2.31b), are used in conjunction with Eqs. (2.35a) and (2.35b), respectively. With use of the integral relation, Eq. (2.34), this leads to

$$[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')] = \frac{i\hbar\mu_0}{\pi}\nabla_{\mathbf{r}'}\times\int_0^\infty \mathrm{d}\omega\,\omega\bigg(\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)] + \mathrm{Im}[\boldsymbol{G}(\mathbf{r}',\mathbf{r},\omega)]\bigg). \tag{2.39}$$

Using the definition of the imaginary part of the Green's function ($\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)] = \frac{1}{2i}(\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) - \boldsymbol{G}^\dagger(\mathbf{r}',\mathbf{r},\omega))$), the Green's functions analytic properties in the frequency upper half plane and the be-

haviour

$$\lim_{|\omega|\to\infty} \frac{\omega^2}{c^2}\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = -\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'), \tag{2.40}$$

which can be seen from Eq. (2.13) by noting that $\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)$, $\boldsymbol{\varepsilon}(\mathbf{r},\omega) \to 1$ as $\omega \to \infty$, the commutation relation can be written as

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{B}}(\mathbf{r}')] = -\frac{i\hbar}{\varepsilon_0}\nabla' \times \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'). \tag{2.41}$$

Through the same method,

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{E}}(\mathbf{r}')] = [\hat{\mathbf{B}}(\mathbf{r}), \hat{\mathbf{B}}(\mathbf{r}')] = 0, \tag{2.42}$$

can be found. Thus, we have shown that the Langevin noise approach to electromagnetic field quantisation in magnetodielectric media is consistent with the requirements from statistical physics and quantum theory. This section has briefly detailed the quantisation of the electromagnetic field in an inhomogeneous absorbing medium in terms of macroscopic response functions. The next section will discuss some other quantisation procedures.

2.1.4 Alternative Approaches to Field Quantisation

The first quantisation of the electromagnetic field in absorbing media applied to homogeneous absorbing dielectrics, the quantisation procedure involved the coupling of applied electromagnetic fields to a matter field which itself is coupled to a reservoir modelled by a continuum of harmonic oscillators [2]. At this stage the field quantisation can be achieved and the matter-field system excitations, which obey a commutation relation and define a Hamiltonian, can be found through a series of transformations and a diagonalisation. A recent extension to this can be found in Ref. [16]. A different method for the quantisation of the electromagnetic field in an absorbing dielectric is the inclusion of auxiliary fields [17], which has been shown to be equivalent to the Langevin noise approach [18].

By introducing a spatially dependent coupling term between the electric field and the harmonic oscillator field, this model was extended to include inhomogeneous dielectric absorbing media [3]. The subsequent addition of an extra reservoir allowed the theory to include any linear magnetodielectric absorbing media [4], recently expanded to magnetoelectric media [19]. The quantisation of the electromagnetic field in linear magnetodielectric media through a mode expansion procedure has also been achieved [20].

The results obtained from the perspective of the microscopic fields agree with those found in macroscopic QED. However, when the theory is in terms of the microscopic parameters it can become a difficult exercise to relate these quantities to the macroscopic properties that usually describe media, although it has been shown that it is possible to include the macroscopic parameters from the beginning of the quantisation process [4]. This thesis will be concerned exclusively with the macroscopic viewpoint.

2.2 Dispersion Forces

In the introduction to this chapter a fruitful application of macroscopic QED was alluded to, the investigation of dispersion forces between particles and absorbing bodies or between particles in the presence of absorbing bodies. Dispersion forces are quantum forces that arise between polarisable and magnetisable objects and are generated by the correlation of quantum fluctuations of the electromagnetic field. These forces can be classified according to various criteria, here the classification is made with respect to the objects involved in the interaction. This means that there are three forms of the dispersion forces; Casimir forces, Casimir-Polder forces and Van der Waals forces. Casimir forces are usually between two bodies that can be considered macroscopic, H. B. G. Casimir initially considered conducting plates [21],

and the most recent measurements of the Casimir force have used gold hemispheres near a plate [22]. Casimir-Polder forces tend to be between a macroscopic body and a particle [23], as shown in Fig. 2.2, and were first measured by detecting the deflection of Sodium atoms passing through a cavity [24]. The dispersion forces between particles are normally refered to as Van der Waals forces. In its most general form the Casimir-Polder force can be time-dependent, affected by finite temperatures and affected by the initial state of the matter-field system, which is not necessarily the ground state. All the calculations of the Casimir-Polder force in this thesis will only be concerned with the zero temperature regime, the matter-field system will be in its ground state and a static approach with respect to time shall be taken. This means that thermal fluctuations, the dynamics of the matter-field system and the decay and lifetimes of the atomic or molecular excited states are neglected. Initially the effects of motion have also been discounted, but this will be returned to in Chapter 6. For an extensive overview of Casimir - Polder forces, see Refs. [25, 26] and the references therein.

2.2.1 Casimir-Polder Potential

The following derivation of the Casimir-Polder potential is based on the procedure given in Ref. [12]. It is assumed that the coupling between the atom and the electromagnetic field is weak which allows a perturbative approach to be taken and the interactions are described in the multipolar coupling framework [27]. The Casimir-Polder force is

$$\hat{\mathbf{F}}_{\mathrm{CP}}(\mathbf{r}_A) = -\nabla U_{\mathrm{CP}}(\mathbf{r}_A), \tag{2.43}$$

where the Casimir-Polder potential can be viewed as the position dependent part of the energy shift, $U_{\mathrm{CP}}(\mathbf{r}_A) = \Delta E(\mathbf{r}_A)$. The subscript A on the position vector, $\mathbf{r}_A$, refers to the position of atom (or molecule). Discarding higher order contributions such as the diamagnetic interaction [25] and assuming that the atom to body distance is large compared to the atomic radius (which allows the use of the long wavelength approximation) the atom - field interaction Hamiltonian is

$$\hat{H}_{AF} = -\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A) - \hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A). \tag{2.44}$$

To calculate the ground state energy shift due to the Casimir-Polder force, 2nd order perturbation theory is used,

$$\Delta E = \sum_{I\neq N} \frac{\langle N|\hat{H}_{AF}|I\rangle\langle I|\hat{H}_{AF}|N\rangle}{E_N - E_I}, \tag{2.45}$$

where $|N\rangle = |n\rangle|\{0\}\rangle$ represents the initial state of the $\hat{H}_F + \hat{H}_{AF}$ system. The subsequent intermediate states are

$$|I\rangle = |k\rangle|\mathbf{1}_\lambda(\mathbf{r},\omega)\rangle, \quad |\mathbf{1}_\lambda(\mathbf{r},\omega)\rangle = \hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)|\{0\}\rangle. \tag{2.46}$$

The term $\hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)$ is a creation operator that acts on the matter - field system state $|\{0\}\rangle$. The energies of the states can be written in terms of frequencies, where $E_N = \hbar\omega_n$ and $E_I = \hbar(\omega_k + \omega)$. The initial state of the particle, $|n\rangle$, is not necessarily the ground state of the particle and so could be an excited state.

Equation (2.44) is substituted into Eq. (2.45), which results in

$$\begin{aligned}\Delta E = \sum_{I\neq N}\Big(&\langle N|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|N\rangle + \langle N|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|N\rangle \\ &+ \langle N|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|N\rangle + \langle N|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|N\rangle\Big)\Big/\Big(E_N - E_I\Big).\end{aligned} \tag{2.47}$$

The Casimir-Polder potential can be split into electric, magnetic and magnetoelectric components. The

demarcation is based on which terms contain only the electric dipole moments, only the magnetic dipole moments or a combination of both. This means that the electric and magnetic components arise from the terms

$$\langle N|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|N\rangle,$$

$$\langle N|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|N\rangle,$$

respectively. The remaining terms

$$\langle N|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|N\rangle,\ \langle N|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|N\rangle$$

are the magnetoelectric contributions to the Casimir-Polder potential. These cross terms are usually neglected because for an achiral particle, $\hat{\mathbf{d}}$ and $\hat{\mathbf{m}}$ have opposite parity and there are no intermediate states that can be reached by both an electric dipole transition and a magnetic dipole transition [25]. However, there are exceptions when the magnetoelectric contributions do not disappear, such as the Casimir-Polder potential between a chiral molecule near a chiral medium, which is the subject of section 5.3. For the present example it is assumed that the atom is achiral, so the magnetoelectric terms are neglected.

By substituting the expansion of the electric field, Eq. (2.31a), into Eq. (2.35a) and the expansion of the magnetic induction field, Eq. (2.31b), into Eq. (2.35b), the relevant matrix elements can be expressed as

$$\langle N|\ \hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle = \mathbf{d}_{nk}\cdot\boldsymbol{G}_\lambda(\mathbf{r}_A,\mathbf{r},\omega) \tag{2.48a}$$

$$\langle N|\ \hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle = \frac{\mathbf{m}_{nk}\cdot\nabla_A\times\boldsymbol{G}_\lambda(\mathbf{r}_A,\mathbf{r},\omega)}{i\omega} \tag{2.48b}$$

where $\mathbf{d}_{nk} = \langle n|\hat{\mathbf{d}}|k\rangle$ and $\mathbf{m}_{nk} = \langle n|\hat{\mathbf{m}}|k\rangle$. The formal sum in Eq. (2.47) (and Eq. (2.45)) is actually a representation of a sum over the modes λ and a sum over the atom's intermediate states k and it contains integrals over space and frequency,

$$\sum_{I\neq N} \to \sum_\lambda \sum_k \int \mathrm{d}^3r \int_0^\infty \mathrm{d}\omega. \tag{2.49}$$

Therefore substituting Eqs. (2.48a) and (2.48b) into Eq. (2.47) leads to

$$\begin{aligned}\Delta E = -\frac{\mu_0}{\pi}\sum_k \mathcal{P}\int_0^\infty \frac{\mathrm{d}\omega}{\omega_{kn}+\omega}\Big[&\omega^2\ \mathbf{d}_{nk}\cdot\mathrm{Im}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega)]\cdot\mathbf{d}_{kn}\\ &+\mathbf{m}_{nk}\cdot\nabla\times\mathrm{Im}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega)]\times\overleftarrow{\nabla}'\cdot\mathbf{m}_{kn}\Big],\end{aligned} \tag{2.50}$$

where the integral relation as shown in Eq. (2.34) has been used, the atomic transition frequencies are defined as $\omega_{kn} = \omega_k - \omega_n$ and $\mathcal{P}$ denotes a Cauchy principle value.

The integral in Eq. (2.50) is dependent upon the imaginary part of the Green's function, which is not analytic in the frequency plane although the Green's function is analytic in the upper half plane. This means that by rewriting the imaginary part of the scattering Green's function in terms of the Green's function ($\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)] = \frac{1}{2i}(\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) - \boldsymbol{G}^\dagger(\mathbf{r}',\mathbf{r},\omega))$), then using the Schwarz reflection principle Eq. (2.15), equation (2.50) can be recast into a form where a contour integral with frequency can be performed, pictorially represented in Fig. 2.1. The full Green's function comprises a position independent bulk part, $\boldsymbol{G}^{(0)}(\mathbf{r}_A,\mathbf{r}_A,\omega)$, and a position dependent scattering part, $\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega)$, discussed in Appendix E. The bulk part of the Green's function can be discarded because it does not contribute to the Casimir-Polder potential, leaving just the scattering part of the Green's function. To perform the

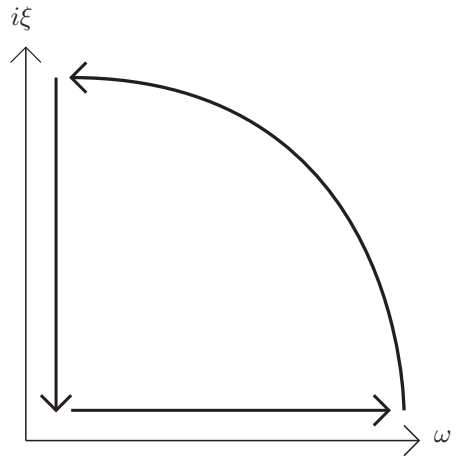


Figure 2.1: This diagram shows the path taken when a contour integration with frequency is performed after Eq. (2.50) is rewritten in terms of the Green's function. As can be seen in Eq. (2.51), the arc between the two axis is zero in this situation. This means that the contribution to the contour from the real freqeuncy axis must be equal to the contribution from the imaginary frequency axis plus the residue, which arises when there are singularities.

contour integration the scattering part of the Green's functions behaviour in the limit of large frequency is needed,

$$\lim_{|\omega|\to\infty} \frac{\omega^2}{c^2}\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega) = \mathbf{0}, \tag{2.51}$$

which can be seen from the homogeneous form of Eq. (2.13) and by noting that $\boldsymbol{\varepsilon}(\mathbf{r},\omega)\to 1$ as $\omega\to\infty$. This means that the infinite arc section of the contour between the real and imaginary frequency ($\omega\to i\xi$) axis is zero.

Writing the Casimir-Polder potential in terms of an electric, $U_e(\mathbf{r}_A)$, and a magnetic component, $U_m(\mathbf{r}_A)$, where

$$U(\mathbf{r}_A) = U_e(\mathbf{r}_A) + U_m(\mathbf{r}_A), \tag{2.52}$$

and performing the integral leads to components of the Casimir-Polder potential for a particle initially in state n of

$$\begin{aligned} U_e^n(\mathbf{r}_A) =& \frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\ \xi^2 \mathrm{tr}[\boldsymbol{\alpha}^n(i\xi)\cdot\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)] \\ & -\mu_0\sum_k \Theta(\omega_{nk})\omega_{nk}^2\mathrm{tr}[(\mathbf{d}_{kn}\otimes\mathbf{d}_{nk})\cdot\mathrm{Re}[\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]], \end{aligned} \tag{2.53a}$$

$$\begin{aligned} U_m^n(\mathbf{r}_A) =& \frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\ \mathrm{tr}[\boldsymbol{\beta}^n(i\xi)\cdot\nabla\times\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)\times\overleftarrow{\nabla}'] \\ & +\mu_0\sum_k \Theta(\omega_{nk})\mathrm{tr}[(\mathbf{m}_{kn}\otimes\mathbf{m}_{nk})\cdot\nabla\times\mathrm{Re}[\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\times\overleftarrow{\nabla}'], \end{aligned} \tag{2.53b}$$

where $\boldsymbol{\alpha}^n(i\xi)$, the electric atomic polarisability, and $\boldsymbol{\beta}^n(i\xi)$, the magnetic atomic polarisability, for an initial particle state n are defined as

$$\boldsymbol{\alpha}^n(i\xi) = \frac{1}{\hbar}\sum_k\left(\frac{\mathbf{d}_{kn}\otimes\mathbf{d}_{nk}}{\omega_{kn}+i\xi} + \frac{\mathbf{d}_{kn}\otimes\mathbf{d}_{nk}}{\omega_{kn}-i\xi}\right), \tag{2.54a}$$

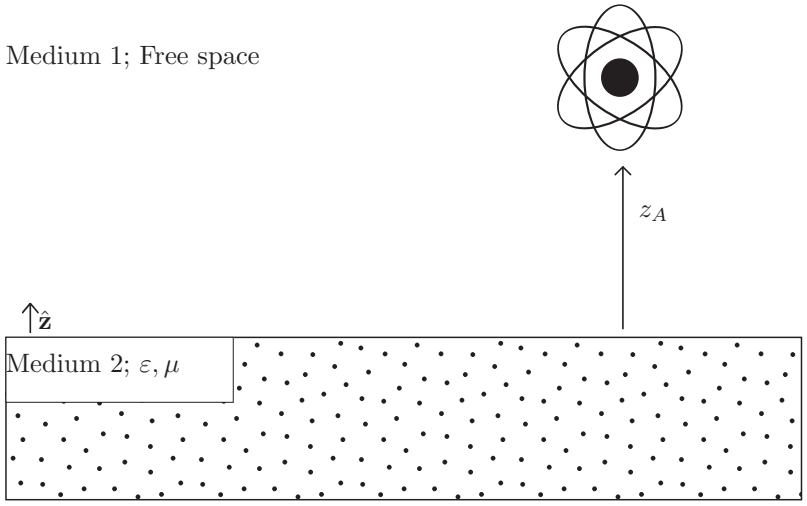


Figure 2.2: This diagram shows the Casimir-Polder force between a particle in free space near the boundary with a magnetodielectric medium.

$$\boldsymbol{\beta}^n(i\xi) = \frac{1}{\hbar}\sum_k \left(\frac{\mathbf{m}_{kn}\otimes\mathbf{m}_{nk}}{\omega_{kn}+i\xi} + \frac{\mathbf{m}_{kn}\otimes\mathbf{m}_{nk}}{\omega_{kn}-i\xi} \right). \tag{2.54b}$$

Here tr[] refers to the trace and $\mathrm{Re}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)] = \frac{1}{2}(\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) + \boldsymbol{G}^\dagger(\mathbf{r}',\mathbf{r},\omega))$ is the real part of the Green's function. The first terms on the right hand sides in Eqs. (2.53a) and (2.53b) are the off-resonant components of the Casimir-Polder potential and the second terms on the right hand sides of Eqs. (2.53a) and (2.53b) are the resonant contributions. The resonant contributions are proportional to the residue at the poles and they only occur when the particle involved in the interaction transitions from an excited state to a lower state. The function $\Theta(\omega_{nk})$ is the Heaviside step function, it ensures that the resonant terms disappear and therefore do not contribute when there is a transition to a higher state. For example, when an atom initially in its ground state interacts with a medium, the resultant Casimir-Polder potential will not include a resonant contribution. The derivations in Eqs. (2.53a) and (2.53b) are valid for a general geometry where a particle, situated in free space, is near an absorbing medium. If the particle is itself embedded in a medium near the boundary with another medium, the coupling of the particle to the local electromagnetic field is important and this is taken into account by local-field corrections [12, 28–30].

The versatility of the Green's function approach means that the derivation of the Casimir-Polder potential is widely applicable to a variety of geometries. As an illustrative example taken from Ref. [12], consider the Casimir-Polder potential between an atom in free space and an absorbing magnetodielectric halfspace, this is pictorially represented in Fig. 2.2. The diagram in Fig. 2.2 shows a geometry where the boundary of the medium is the $(x-y)$-plane at $z=0$. The atom is in the free space region, labelled 1, which fills the space $z>0$, and the medium, denoted by subscript 2, fills out the space $z<0$. The scattering part of the Green's function is [25]

$$\begin{aligned}\boldsymbol{G}^{(1)}(\mathbf{r},\mathbf{r}',\omega) = \frac{i}{8\pi^2}\int \frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\big[&R_{MM}\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}) \\ &+ R_{NN}\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})\big],\end{aligned} \tag{2.55}$$

where k_{zj} refers to the z-component of the wavevector in region j and $\mathbf{k}_s$ is the vector of the transverse component of the wave vector, which is $\mathbf{k}_s = (k_x, k_y, 0)$ in this geometry. The reflection coefficients,

R_{MM} and R_{NN} are the well known Fresnel coefficients

$$R_{MM} = \frac{\mu(\omega)k_{z1} - k_{z2}}{\mu(\omega)k_{z1} + k_{z2}}, \tag{2.56a}$$

$$R_{NN} = \frac{\varepsilon(\omega)k_{z1} - k_{z2}}{\varepsilon(\omega)k_{z1} + k_{z2}}, \tag{2.56b}$$

where $\varepsilon(\omega) = \varepsilon_2(\omega)$ and $\mu(\omega) = \mu_2(\omega)$. The functions $\mathbf{M}(\mathbf{k}_s, k_{z1}, \mathbf{r})$ and $\mathbf{N}(\mathbf{k}_s, k_{z1}, \mathbf{r})$ are vector wave functions,

$$\mathbf{M}(\mathbf{k}_s, k_{z1}, \mathbf{r}) = \nabla \times \hat{\mathbf{z}} e^{i\mathbf{k}\mathbf{r}} \tag{2.57a}$$

$$\mathbf{N}(\mathbf{k}_s, k_{z1}, \mathbf{r}) = \frac{1}{k} \nabla \times (\nabla \times \hat{\mathbf{z}} e^{i\mathbf{k}\mathbf{r}}). \tag{2.57b}$$

Appendix E details the derivation of this scattering Green's function, the reflection coefficients and the vector wave functions. The atom and the medium are both assumed to be isotropic. For the medium this means that the response functions reduce to $\boldsymbol{\varepsilon}(\omega) = \varepsilon(\omega)\boldsymbol{I}$ and $\boldsymbol{\mu}(\omega) = \mu(\omega)\boldsymbol{I}$, this assumption of isotropic media has already been employed in the reflection coefficients shown above. The electric and magnetic atomic polarisabilities reduce to

$$\boldsymbol{\alpha}^n(i\xi) = \alpha^n(i\xi)\boldsymbol{I}, \quad \alpha^n(i\xi) = \frac{2}{3\hbar} \sum_k \frac{\omega_{kn} |d_{nk}|^2}{\omega_{kn}^2 + \xi^2}, \tag{2.58a}$$

$$\boldsymbol{\beta}^n(i\xi) = \beta^n(i\xi)\boldsymbol{I}, \quad \beta^n(i\xi) = \frac{2}{3\hbar} \sum_k \frac{\omega_{kn} |m_{nk}|^2}{\omega_{kn}^2 + \xi^2}, \tag{2.58b}$$

respectively, where the isotropic averages

$$\mathbf{d}_{kn} \otimes \mathbf{d}_{nk} = \frac{\mathbf{d}_{kn} \cdot \mathbf{d}_{nk}}{3} = \frac{|\mathbf{d}_{nk}|^2}{3}$$

$$\mathbf{m}_{kn} \otimes \mathbf{m}_{nk} = \frac{\mathbf{m}_{kn} \cdot \mathbf{m}_{nk}}{3} = \frac{|\mathbf{m}_{nk}|^2}{3}$$

have been used. The Casimir-Polder potential can now written as

$$\begin{aligned} U_e^n(z_A) =& \frac{\hbar\mu_0}{8\pi^2} \int_0^\infty \mathrm{d}\xi\, \xi^2 \alpha^n(i\xi) \int_{\xi/c}^\infty \mathrm{d}\tilde{k}_{z1} e^{-2\tilde{k}_{z1} z_A} \left[\frac{\mu(i\xi)\tilde{k}_{z1} - \tilde{k}_{z2}}{\mu(i\xi)\tilde{k}_{z1} + \tilde{k}_{z2}} + \left(1 - \frac{2\tilde{k}_{z1}^2 c^2}{\xi^2}\right) \frac{\varepsilon(i\xi)\tilde{k}_{z1} - \tilde{k}_{z2}}{\varepsilon(i\xi)\tilde{k}_{z1} + \tilde{k}_{z2}} \right] \\ &+ \frac{\mu_0}{12\pi} \sum_k \Theta(\omega_{nk}) \omega_{nk}^2 |\mathbf{d}_{nk}|^2 \mathrm{Im} \Bigg[\int_0^\infty \frac{\mathrm{d}k_s}{k_{z1}} e^{2ik_{z1} z_A} \left[\frac{\mu(\omega_{nk})k_{z1} - k_{z2}}{\mu(\omega_{nk})k_{z1} + k_{z2}} \right. \\ &\left. + \left(1 - \frac{2k_{z1}^2 c^2}{\omega_{nk}^2}\right) \frac{\varepsilon(\omega_{nk})k_{z1} - k_{z2}}{\varepsilon(\omega_{nk})k_{z1} + k_{z2}} \right] \Bigg] \end{aligned} \tag{2.59a}$$

$$\begin{aligned} U_m^n(z_A) =& \frac{\hbar\mu_0}{8\pi^2} \int_0^\infty \mathrm{d}\xi\, \xi^2 \frac{\beta^n(i\xi)}{c^2} \int_{\xi/c}^\infty \mathrm{d}\tilde{k}_{z1} e^{-2\tilde{k}_{z1} z_A} \left[\frac{\varepsilon(i\xi)\tilde{k}_{z1} - \tilde{k}_{z2}}{\varepsilon(i\xi)\tilde{k}_{z1} + \tilde{k}_{z2}} + \left(1 - \frac{2\tilde{k}_{z1}^2 c^2}{\xi^2}\right) \frac{\mu(i\xi)\tilde{k}_{z1} - \tilde{k}_{z2}}{\mu(i\xi)\tilde{k}_{z1} + \tilde{k}_{z2}} \right] \\ &- \frac{\mu_0}{12\pi c^2} \sum_k \Theta(\omega_{nk}) \omega_{nk}^2 |\mathbf{m}_{nk}|^2 \mathrm{Im} \Bigg[\int_0^\infty \frac{\mathrm{d}k_s}{k_{z1}} e^{2ik_{z1} z_A} \left[\frac{\varepsilon(\omega_{nk})k_{z1} - k_{z2}}{\varepsilon(\omega_{nk})k_{z1} + k_{z2}} \right. \\ &\left. + \left(1 - \frac{2k_{z1}^2 c^2}{\omega_{nk}^2}\right) \frac{\mu(\omega_{nk})k_{z1} - k_{z2}}{\mu(\omega_{nk})k_{z1} + k_{z2}} \right] \Bigg]. \end{aligned} \tag{2.59b}$$

The expressions with a ˜ refer to the imaginary parts of the respective quantity, i.e., $\tilde{k}_z = \mathrm{Im}k_z$, etc., these appear in the off-resonant contributions which are in terms of the complex frequency, the details

are shown in Appendix E. In the off-resonant contribution to the Casimir-Polder potential the response functions are evaluated along the imaginary frequency axis where they must be real and positive.

In general the Casimir-Polder potential has to be calculated numerically, but some assumptions can be made in special circumstances to simplify the potential. For example, the behaviour of the electromagnetic fields in the close and far limits of distance between the atom and the medium can be used. If $z_A\omega_{\min}/c \gg 1$ ($\omega_{\min}$ is the minimum relevant particle transition frequency) holds, it is assumed that the atom is far enough away from the medium that z_A is greater than the effective transition frequency $\omega_{\min}/c$ and the atomic polarisations can be approximated by their static forms ($\alpha(i\xi) \approx \alpha(0)$, $\beta(i\xi) \approx \beta(0)$), this is called the retarded limit. In the opposite, the non-retarded limit, the relation $z_A\omega_{\max}/c \ll 1$ ($\omega_{\max}$ is the maximum relevant particle transition frequency) holds. Here the distance between the particle and the medium is smaller than the transition wavelength. This means that in the non-retarded limit the exponential term approximates to unity and the main contribution to the integral comes from the k_z^2 term. This thesis is primarily concerned with the non-retarded limit, so an example of its application will highlight the methods to be used further on. The dispersion relations for free space and the magnetodielectric medium are

$$k_1 = \frac{\omega}{c} \tag{2.60a}$$

$$k_2 = \frac{\omega}{c}\sqrt{\varepsilon(\omega)\mu(\omega)} \tag{2.60b}$$

respectively, these are derived in Appendix E. This means that $\tilde{k}_{z2}$ can be written in terms of $\tilde{k}_{z1}$, $\tilde{k}_1$ and $\tilde{k}_2$, (by noting that k_s is the same for both) as

$$\tilde{k}_{z2} = \sqrt{\tilde{k}_{z1}^2 + \frac{\xi^2}{c^2}(\varepsilon(i\xi)\mu(i\xi) - 1)} \tag{2.61}$$

In the non-retarded limit, $\frac{\xi^2}{c^2}(\varepsilon_2\mu_2 - 1)$ is small and hence $\tilde{k}_{z2}$ can be Taylor expanded around $\tilde{k}_{z1}$ to second order to obtain

$$\tilde{k}_{z2} \approx \tilde{k}_{z1}\left(1 + \frac{\frac{\xi^2}{c^2}(\varepsilon(i\xi)\mu(i\xi) - 1)}{2\tilde{k}_{z1}^2} + \dots\right). \tag{2.62}$$

For many calculations this can be truncated at first order, that is $\tilde{k}_{z2} \approx \tilde{k}_{z1} \approx k_s$. This then gives the Casimir-Polder potential, to first order in the non-retarded limit for an atom initially in state n, as

$$U_{eO}^n(z_A) = -\frac{\hbar}{16\pi^2\varepsilon_0(z_A)^3}\int_0^\infty \mathrm{d}\xi\, \alpha^n(i\xi)\left(\frac{\varepsilon(i\xi) - 1}{\varepsilon(i\xi) + 1}\right), \tag{2.63a}$$

$$U_{mO}^n(z_A) = -\frac{\hbar}{16\pi^2\varepsilon_0(z_A)^3}\int_0^\infty \mathrm{d}\xi \frac{\beta^n(i\xi)}{c^2}\left(\frac{\mu(i\xi) - 1}{\mu(i\xi) + 1}\right), \tag{2.63b}$$

for the off-resonant terms and

$$U_{eR}^n(z_A) = -\frac{1}{24\pi\varepsilon_0(z_A)^3}\sum_k \Theta(\omega_{nk})|d_{nk}|^2\,\mathrm{Re}\left[\frac{\varepsilon(\omega_{nk}) - 1}{\varepsilon(\omega_{nk}) + 1}\right], \tag{2.64a}$$

$$U_{mR}^n(z_A) = \frac{\mu_0}{24\pi(z_A)^3}\sum_k \Theta(\omega_{nk})|m_{nk}|^2\,\mathrm{Re}\left[\frac{\mu(\omega_{nk}) - 1}{\mu(\omega_{nk}) + 1}\right], \tag{2.64b}$$

for the resonant terms. The dominant contributions are the terms proportional to k_{z1}^2 and so only those terms were considered in the above calculation. As can be seen, the Casimir-Polder potential is dependent on the electric or magnetic dipole matrix elements of the atom and the electric or magnetic response functions of the medium. Therefore the strength of the Casimir-Polder potential is proportional to the properties of the atom and the medium. The spatial scaling law, which describes the behaviour

of Casimir-Polder potential as the distance between the atom and the macroscopic body varies, is an important paramenter in dispersion forces [31]. Here, the spatial scaling in the resonant and non-resonant terms is $1/(z_A)^3$. The similar structure between the electric and magnetic components should be noted, that one can be transformed into the other with substitutions such as $\varepsilon \leftrightarrow \mu$, etc. is a consequence of duality invariance which will be discussed in section 4.3.

The versatility of the Green's function approach means that the Casimir-Polder potential in other geometric scenarios can also be found through this derivation. These include a particle near a plate rather than a semi-infinite halfspace, which means that scattering off the far boundary of the plate need to be considered [32]. In more complicated geometries, such as those with many arbitrarily shaped bodies, the Green's function can be expressed as a Born series expansion [12].

This chapter has outlined a procedure for quantising the electromagnetic field in linear absorbing media and has derived a dispersion force, the Casimir-Polder force, that acts between polarisable and magnetisable objects. The next chapter will show the generalisation of the quantisation method to all linear media, including non-reciprocal media.

Chapter 3

Non-Reciprocal Linear Media

The linear response of macroscopic media to externally applied electromagnetic fields is frequently discussed in terms of a local electric response, through an electric permittivity, and a local magnetic response, through a magnetic permeability. However, there are media where this description is inadequate. For example, there exist materials where an externally applied electric field produces a magnetic response and a vice versa. The general name for media with these properties is bianisotropic media, which is discussed in chapter 4. Some further examples where the basic description of macroscopic media is insufficient include media with non-local spatial responses and media whose response is non-reciprocal, both of which are discussed in the present chapter.

In spatially non-local (sometimes refered to as spatially dispersive) media, the response of a medium to an applied electromagnetic field can be dependent upon more positions than just the observation point. In media with non-local spatial responses it is known that the decomposition into electric and magnetic effects is poorly defined [33], as the magnetic response can be seen as a special example of a non-local electric response [34]. A well known example of a non-local spatial response is a medium, whose response is spatially local in its rest frame, in motion. As a brief example, a non-local response can be represented in frequency space by

$$\mathbf{D}(\mathbf{r},\omega) = \int \mathrm{d}^3 r' \boldsymbol{\varepsilon}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{E}(\mathbf{r}',\omega), \tag{3.1}$$

where $\mathbf{r}$ and $\mathbf{r}'$ are the observation and source positions respectively. This clearly shows that the displacement field at position $\mathbf{r}$ depends upon the electric field at positions $\mathbf{r}'$ and not just at $\mathbf{r}$. In contrast, a local response is commonly represent in the form

$$\mathbf{D}(\mathbf{r},\omega) = \boldsymbol{\varepsilon}(\mathbf{r},\omega)\cdot\mathbf{E}(\mathbf{r},\omega), \tag{3.2}$$

which shows that the displacement field at $\mathbf{r}$ only upon the electric field at $\mathbf{r}$.

In nanoplasmonics the separation between objects can be very small ($\lesssim \mu m$) and as such the materials become non-local at this scale. This has lead to interest in non-local media from a theoretical transformation optics view [35,36] and the subsequent proposed construction of novel metamaterials that exhibit non-local responses [37,38]. It has been shown that the effective macroscopic response functions can be derived for a specific microscopic structure taking into account spatial dispersion [39]. In the context of macroscopic QED, recent work has presented a quantisation that encompasses spatial dispersion [40,41].

A common assumption when dealing with macroscopic media is that the media are reciprocal. This is a manifestation of time-reversibility and it implies that the physics of the system remain the same when time is reversed. This assumption has been shown to break down in certain classes of media, such

as moving media (see chapter 6 for details), Tellegen media [42,43] and systems under the influence of an external magnetic field. In fact, an initial motivation for non-reciprocal media was for use in electronics, where devices such as isolators use non-reciprocal media to only allow power to be transmitted in one direction. Continuous work in electronics to produce new types of electromagnetic isolator have recently used metamaterials to create a non-reciprocal medium [44], as well as new ways to use commercially available magnets [45].

Initial work by L. Onsager in the 1930's mainly considered reversible microscopic processes, but also contained a discussion of irreversible processes with examples such as a free particle under the influence of an external magnetic field and the Coriolis force. Onsager stated that "In such cases, where the macroscopic laws of motion are non-reversible, the microscopic motion cannot be reversible." [46, 47]. Therefore the non-reciprocal microscopic behaviour can be described in terms of the properties of macroscopic variables, such as response functions and a Green's function. Hence in this context, reciprocity (sometimes referred to as Onsager reciprocity) has come to be seen as the time symmetry invariance of a quantum optical system. With reference to macroscopic QED, the exact microscopic processes that cause a non-reciprocal (irreversible) response do not need to be known in each circumstance. As long as the macroscopic response functions take into account the non-reciprocal behaviour of the media, the field quantisation from a macroscopic viewpoint can be performed consistently.

Past controversy [48–52] aside, there is still interest in non-reciprocal media. As well as the ongoing research from an electronics perspective, recent interest in non-reciprocal media has come from the increasing research into phenomena such as the fractional Quantum Hall effect [53, 54], which can also have a non-local response [55]. This means that a general theory of quantised linear media would have to be applicable to spatially non-local as well as locally responding media and it would have to apply to reciprocal and non-reciprocal media. Previous theories in the linear response regime have either catered for a non-local spatial response [40] or have accounted for non-reciprocal media [19], but never in combination.

This chapter will describe the quantisation of the electromagnetic field in general linear absorbing media in which no assumptions have been made with regard to the reciprocity of the media, or the spatial response of the media, and so will apply to a wide variety of classes of media. The quantisation will be achieved by introducing a complex conductivity tensor, which contains the medium's response to externally applied electromagnetic fields, into Maxwell's equations through a generalised version of Ohm's law. The results in this chapter were initially published in Ref. [56].

3.1 Field Quantisation in General Linear Media

In previous treatments of electromagnetic field quantisation in absorbing media [12], it has been assumed that the medium under consideration was reciprocal. In order to generalise these theories, the quantisation must apply to non-reciprocal media as well. By only considering the response of a medium to externally applied electromagnetic fields to linear order and by enforcing causality, a general theory for absorbing media can be derived. As was shown for general linear reciprocal media [40], this can be achieved with the complex conductivity tensor, $\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', \omega)$. The two spatial variables in the conductivity tensor are in general independent which allows for non-local media to be included in this description of general linear media. Although knowledge of the conductivity tensor is required, this is the only basic piece of information about the medium that is needed. By describing the medium's linear response to electromagnetic fields in terms of the conductivity tensor, it is not necessary to separate these responses into electric (polarisation) and magnetic (magnetisation) effects. This is important, as the distinction between a polarisation and magnetisation is not well defined for non-local media [33]. The conductivity

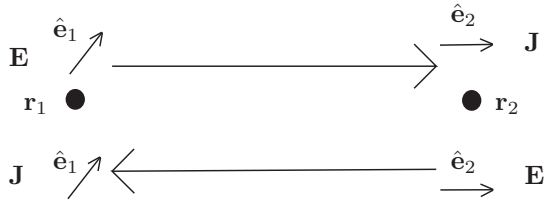


Figure 3.1: This sketch shows the optical path between a source and observation point and the reverse orientation. In this example the connection between the source and observation point is described by the conductivity tensor, $\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', \omega)$. The top part of the diagram shows a relationship between a source term and an induced current. In the lower part of the diagram the reverse is found by switching the source and observation points *and* the orientation of the vectors. In reciprocal media, this is the same as reversing the optical path and this reflects the fact that reciprocity is also a statement of time symmetry. In non-reciprocal media, this reverse orientiation is not equivalent to a time reversal of the optical path and so the time symmetry invariance does not hold.

tensor is introduced by a generalised Ohm's law (in frequency space)

$$\mathbf{j}_{\text{in}}(\mathbf{r}, \omega) = \int \mathrm{d}^3 r' \, \boldsymbol{Q}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{E}(\mathbf{r}', \omega) + \mathbf{j}_{\text{N}}(\mathbf{r}, \omega), \tag{3.3}$$

where $\mathbf{j}_{\text{in}}$ is the current induced in the medium. This comprises of a component dependent on the interaction of the medium with an applied electric field and a noise current which arises due to the dissipation into the medium. The conductivity tensor must fulfil the causality requirements that $\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', t) = \mathbf{0}$ for $t < 0$. This ensures that the effect of the electromagnetic source at position $\mathbf{r}'$ cannot produce a response in the medium at an arbitrary observation position $\mathbf{r}$ faster than the speed of light and that the response must always occur after the electromagnetic field has been applied. As $\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', t)$ must be a real quantity, a further condition on the conductivity tensor is the Schwarz reflection principle,

$$\boldsymbol{Q}^*(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{Q}(\mathbf{r}, \mathbf{r}', -\omega^*) \tag{3.4}$$

which the conductivity tensor must fulfil for all values of $\mathbf{r}$, $\mathbf{r}'$ and ω, this is derived in appendix B. Although this initial work on the quantisation follows the method as discussed in [40], the theories diverge here.

In reciprocal media, the conductivity tensor has to obey a condition of the form

$$\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{Q}^{\mathrm{T}}(\mathbf{r}', \mathbf{r}, \omega). \tag{3.5}$$

For the general quantisation procedure to be developed here, Eq. (3.5) does not necessarily hold. The effect of non-reciprocity in an electromagnetic system can be seen as follows. The conductivity tensor in Eq. (3.3) governs the relationship between an electric field, $\mathbf{E}$, at a source point $\mathbf{r}_1$ orientated in a direction $\hat{\mathbf{e}}_1$ with a current, $\mathbf{J}$, at the observation point $\mathbf{r}_2$ and which is orientated in an arbitrary direction $\hat{\mathbf{e}}_2$. If the reciprocity condition, Eq. (3.5), does hold, this would automatically imply that an electric field, $\mathbf{E}$, at $\mathbf{r}_2$ in orientation $\hat{\mathbf{e}}_2$, would induce a the current of value $\mathbf{J}$ at $\mathbf{r}_1$ in the orientation $\hat{\mathbf{e}}_1$. This is illustrated in Fig. 3.1. This switching of the source and observation points is a reversal of the optical path taken, but it can also be viewed as a reversal of time, $t \to -t$, and the reciprocal condition implies that the optical paths are equivalent. Therefore when Eq. (3.5) does not hold, it cannot be said that the two paths are equivalent and so the time symmetry is broken.

An implication of removing the assumption that the media are necessarily reciprocal is that the previously used real and imaginary parts of a tensor field are no longer equivalent to the Hermitian and

anti-Hermitian components of the tensor field. It is therefore expedient to define generalised real and imaginary parts of a tensor field, $\boldsymbol{O}(\mathbf{r},\mathbf{r}')$, i.e. the Hermitian and anti-Hermitian parts, as

$$\mathcal{R}\mathrm{e}[\boldsymbol{O}(\mathbf{r},\mathbf{r}')] = \frac{1}{2}[\boldsymbol{O}(\mathbf{r},\mathbf{r}') + \boldsymbol{O}^{\dagger}(\mathbf{r}',\mathbf{r})], \tag{3.6a}$$

$$\mathcal{I}\mathrm{m}[\boldsymbol{O}(\mathbf{r},\mathbf{r}')] = \frac{1}{2i}[\boldsymbol{O}(\mathbf{r},\mathbf{r}') - \boldsymbol{O}^{\dagger}(\mathbf{r}',\mathbf{r})], \tag{3.6b}$$

respectively. If the medium in question is reciprocal ($\boldsymbol{O}(\mathbf{r},\mathbf{r}') = \boldsymbol{O}^{\dagger}(\mathbf{r}',\mathbf{r})$ holds for all $\mathbf{r},\mathbf{r}'$) then Eqs. (3.6a) and (3.6b) reduce to the standard real and imaginary parts of a tensor field. However this is not true in the general case, where $\mathcal{R}\mathrm{e}$ and $\mathcal{I}\mathrm{m}$ refer to the Hermitian and anti-Hermitian components of the tensor field. Now that the conductivity tensor can be written in terms of generalised real and imaginary parts,

$$\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = \mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)] + i\mathcal{I}\mathrm{m}[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)], \tag{3.7}$$

the causal conditions imply that the conductivity tensor is constrained by Kramers-Kronig type relation, as shown in appendix A.

D. Melrose in Ref. [33] uses an equivalent dielectric tensor, $\boldsymbol{K}(\mathbf{k},\omega)$, to show that the Hermitian part of this response function describes the non-dissipative contribution and the anti-Hermitian part is the dissipative component. The connection between Melrose' equivalent dielectric tensor and the conductivity is

$$\boldsymbol{K}(\mathbf{k},\omega) = \boldsymbol{I} + \frac{i}{\omega\varepsilon_0}\boldsymbol{Q}(\mathbf{k},\omega),$$

therefore these relationships are reversed for the conductivity tensor. The Hermitian, or generalised real part of the conductivity tensor corresponds to the dissipative component and the anti-Hermitian part is the non-dissipative contribution.

Taking this into consideration, the generalised real part of the conductivity tensor is therefore used to postulate a commutation relation of the noise current, which is treated as an operator,

$$\left[\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{j}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \frac{\hbar\omega}{\pi}\mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)]\delta(\omega-\omega'). \tag{3.8}$$

The noise current is now considered as the dynamical variables of the matter-field system. This result is the generalisation of the commutation relation for reciprocal media, described in Ref. [40]. Although this commutation relation has been postulated, it can be derived from first principles through the procedures outlined in Refs. [4, 19]. By combining the generalised Ohm's law, Eq. (3.3), with the macroscopic Maxwell's equations, Eqs. (2.1), the inhomogeneous Helmholtz equation for the electric field can be obtained

$$\left[\nabla\times\nabla\times\boldsymbol{I} - \frac{\omega^2}{c^2}\boldsymbol{I}\right]\cdot\hat{\mathbf{E}}(\mathbf{r},\omega) - i\mu_0\omega\int \mathrm{d}^3r'\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{E}}(\mathbf{r}',\omega) = i\mu_0\omega\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega). \tag{3.9}$$

In the source quantity representation the solution for the electric field is given as

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = i\mu_0\omega\int \mathrm{d}^3r'\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{3.10}$$

where the Green's function is uniquely defined as the solution to the Helmholtz equation

$$\left[\nabla\times\nabla\times\boldsymbol{I} - \frac{\omega^2}{c^2}\boldsymbol{I}\right]\cdot\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) - i\mu_0\omega\int \mathrm{d}^3s\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'). \tag{3.11}$$

The Green's function is constrained by the boundary conditions $|\mathbf{r}-\mathbf{r}'|\to\infty$, $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\to 0$, which ensure the correct physical behaviour as the source and observation points diverge. The definition of the

Green's function in Eq. (3.11) shows that the properties of the conductivity tensor are inherited by the Green's function. This includes the Schwarz reflection principle

$$\boldsymbol{G}^*(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}(\mathbf{r},\mathbf{r}',-\omega^*) \quad \forall\ \mathbf{r},\mathbf{r}',\omega \tag{3.12}$$

and the causal behaviour, $\boldsymbol{G}(\mathbf{r},\mathbf{r}',t) = \mathbf{0}$ for $t < 0$. Crucially, the non-reciprocity of the conductivity is also inherited, so that the relationship

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega), \tag{3.13}$$

derived for a reciprocal system in Appendix C, does not necessarily hold. As the Green's function describes the propagation through a medium and characterises the electric field produced by a current source, any of the electromagnetic fields propagating in non-reciprocal media can now be characterised. This is of course a necessary requirement for any quantum theory of light in non-reciprocal media. The magnetic induction field is

$$\hat{\mathbf{B}}(\mathbf{r},\omega) = \mu_0 \int \mathrm{d}^3 r' \nabla \times \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \cdot \hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{3.14}$$

which is obtained from substituting Eq. (3.10) into Eq. (2.1b).

To continue the quantum description of the electromagnetic field in a non-reciprocal medium, the absorption into the medium needs to be taken into account. The first step is finding an integral relation, connecting the fluctuations in the system with the dissipation into the medium. This can be obtained by making use of the generalised real and imaginary parts of a tensor field defined above and is of the form

$$\mu_0\omega \int \mathrm{d}^3 s \int \mathrm{d}^3 s'\, \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega) \cdot \mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{s},\mathbf{s}',\omega)] \cdot \boldsymbol{G}^{\dagger}(\mathbf{r}',\mathbf{s}',\omega) = \mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)], \tag{3.15}$$

the full derivation can be found in appendix D. For a consistent description of absorbing media, the noise currents and the fields must satisfy the fluctuation dissipation theorem. This is an important concept in statistical physics and it states that the mediums response to applied fields is linked to and can be expressed in terms of the strength of the field fluctuations [57,58]. The fluctuations of the noise currents need to be proportional to the dissipative part of the medium's response functions to be consistent with the fluctuation dissipation theorem. To proceed, the ground state of the matter-field system is denoted by $|\{0\}\rangle$ and is defined such that

$$\hat{\mathbf{j}}_{\mathrm{N}}|\{0\}\rangle = 0, \tag{3.16}$$

and the average of the fluctuations of the noise current is given as

$$\langle \hat{\mathbf{j}}_{\mathrm{N}} \rangle = 0. \tag{3.17}$$

To show that the commutation relation Eq. (3.8) is consistent with the fluctuation dissipation theorem the symmeterised operator product, defined as [25]

$$\left\{ \hat{\mathbf{a}}, \hat{\mathbf{b}}^{\dagger} \right\} = \frac{1}{2}(\hat{a}\hat{b} + \hat{b}\hat{a}), \tag{3.18}$$

is introduced. This shows the correlation between the quantities $\hat{\mathbf{a}}$ and $\hat{\mathbf{b}}$. By defining $\Delta\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ as

$$\Delta\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega) = \hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega) - \langle \hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega) \rangle, \tag{3.19}$$

the commutation relation Eq. (3.8) is used to show that the noise currents satisfy the relation

$$\left\langle \left\{ \Delta\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega), \Delta\hat{\mathbf{j}}_{\mathrm{N}}^{\dagger}(\mathbf{r}',\omega') \right\} \right\rangle = \frac{\hbar}{\pi} \mathcal{I}\mathrm{m}[i\omega \boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)]\delta(\omega-\omega'). \tag{3.20}$$

As discussed previously, it is know that the dissipation is described by the Hermitian part of the conductivity tensor, this confirms that the quantisation scheme does fulfil the fluctuation dissipation theorem. By making use of the definition of the electric field, Eq. (3.10), and the relation, Eq. (3.20), it is straight forward to show that the electric field also satisfies the fluctuation dissipation theorem

$$\left\langle \left\{ \Delta\hat{\mathbf{E}}(\mathbf{r},\omega), \Delta\hat{\mathbf{E}}^{\dagger}(\mathbf{r}',\omega') \right\} \right\rangle = \frac{\hbar}{\pi} \mathcal{I}\mathrm{m}\big[\mu_0\omega^2 \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\big]\delta(\omega-\omega'). \tag{3.21}$$

An important aspect of any quantum theory is the Hamiltonian which can be constructed from the noise current operators. To obtain the correct free time evolution the Hamiltonian $\hat{H}$ is constrained by

$$[\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{H}] = \hbar\omega \hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega). \tag{3.22}$$

This gives the Hamiltonian of the matter-field system as

$$\hat{H}_F = \pi \int_0^{\infty} \mathrm{d}\omega \int \mathrm{d}^3 r \int \mathrm{d}^3 r' \hat{\mathbf{j}}_{\mathrm{N}}^{\dagger}(\mathbf{r},\omega) \cdot (\mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)])^{-1} \hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{3.23}$$

plus some irrelevant constants that arise due to the constraints put on the Hamiltonian. At this point the theory has field operators $\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ with a well defined commutation relation, an integral relation, it fulfils the fluctuation dissipation theorem and has a Hamiltonian. The next step is to calculate the equal time commuatation relations between the electric and magnetic induction fields, these need to replicate the results obtained from the equivalent free space QED commutation relations to verify the quantisation procedure. By using the definition of the electric field, Eq. (3.10), and the integral relation, Eq. (3.15), the commutation relation between the frequency components of the electric field and its Hermitian conjugate is shown to be

$$[\hat{\mathbf{E}}(\mathbf{r},\omega), \hat{\mathbf{E}}^{\dagger}(\mathbf{r}',\omega')] = \frac{\hbar\omega\omega'\mu_0}{\pi} \mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)]\delta(\omega-\omega'), \tag{3.24}$$

as required. It can also be shown that the associated commutation relations,

$$[\hat{\mathbf{E}}(\mathbf{r},\omega), \hat{\mathbf{E}}(\mathbf{r}',\omega')] = [\hat{\mathbf{E}}^{\dagger}(\mathbf{r},\omega), \hat{\mathbf{E}}^{\dagger}(\mathbf{r}',\omega')] = 0, \tag{3.25}$$

are also correct. Hence, the commutation relation of the electric field operators as a function of position, $\hat{\mathbf{E}}(\mathbf{r})$ and $\hat{\mathbf{E}}(\mathbf{r}')$, is found to be

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{E}}(\mathbf{r}')] = 0, \tag{3.26}$$

as required. To show the commutation relation between the electric field and the magnetic induction field, Faraday's law Eq. (2.1b) is used with Eq. (2.35a) to obtain

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{B}}(\mathbf{r}')] = \left[\left(\int_0^{\infty} \mathrm{d}\omega \hat{\mathbf{E}}(\mathbf{r},\omega) + \mathrm{H.c.} \right), \left(\int_0^{\infty} \frac{\mathrm{d}\omega'}{i\omega'} \nabla \times \hat{\mathbf{E}}(\mathbf{r},\omega) + \mathrm{H.c.} \right) \right], \tag{3.27}$$

which is then written as

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{B}}(\mathbf{r}')] = \frac{i\hbar\mu_0}{\pi} \nabla_{\mathbf{r}'} \times \int_0^{\infty} \mathrm{d}\omega\, \omega \Big(\mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)] + \mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r}',\mathbf{r},\omega)] \Big), \tag{3.28}$$

by making use of Eqs. (3.24) and (3.25). The right hand side of Eq. (3.28) needs to be treated carefully as this is the anti-Hermitian part of the dyadic Green's function. However, as the Green's function is analytic in the upper half plane, the generalised imaginary part can be expanded in terms of the Green's function ($\mathcal{I}\mathrm{m}[\boldsymbol{G}] = \frac{1}{2i}(\boldsymbol{G} - \boldsymbol{G}^\dagger)$) and then the integration contour in the upper half plane can be closed with use of the Schwarz reflection principle. It can be seen from Eq. (3.11) that as $|\omega| \to \infty$, $\boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) \to -\frac{c^2}{\omega^2}\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')$.

Therefore the commutation relation can now be written as

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{B}}(\mathbf{r}')] = -\frac{i\hbar}{\varepsilon_0}\nabla_{\mathbf{r}'} \times \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}') \tag{3.29}$$

which is the result from free space QED, as required. The commutation relation between the other fields follows from this, as can be shown by use of Maxwell's equations.

So far the quantisation has been achieved in terms of the noise currents, but annihilation and creation operators of the matter-field system can be specified. These are commonly denoted by $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$, respectively. They are related to the noise currents by

$$\hat{\mathbf{j}}_\mathrm{N}(\mathbf{r}, \omega) = \sqrt{\frac{\hbar\omega}{\pi}} \int \mathrm{d}^3 r' \boldsymbol{R}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}', \omega) \tag{3.30}$$

where $\boldsymbol{R}(\mathbf{r}, \mathbf{r}', \omega)$ is the (positive) square root of the generalised real part of the conductivity, $\mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', \omega)]$,

$$\mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{r}, \mathbf{r}', \omega)] = \int \mathrm{d}^3 s \boldsymbol{R}(\mathbf{r}, \mathbf{s}, \omega) \cdot \boldsymbol{R}^\dagger(\mathbf{r}', \mathbf{s}, \omega). \tag{3.31}$$

With the commutation relation for the noise currents, Eq. (3.8), and the relationship between the system excitations and the noise currents, Eq. (3.30), the commutation relation

$$[\hat{\mathbf{f}}(\mathbf{r}, \omega), \hat{\mathbf{f}}^\dagger(\mathbf{r}', \omega')] = \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega') \tag{3.32}$$

is ensured. By substituting Eqs. (3.30) and (3.31) into Eq. (3.23), the Hamiltonian of the matter-field system can be rewritten as

$$\hat{H}_F = \int \mathrm{d}^3 r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}', \omega'). \tag{3.33}$$

The free evolution of the dynamical variables is determined as $\hat{\mathbf{f}}(\mathbf{r}, \omega, t) = \hat{\mathbf{f}}(\mathbf{r}, \omega)e^{-i\omega t}$. This means that, by construction, Maxwell's equations for the electromagnetic field operators in the Heisenberg picture are valid. This can be shown by use of Heisenberg's equation of motion,

$$\dot{\hat{\mathbf{O}}} = \frac{i}{\hbar}[\hat{H}, \hat{\mathbf{O}}] \tag{3.34}$$

for the time evolution of an operator, $\hat{\mathbf{O}}$. This means that the noise currents are governed by

$$\dot{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r}, \omega) = \frac{i}{\hbar}[\hat{H}_F, \hat{\mathbf{j}}_\mathrm{N}(\mathbf{r}, \omega)] = -i\omega\hat{\mathbf{j}}_\mathrm{N}(\mathbf{r}, \omega) \tag{3.35}$$

and the equivalent relation for the system excitations

$$\dot{\hat{\mathbf{f}}}(\mathbf{r}, \omega) = \frac{i}{\hbar}[\hat{H}_F, \hat{\mathbf{f}}(\mathbf{r}, \omega)] = -i\omega\hat{\mathbf{f}}(\mathbf{r}, \omega). \tag{3.36}$$

The time derivative of the magnetic induction field, written in the form Eq. (3.14), is then shown to be

$$\begin{aligned}\dot{\hat{\mathbf{B}}}(\mathbf{r},\omega) &= \frac{i}{\hbar}[\hat{H}_F, \hat{\mathbf{B}}(\mathbf{r},\omega)] = \frac{i}{\hbar}\int \mathrm{d}^3 s \nabla\times\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)[\hat{H}_F, \hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{s},\omega)] \\ &= -\nabla\times\hat{\mathbf{E}}(\mathbf{r},\omega),\end{aligned} \tag{3.37}$$

which is Faraday's law. Ampère's law is obtained similarly

$$\dot{\hat{\mathbf{D}}}(\mathbf{r},\omega) = \frac{i}{\hbar}[\hat{H}_F, \hat{\mathbf{D}}(\mathbf{r},\omega)] = \nabla\times\hat{\mathbf{H}}(\mathbf{r},\omega). \tag{3.38}$$

In this chapter a quantum theory for the electromagnetic field in a general linear absorbing medium has been presented. By describing the medium's response in terms of a conductivity tensor the theory can be applied to both spatially dispersive and non-reciprocal media where only information about the conductivity tensor itself is required. The next chapter will follow the quantisation of the electromagnetic field in bianisotropic media described by explicit response functions for electric, magnetic and electromagnetic effects and will discuss the duality implications of this system.

Chapter 4

Bianisotropic Linear Media

For a simple linear medium, the macroscopic response to an applied external electromagnetic field is usually described by an electric permittivity and a magnetic permeability. These take the form of tensors that couple the applied fields to the fields in the medium. The displacement field is coupled to the electric field by the permittivity and the magnetic induction field is coupled to the magnetic field by the magnetic permeability. This can be represented in frequency space as

$$\hat{\mathbf{D}}(\mathbf{r},\omega) = \varepsilon_0 \boldsymbol{\varepsilon}(\mathbf{r},\omega)\cdot\hat{\mathbf{E}}(\mathbf{r},\omega), \tag{4.1a}$$

$$\hat{\mathbf{H}}(\mathbf{r},\omega) = \frac{1}{\mu_0}\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)\cdot\hat{\mathbf{B}}(\mathbf{r},\omega), \tag{4.1b}$$

where any noise polarisation and noise magnetisation fields have been omitted for convenience. However, this description of the interaction between applied fields and a medium does not take into account media where an applied electric field causes a magnetic response and an applied magnetic field causes an electric response. These magnetoelectric effects can be described by cross susceptibility tensors, which in conjunction with the electric permittivity and magnetic permeability characterise the medium. The general term for this class of media is bianisotropic, which has been discussed in Refs. [59–61], and more recently in Ref. [19]. Some common examples of bianisotropic media are chiral media, discussed in chapter 5, and moving media, discussed in chapter 6.

Although magnetoelectric effects were known to occur when a medium is in motion (see Ref. [62]), naturally occuring magnetoelectric effects in cystals [63] and other media [64,65] have also been studied. The interest in bianisotropic media can be partially attributed to the potential of metamaterials, where a medium with a bianisotropic response can be constructed [66]. Of particular interest are bianisotropic media that could generate repulsive Casimir forces [67,68].

Although the clear separation of electric and magnetic effects is not always trivial, there are instances where this becomes convenient. For example, in the absense of free charges the equations governing the electric and magnetic fields are similar and they can be constructed as duals of each other. This means that under a suitable transformation, the electric and magnetic laws can be interchanged. If the laws are invariant under this transformation, a duality transformation, then the system is said to possess duality symmetry. While the duality symmetry of systems has long been a topic of research [69,70], it has been recently shown that duality is a symmetry of macroscopic QED for an isotropic magnetodielectric [71].

The duality invariance of electromagnetic systems can be extended to encorporate the Green's functions, which have their own tranformation rules. These can be used in conjunction with transformation laws for atomic properties to find the dual of dispersion forces, decay rates, etc.

In this chapter the quantisation of the electromagnetic field in absorbing media in terms of electromag-

netic response functions is achieved. Section 4.3 extends the previous duality treatment of macroscopic QED to include bianisotropic media and discusses the conditions under which duality is a valid symmetry. The chapter finishes with the duality transformations of the Green's function in free space. The results presented in this chapter were published in Ref. [56].

4.1 Field Quantisation in Bianisotropic Media

A bianisotropic medium can be described by its electric permittivity $\boldsymbol{\varepsilon}$, magnetic permeability, $\boldsymbol{\mu}$, and two cross coupling tensors, $\boldsymbol{\xi}$ and $\boldsymbol{\zeta}$. In isotropic, anisotropic and bianisotropic media, the permittivity couples the electric field, $\hat{\mathbf{E}}$ with the displacement field, $\hat{\mathbf{D}}$, and the permeability couples the magnetic field, $\hat{\mathbf{H}}$, to the magnetic induction field, $\hat{\mathbf{B}}$. In bianisotropic media there are additional cross tensors which couple the magnetic field to the displacement field, $\boldsymbol{\xi}$, and the electric field to the magnetic induction field, $\boldsymbol{\zeta}$, this is labelled the 'DB' convention. There is an alternative description of bianisotropic media in which the cross tensor $\boldsymbol{\xi}$ couples the magnetic induction field to the displacement field and $\boldsymbol{\zeta}$ couples the electric field to the magnetic field, called the 'DH' convention. The two conventions are physically equivalent and the response tensors from each convention are linked by the simple relationships

$$\boldsymbol{\varepsilon}_{DH} = \boldsymbol{\varepsilon}_{DB} - \boldsymbol{\xi}_{DB} \star \boldsymbol{\mu}_{DB}^{-1} \star \boldsymbol{\zeta}_{DB}, \tag{4.2a}$$

$$\boldsymbol{\xi}_{DH} = \boldsymbol{\xi}_{DB} \star \boldsymbol{\mu}_{DB}^{-1}, \tag{4.2b}$$

$$\boldsymbol{\zeta}_{DH} = \boldsymbol{\mu}_{DB}^{-1} \star \boldsymbol{\zeta}_{DB}, \tag{4.2c}$$

$$\boldsymbol{\mu}_{DH} = \boldsymbol{\mu}_{DB}. \tag{4.2d}$$

Here, the '$\star$' operation is notational shorthand for a spatial convolution, for example,

$$\boldsymbol{\varepsilon} \star \hat{\mathbf{E}} = [\boldsymbol{\varepsilon}(\omega) \star \hat{\mathbf{E}}(\omega)](\mathbf{r}, \mathbf{r}') = \int \mathrm{d}^3 r' \boldsymbol{\varepsilon}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}(\mathbf{r}', \omega). \tag{4.3}$$

The two spatial variables are required to be independent when the medium has a non-local spatial response as they correspond to an observation point and a source point which are spatially separated. If the medium has a local spatial response, the source and observation points approximately coincide and the response functions will revert to an expression of the form $\boldsymbol{\varepsilon}(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{\varepsilon}(\mathbf{r}, \omega)\delta(\mathbf{r} - \mathbf{r}')$, etc., which means that the spatial convolution reduces to

$$\int \mathrm{d}^3 r' \boldsymbol{\varepsilon}(\mathbf{r}, \omega)\delta(\mathbf{r} - \mathbf{r}') \cdot \hat{\mathbf{E}}(\mathbf{r}', \omega) = \boldsymbol{\varepsilon}(\mathbf{r}, \omega) \cdot \hat{\mathbf{E}}(\mathbf{r}, \omega), \tag{4.4}$$

which is the standard form. Although there is no physical reason for choosing one convention over the other, the 'DB' response functions allow the construction of a duality invariant system. This can be seen by comparing Ampère's law, Eq. (2.1d), with Faraday's law, Eq. (2.1b) and noticing that the time dependent components are the displacement and magnetic induction fields respectively, and the fields subject to a curl operation are the magnetic and electric fields. This symmetry will be discussed in the present chapter.

In bianisotropic media the cross susceptibilies are introduced into the polarisation and magnetisation fields, which are defined as [33]

$$\hat{\mathbf{P}}_{DH} = \varepsilon_0 \boldsymbol{\chi}_{ee} \star \hat{\mathbf{E}} + \frac{1}{Z_0} \boldsymbol{\chi}_{em} \star \hat{\mathbf{B}} + \hat{\mathbf{P}}_{\mathrm{N}}, \tag{4.5a}$$

$$\hat{\mathbf{M}}_{DH} = \frac{1}{Z_0}\boldsymbol{\chi}_{me}\star\hat{\mathbf{E}} + \frac{1}{\mu_0}\boldsymbol{\chi}_{mm}\star\hat{\mathbf{B}} + \hat{\mathbf{M}}_{\mathrm{N}}, \tag{4.5b}$$

where $Z_0 = \sqrt{\mu_0/\varepsilon_0}$. The substitutions,

$$\boldsymbol{\chi}_{ee} = \boldsymbol{\varepsilon}_{DH} - \boldsymbol{I}, \tag{4.6a}$$

$$\boldsymbol{\chi}_{mm} = \boldsymbol{I} - \boldsymbol{\mu}_{DH}^{-1}, \tag{4.6b}$$

$$\boldsymbol{\chi}_{em} = \boldsymbol{\zeta}_{DH}, \tag{4.6c}$$

$$\boldsymbol{\chi}_{me} = \boldsymbol{\xi}_{DH}, \tag{4.6d}$$

are made so the polarisation field and magnetisation field can be written as

$$\hat{\mathbf{P}}_{DH} = \varepsilon_0(\boldsymbol{\varepsilon}_{DH} - \boldsymbol{I})\star\hat{\mathbf{E}} + \frac{1}{Z_0}\boldsymbol{\zeta}_{DH}\star\hat{\mathbf{B}} + \hat{\mathbf{P}}_{\mathrm{N}}, \tag{4.7a}$$

$$\hat{\mathbf{M}}_{DH} = \frac{1}{Z_0}\boldsymbol{\xi}_{DH}\star\hat{\mathbf{E}} + \frac{1}{\mu_0}(\boldsymbol{I} - \boldsymbol{\mu}_{DH}^{-1})\star\hat{\mathbf{B}} + \hat{\mathbf{M}}_{\mathrm{N}}. \tag{4.7b}$$

The constitutive relations in the 'DH' convention are obtained by substituting Eqs. (4.7a) and (4.7b) into Eq. (2.3),

$$\hat{\mathbf{D}} = \varepsilon_0\boldsymbol{\varepsilon}_{DH}\star\hat{\mathbf{E}} + \frac{1}{Z_0}\boldsymbol{\zeta}_{DH}\star\hat{\mathbf{B}} + \hat{\mathbf{P}}_{\mathrm{N}}, \tag{4.8a}$$

$$\hat{\mathbf{H}} = \frac{1}{\mu_0}\boldsymbol{\mu}_{DH}^{-1}\star\hat{\mathbf{B}} - \frac{1}{Z_0}\boldsymbol{\xi}_{DH}\star\hat{\mathbf{E}} - \hat{\mathbf{M}}_{\mathrm{N}}. \tag{4.8b}$$

In the 'DB' convention, the polarisation and magnetisation fields are found by substituting Eqs. (4.2a) to (4.2d) into Eqs. (4.7a) and (4.7b),

$$\hat{\mathbf{P}} = \varepsilon_0(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} - \boldsymbol{I})\star\hat{\mathbf{E}} + \frac{1}{Z_0}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\hat{\mathbf{B}} + \hat{\mathbf{P}}_{\mathrm{N}}, \tag{4.9a}$$

$$\hat{\mathbf{M}} = \frac{1}{Z_0}\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\hat{\mathbf{E}} + \frac{1}{\mu_0}(\boldsymbol{I} - \boldsymbol{\mu}^{-1})\star\hat{\mathbf{B}} + \hat{\mathbf{M}}_{\mathrm{N}}. \tag{4.9b}$$

Throughout the rest of the thesis the 'DB' convention will be used with the notation suppressed. By using the inhomogeneous macroscopic Maxwell's equations, Eqs. (2.1), and the general terms for the constitutive relations, Eq. (2.3), the constitutive relations are written as

$$\hat{\mathbf{D}} = \varepsilon_0\boldsymbol{\varepsilon}\star\hat{\mathbf{E}} + \frac{1}{c}\boldsymbol{\xi}\star\hat{\mathbf{H}} + \hat{\mathbf{P}}_{\mathrm{N}} + \frac{1}{c}\boldsymbol{\xi}\star\hat{\mathbf{M}}_{\mathrm{N}}, \tag{4.10a}$$

$$\hat{\mathbf{B}} = \frac{1}{c}\boldsymbol{\zeta}\star\hat{\mathbf{E}} + \mu_0\boldsymbol{\mu}\star\hat{\mathbf{H}} + \mu_0\boldsymbol{\mu}\star\hat{\mathbf{M}}_{\mathrm{N}}. \tag{4.10b}$$

All the response functions have been defined as dimensionless and they are assumed to be causal, so the real and imaginary parts are linked by a Kramers-Kronig relation, and they exhibit loss, i.e., $\mathcal{I}\mathrm{m}[\boldsymbol{\varepsilon}] > 0$, etc. It should also be noted that the bianisotropic response functions are not independent of each other, for example, their dissipative parts are constrained by the dissipative parts of the permittivity and permeability [19].

The wave equation for an electric field propagating through such a medium is

$$\left[\nabla\times\boldsymbol{\mu}^{-1}\star\nabla\times - \frac{i\omega}{c}\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} \;+ \frac{i\omega}{c}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\right]\star\hat{\mathbf{E}} = i\mu_0\omega\hat{\mathbf{j}}_{\mathrm{N}}. \tag{4.11}$$

where it has been assumed that the fields are all time harmonic. The wave equation for the electric field

is solved by

$$\hat{\mathbf{E}} = i\mu_0\omega \boldsymbol{G} \star \hat{\mathbf{j}}_{\mathrm{N}}, \tag{4.12}$$

and so the Green's function, $\boldsymbol{G}$, is a solution to

$$\left[\nabla\times\boldsymbol{\mu}^{-1}\star\nabla\times - \frac{i\omega}{c}\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} \; + \frac{i\omega}{c}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\right]\star\boldsymbol{G} = \boldsymbol{\delta}. \tag{4.13}$$

This Green's function is causal, constrained in the limit of diverging source and observation points and obeys the Schwarz reflection principle, Eq. (2.15). Using the constitutive relations Eqs. (4.10b) and (4.10a), the other electromagnetic fields can be written in terms of this Green's function as

$$\hat{\mathbf{B}} = \mu_0\nabla\times\boldsymbol{G}\star\hat{\mathbf{j}}_{\mathrm{N}}, \tag{4.14a}$$

$$\hat{\mathbf{D}} = \frac{i\omega}{c^2}\left(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\right)\star\boldsymbol{G}\star\hat{\mathbf{j}}_{\mathrm{N}} + \frac{1}{c}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G}\star\hat{\mathbf{j}}_{\mathrm{N}}, \tag{4.14b}$$

$$\hat{\mathbf{H}} = \boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G}\star\hat{\mathbf{j}}_{\mathrm{N}} - \frac{i\omega}{c}\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\boldsymbol{G}\star\hat{\mathbf{j}}_{\mathrm{N}}. \tag{4.14c}$$

When the Green's function for the electric field is known, Eq. (4.12), these results can be used to find the Green's functions that solve for the other fields in the source quantity representation, without solving a new wave equation. They will also be a useful reference when dealing with boundary conditions in a later chapter. As the Helmholtz equation, Eq. (4.13), is of the general form $\boldsymbol{H}\star\boldsymbol{G} = \boldsymbol{\delta}$, a useful integral relation in terms of the response functions can be obtained, as is described in Appendix D.

The definition of the induced current in a medium is

$$\hat{\mathbf{j}}_{\mathrm{in}} = -i\omega\hat{\mathbf{P}} + \nabla\times\hat{\mathbf{M}}, \tag{4.15}$$

which can be compared with the generalised Ohms law given in Eq. (3.3) to find an explicit form of the complex conductivity tensor in terms of the medium's response tensors as

$$\boldsymbol{Q} = \frac{1}{i\mu_0\omega}\nabla\times(\boldsymbol{\mu}^{-1} - \boldsymbol{I})\times\overleftarrow{\nabla} + \frac{1}{Z_0}\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} + \frac{1}{Z_0}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\times\overleftarrow{\nabla} - i\varepsilon_0\omega(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} - \boldsymbol{I}). \tag{4.16}$$

In the previous chapter it was noted that in spatially non-local media the distinction between electric and magnetic effects is not uniquely defined, so it would be possible to express the conductivity tensor without explicit reference to the magnetic effects and yet it would still hold all the relevant information on the medium. However, the formal separation of magnetic and electric effects can be useful in classifying and comparing classes of media and they can be determined experimentally [72]. This is why clearly defined response functions governing electric and magnetic effects are still useful when dealing with bianisotropic media. The general integral relation Eq. (3.15) and the noise current commutation relation Eq. (3.8) are dependent on the generalised real part of the conductivity, which is

$$\begin{aligned}\mathcal{R}\mathrm{e}[\boldsymbol{Q}] = &-\frac{1}{\omega\mu_0}\nabla\times\mathcal{I}\mathrm{m}[\boldsymbol{\mu}^{-1}]\times\overleftarrow{\nabla} - \omega\varepsilon_0\mathcal{I}\mathrm{m}[\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}] + \frac{1}{2Z_0}\Big(\nabla\times(\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} - \boldsymbol{\mu}^{-1\dagger}\star\boldsymbol{\xi}^{\dagger}) \\ &+ (\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1} - \boldsymbol{\zeta}^{\dagger}\star\boldsymbol{\mu}^{-1\dagger})\times\overleftarrow{\nabla}\Big),\end{aligned} \tag{4.17}$$

in terms of the explicit response functions. The constitutive relations Eqs. (4.10a) and (4.10b) account for the absorption into the medium by components of the noise current, a noise polarisation and a noise magnetisation, which form the noise current by the prescription shown in Eq. (2.9). This decomposition

is not unique but it is consistent as long the condition

$$\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)=\sum_{\lambda}\hat{\mathbf{j}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)=\sum_{\lambda'}\hat{\mathbf{J}}_{\mathrm{N}\lambda'}(\mathbf{r},\omega), \tag{4.18}$$

is met [40]. In Eq. (4.18), λ, λ' denote different decomposition modes, such as $\lambda = e, m$ etc., and $\hat{\mathbf{j}}_{\mathrm{N}\lambda}$, $\hat{\mathbf{J}}_{\mathrm{N}\lambda'}$ are components of the noise current in these different decompositions. By substituting the definition Eq. (2.9) into the commutation relation Eq. (3.8) and comparing terms, the commutation relations between the components of the noise current are

$$\left[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{P}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right]=\frac{\varepsilon_0\hbar}{\pi}\mathcal{I}\mathrm{m}\left[\boldsymbol{\varepsilon}(\omega)-\boldsymbol{\xi}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\zeta}(\omega)\right](\mathbf{r},\mathbf{r}')\delta(\omega-\omega'), \tag{4.19a}$$

$$\left[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{M}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right]=\frac{\hbar}{2\pi i Z_0}\left\{[\boldsymbol{\xi}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)]-[\boldsymbol{\zeta}^{\dagger}(\omega)\star\boldsymbol{\mu}^{-1\dagger}(\omega)]\right\}(\mathbf{r},\mathbf{r}')\delta(\omega-\omega'), \tag{4.19b}$$

$$\left[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{P}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right]=\frac{\hbar}{2\pi i Z_0}\left\{[\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\zeta}(\omega)]-[\boldsymbol{\mu}^{-1\dagger}(\omega)\star\boldsymbol{\xi}^{\dagger}(\omega)]\right\}(\mathbf{r},\mathbf{r}')\delta(\omega-\omega'), \tag{4.19c}$$

$$\left[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{M}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right]=-\frac{\hbar}{\pi\mu_0}\mathcal{I}\mathrm{m}[\boldsymbol{\mu}^{-1}(\mathbf{r},\mathbf{r}',\omega)]\delta(\omega-\omega'). \tag{4.19d}$$

The splitting of the commutation relation, Eq. (3.8), into the commutation relations, Eq. (4.19), is only valid for the decomposition outlined in Eq. (2.9) and a different splitting of the noise current will generate different commutation relations between the components of the noise current.

With the noise current split into a noise polarisation and noise magnetisation and the associated commutation relations, the bosonic matter-field system operators introduced in Eq. (3.30) can be decomposed into electric and magnetic modes, denoted by $\hat{\mathbf{f}}_\lambda$ and $\mathbf{f}^{\dagger}_\lambda$ for $\lambda = e, m$. They are introduced under the specification that

$$\begin{pmatrix}\hat{\mathbf{P}}_{\mathrm{N}}\\ \hat{\mathbf{M}}_{\mathrm{N}}\end{pmatrix}=\sqrt{\frac{\hbar}{\pi}}\mathcal{R}\star\begin{pmatrix}\hat{\mathbf{f}}_e\\ \hat{\mathbf{f}}_m\end{pmatrix} \tag{4.20}$$

where $\mathcal{R}$ is a (6×6)-matrix which is a (positive) square root of

$$\mathcal{R}\star\mathcal{R}^{\dagger}=\begin{pmatrix}\varepsilon_0\mathcal{I}\mathrm{m}[\boldsymbol{\varepsilon}-\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}] & \dfrac{\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}-\boldsymbol{\zeta}^{\dagger}\star\boldsymbol{\mu}^{-1\dagger}}{2iZ_0}\\ \dfrac{\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}-\boldsymbol{\mu}^{-1\dagger}\star\boldsymbol{\xi}^{\dagger}}{2iZ_0} & -\dfrac{\mathcal{I}\mathrm{m}[\boldsymbol{\mu}^{-1}]}{\mu_0}\end{pmatrix}. \tag{4.21}$$

With the above definitions, it can be shown that the commutation relation between the system excitations is

$$\left[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega),\hat{\mathbf{f}}^{\dagger}_{\lambda'}(\mathbf{r}',\omega')\right]=\delta_{\lambda\lambda'}\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega') \tag{4.22}$$

and the Hamiltonian of the matter-field system is now

$$\hat{H}_{\mathrm{F}}=\sum_{\lambda=e,m}\int\mathrm{d}^3r\int_0^{\infty}\mathrm{d}\omega\,\hbar\omega\,\hat{\mathbf{f}}^{\dagger}_\lambda(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega). \tag{4.23}$$

The difference between the quantisation scheme in chapter 3, in terms of a single bosonic vector field $\hat{\mathbf{f}}(\mathbf{r},\omega)$, and the quantisation in terms of $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ should be noted. The single bosonic vector field $\hat{\mathbf{f}}(\mathbf{r},\omega)$ can be split into different modes

$$\hat{\mathbf{f}}(\mathbf{r},\omega)=\sum_{\lambda}\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega),\quad \hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)=\int\mathrm{d}^3r'\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}(\mathbf{r}',\omega), \tag{4.24}$$

where $\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega)$ is a projection operator. Substituting Eq. (4.24) into Eq. (3.32) leads to a non-bosonic

commutation relation of the form

$$\left[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega),\hat{\mathbf{f}}^\dagger_{\lambda'}(\mathbf{r}',\omega')\right]=\delta_{\lambda\lambda'}\delta(\omega-\omega')\boldsymbol{P}(\mathbf{r},\mathbf{r}',\omega), \tag{4.25}$$

and a Hamiltonian, Eq. (4.23) can be specified. The quantisation procedure can be based on Eq. (4.22) instead of Eq. (4.25), although after this it is not possible to reverse engineer the system back to one in which $\hat{\mathbf{f}}(\mathbf{r},\omega)$ is the sole dynamical variable [40].

4.2 Reciprocal Media

Up to this point there have not been any assumptions made about the medium's response to applied electric fields, other than that the response is linear and causal. If the medium is reciprocal a relationship between the cross response tensors, $\boldsymbol{\xi}$ and $\boldsymbol{\zeta}$, can be found by considering the explicit form of the complex conductivity tensor as shown in Eq. (4.16). If the conductivity tensor describes the response of a reciprocal medium, it is subject to the constraint $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{Q}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)$. As the full conductivity tensor is reciprocal it can be assumed that each term in the explicit form is reciprocal, so

$$\boldsymbol{\varepsilon}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{\varepsilon}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega), \tag{4.26a}$$

$$\boldsymbol{\mu}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{\mu}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega), \tag{4.26b}$$

are constraints on the permittivity and the permeability. The conductivity tensor can be split into 3 parts; a term without a curl operation, components with one curl operation and a term with two curl operations. Considering the components with two curl operations from $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{Q}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)$, it can be seen that

$$\nabla\times\boldsymbol{\mu}^{-1}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\nabla}'=\nabla'\times(\boldsymbol{\mu}^{-1})^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)\times\overleftarrow{\nabla}, \tag{4.27}$$

when Eq. (4.26b) is used. The subtraction of the components of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ that contain a single curl operator from the components in $\boldsymbol{Q}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)$ that contain a single curl operator must be equal to zero. In coordinate space this is

$$\begin{aligned}&\int \mathrm{d}^3s\big[\nabla\times\big(\boldsymbol{\mu}^{-1}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\zeta}(\mathbf{s},\mathbf{r}',\omega)+\boldsymbol{\mu}^{-1\mathrm{T}}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\xi}^{\mathrm{T}}(\mathbf{s},\mathbf{r}',\omega)\big)\\&+\big(\boldsymbol{\xi}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\mu}^{-1}(\mathbf{s},\mathbf{r}',\omega)+\boldsymbol{\zeta}^{\mathrm{T}}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\mu}^{-1\mathrm{T}}(\mathbf{s},\mathbf{r}',\omega)\big)\times\overleftarrow{\nabla}'\big]=\mathbf{0}.\end{aligned} \tag{4.28}$$

As both terms need to be zero independently, this leads to the relationship between the cross tensors that must be satisfied in a reciprocal medium,

$$\boldsymbol{\zeta}(\mathbf{r},\mathbf{s},\omega)=-\boldsymbol{\xi}^{\mathrm{T}}(\mathbf{s},\mathbf{r},\omega). \tag{4.29}$$

Finally, the components with no curl operations from $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{Q}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)$ must be equal

$$\boldsymbol{\varepsilon}(\mathbf{r},\mathbf{r}',\omega)-[\boldsymbol{\xi}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\zeta}(\omega)](\mathbf{r},\mathbf{r}')=\boldsymbol{\varepsilon}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)-[\boldsymbol{\zeta}^{\mathrm{T}}(\omega)\star(\boldsymbol{\mu}^{-1}(\omega))^{\mathrm{T}}\star\boldsymbol{\xi}^{\mathrm{T}}(\omega)](\mathbf{r}',\mathbf{r}), \tag{4.30}$$

which can be confirmed by substituting in Eqs. (4.26a) and (4.29). An alternative approach is to consider the terms from the Post constitutive relations [59] where the cross tensors are split into reciprocal and non-reciprocal parts and are assigned the forms

$$\boldsymbol{\zeta}=\boldsymbol{\chi}+i\boldsymbol{\kappa}, \tag{4.31a}$$

$$\boldsymbol{\xi} = \boldsymbol{\chi}^{\mathrm{T}} - i\boldsymbol{\kappa}^{\mathrm{T}}, \tag{4.31b}$$

where $\boldsymbol{\chi}$ describes that non-reciprocal response and $\boldsymbol{\kappa}$ is the reciprocal (and chiral) response. Rearranging these to give

$$\boldsymbol{\chi} = \frac{1}{2i}(\boldsymbol{\zeta} + \boldsymbol{\xi}^{\mathrm{T}}), \tag{4.32a}$$

$$\boldsymbol{\kappa} = \frac{1}{2i}(\boldsymbol{\zeta} - \boldsymbol{\xi}^{\mathrm{T}}), \tag{4.32b}$$

there is an explicit term for the non-reciprocal response of a medium in terms of its cross susceptibility tensors. When the medium is reciprocal, $\boldsymbol{\chi}$ is zero, which means that the cross tensors are related by $\boldsymbol{\zeta} = -\boldsymbol{\xi}^{\mathrm{T}}$, consistent with the expression found earlier in Eq. (4.29).

4.3 Duality

It has been shown that Maxwell's equations (without external sources) in a macroscopic QED framework can be written in a duality invariant form for an isotropic, linear medium [71]. In this section, whose results are presented in Ref. [56], the duality system is extended to incorporate a general linear bianisotropic medium. The duality transformation on an object is

$$\begin{pmatrix} \mathbf{x} \\ \mathbf{y} \end{pmatrix}^{\circledast} = D(\theta) \begin{pmatrix} \mathbf{x} \\ \mathbf{y} \end{pmatrix}, \qquad D(\theta) = \begin{pmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{pmatrix}, \tag{4.33}$$

where the superscript $^{\circledast}$ denotes the objects dual. An object is said to be invariant under the duality transformation if it retains the same mathematical structure, i.e., the relationships between the dual objects are the same as the relationships between the original objects. Starting with Maxwell's sourceless inhomogeneous equations, these can be written in a duality invariant form as

$$\nabla \cdot \begin{pmatrix} Z_0\hat{\mathbf{D}} \\ \hat{\mathbf{B}} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \tag{4.34a}$$

$$\nabla \times \begin{pmatrix} \hat{\mathbf{E}} \\ Z_0\hat{\mathbf{H}} \end{pmatrix} - i\omega \boldsymbol{Z} \begin{pmatrix} Z_0\hat{\mathbf{D}} \\ \hat{\mathbf{B}} \end{pmatrix} = \begin{pmatrix} \mathbf{0} \\ \mathbf{0} \end{pmatrix}, \quad \boldsymbol{Z} = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}. \tag{4.34b}$$

The pairings of ($\hat{\mathbf{E}}$, $\hat{\mathbf{H}}$) and ($\hat{\mathbf{D}}$, $\hat{\mathbf{B}}$) are made for mathematical reasons only, following the symmetries of Maxwell's equations. This symmetry is the motivation for the choice of response functions in the 'DB' convention as discussed earlier. For Maxwell's equations to be duality invariant, applying Eq. (4.33) to Eq. (4.34) must not change the structure of the equations. As the duality operator commutes with the nabla operator ∇ and the symplectic matrix $\boldsymbol{Z}$ it is clear that Maxwell's equations in the form Eqs. (4.34) are duality invariant. The extention of duality invariance to a general linear bianisotropic medium starts by writing the constitutive relations Eqs. (4.10a) and (4.10b) in the condensed form

$$\begin{pmatrix} Z_0\hat{\mathbf{D}} \\ \hat{\mathbf{B}} \end{pmatrix} = \frac{1}{c} \begin{pmatrix} \boldsymbol{\varepsilon} & \boldsymbol{\xi} \\ \boldsymbol{\zeta} & \boldsymbol{\mu} \end{pmatrix} \star \begin{pmatrix} \hat{\mathbf{E}} \\ Z_0\hat{\mathbf{H}} \end{pmatrix} + \begin{pmatrix} \boldsymbol{I} & \boldsymbol{\xi} \\ 0 & \boldsymbol{\mu} \end{pmatrix} \star \begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}. \tag{4.35}$$

The duals of the electromagnetic fields are found by multiplying the field pairs on the left hand side by the duality matrix, $D(\theta)$,

$$\begin{pmatrix} \hat{\mathbf{E}} \\ Z_0\hat{\mathbf{H}} \end{pmatrix}^{\circledast} = D(\theta) \begin{pmatrix} \hat{\mathbf{E}} \\ Z_0\hat{\mathbf{H}} \end{pmatrix} \tag{4.36a}$$

$$\begin{pmatrix} Z_0\hat{\mathbf{D}} \\ \hat{\mathbf{B}} \end{pmatrix}^{\circledast} = D(\theta)\begin{pmatrix} Z_0\hat{\mathbf{D}} \\ \hat{\mathbf{B}} \end{pmatrix}. \tag{4.36b}$$

Now that the transformations of the electromagnetic fields are known, to ensure that the constitutive relations, Eqs. (4.35), are invariant under the duality transformation, the response tensors must transform as

$$\begin{pmatrix} \boldsymbol{\varepsilon} & \boldsymbol{\xi} \\ \boldsymbol{\zeta} & \boldsymbol{\mu} \end{pmatrix}^{\circledast} = D(\theta)\begin{pmatrix} \boldsymbol{\varepsilon} & \boldsymbol{\xi} \\ \boldsymbol{\zeta} & \boldsymbol{\mu} \end{pmatrix}D^{-1}(\theta) \tag{4.37}$$

which can be rewritten as

$$\begin{pmatrix} \boldsymbol{\varepsilon} \\ \boldsymbol{\zeta} \\ \boldsymbol{\xi} \\ \boldsymbol{\mu} \end{pmatrix}^{\circledast} = \mathcal{D}(\theta)\begin{pmatrix} \boldsymbol{\varepsilon} \\ \boldsymbol{\zeta} \\ \boldsymbol{\xi} \\ \boldsymbol{\mu} \end{pmatrix} \tag{4.38}$$

where

$$\begin{aligned} \mathcal{D}(\theta) &= D(\theta) \otimes D(\theta) \\ &= \begin{pmatrix} \cos^2\theta & \sin\theta\cos\theta & \sin\theta\cos\theta & \sin^2\theta \\ -\sin\theta\cos\theta & \cos^2\theta & -\sin^2\theta & \sin\theta\cos\theta \\ -\sin\theta\cos\theta & -\sin^2\theta & \cos^2\theta & \sin\theta\cos\theta \\ \sin^2\theta & -\sin\theta\cos\theta & -\sin\theta\cos\theta & \cos^2\theta \end{pmatrix}. \end{aligned} \tag{4.39}$$

The second term on the right hand side of Eq. (4.35) must transform as

$$\begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}^{\circledast} = (\mathcal{A}^{\circledast})^{-1}\star D(\theta)\mathcal{A}\star\begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}. \tag{4.40}$$

where

$$\mathcal{A} = \begin{pmatrix} \boldsymbol{I} & \boldsymbol{\xi} \\ \mathbf{0} & \boldsymbol{\mu} \end{pmatrix}, \tag{4.41a}$$

$$\mathcal{A}^{-1} = \begin{pmatrix} \boldsymbol{I} & -\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1} \\ \mathbf{0} & \boldsymbol{\mu}^{-1} \end{pmatrix}, \tag{4.41b}$$

$$\mathcal{A}^{\circledast} = \begin{pmatrix} \boldsymbol{I} & \boldsymbol{\xi}^{\circledast} \\ \mathbf{0} & \boldsymbol{\mu}^{\circledast} \end{pmatrix}. \tag{4.41c}$$

to guarantee that the system is invariant under the duality transformation. The matrix $\mathcal{A}$ has been introduced solely as a notational shorthand. The term $(\mathcal{A}^{\circledast})^{-1}$ refers to the inverse of the dual of $\mathcal{A}$. Equation (4.40) is written in terms of the dual of $\mathcal{A}$ for notational convenience only, as the dual of all the terms in $\mathcal{A}$ are known from Eq. (4.38).

For a local and isotropic medium, the clear relationship between the noise current components and the matter-field system excitations (Eqs. (2.24a) and (2.24b) in section 2.1.3) allows the duality transformations of these excitations to be found. However, in the general bianisotropic case the explicit relationships between the noise polarisation and magnetisation and the electric and magnetic excitations is not trivial (the relationship is shown in Eq. (4.20)) and therefore the general duality transformations are not shown.

This duality invariant description is valid for the most general linear bianisotropic media, including non-reciprocal media. In this general case there are no constraints on the possible values that the rotation angle θ can take for duality invariance to hold, therefore it is said to have a continuous duality symmetry, the only other circumstance where duality symmetry can be described as continuous is the

case of impedance matching. If the medium is reciprocal, the relationship $\boldsymbol{\zeta} = -\boldsymbol{\xi}^{\mathrm{T}}$ between the cross tensors can be applied to Eq. (4.38) which results in

$$\begin{pmatrix} \boldsymbol{\varepsilon} & -\boldsymbol{\zeta}^{\mathrm{T}} \\ \boldsymbol{\zeta} & \boldsymbol{\mu} \end{pmatrix}^{\circledast} = \begin{pmatrix} \boldsymbol{\varepsilon}\cos^2\theta + \boldsymbol{\mu}\sin^2\theta & \cos\theta\sin\theta(\boldsymbol{\mu}-\boldsymbol{\varepsilon}) \\ \cos\theta\sin\theta(\boldsymbol{\mu}-\boldsymbol{\varepsilon}) & \boldsymbol{\varepsilon}\sin^2\theta + \boldsymbol{\mu}\cos^2\theta \end{pmatrix} - \boldsymbol{\zeta}\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}. \tag{4.42}$$

It can be seen that the first term on the right hand side of Eq. (4.42) are the duals of the response functions that are obtained for anisotropic media, which is a simple extension of the results in Ref. [71]. The matrix in the second term on the right hand side is the cross tensor multiplied by the symplectic matrix, which commutes with the duality operator. Therefore, for reciprocal bianisotropic media, the duality symmetry is discrete and holds only for a restricted group of values for θ, which are $\theta = \frac{n\pi}{2}$ where $n \in \mathbb{Z}$. The reason for this is that for the reciprocal condition, Eq. (4.29), only holds for the duals of the cross response functions when the opposite diagonal terms in the first matrix on the right hand side of Eq. (4.42) are zero. While the angle, θ, can take on 4 possible values, each value corresponds to a varying degree of mixing between the electric permittivity and magnetic permability and electric and magnetic effects in general. In this case the duality operation does not have an effect on terms involving the cross tensor, it remains the same regardless of the mixing angle between the electric and magnetic effects. An explanation of this is that the coupling tensor is, by definition, mixing electric and magnetic effects in an equal and opposite manner, so a global exchange of electric and magnetic properties will only affect the exclusively electric or magnetic terms. The exception to this is the special case of impedance matching when $\varepsilon = \mu$, from Eq. (4.42) it can be seen that in this scenario the duality symmetry is continuous.

The description of general bianisotropic media encompasses the behaviour of some special classes of media, which will be discussed. For example, if the medium is an isotropic magnetodielectric the response functions take the form

$$\boldsymbol{\varepsilon} = \varepsilon\boldsymbol{I},\ \boldsymbol{\mu} = \mu\boldsymbol{I},\ \boldsymbol{\zeta} = \boldsymbol{\xi} = \boldsymbol{0}. \tag{4.43}$$

The medium is reciprocal so the duality symmetry will be discrete, as is discussed in Ref. [71]. The components of the noise current, $\hat{\mathbf{P}}_{\mathrm{N}}$ and $\hat{\mathbf{M}}_{\mathrm{N}}$ commute with each other and the generalised real and imaginary parts, seen in Eq. (4.19), revert to the standard real and imaginary parts. In isotropic and anisotropic media, there is no cross coupling between applied electric and magnetic induction fields and the associated displacement and magnetic fields. This means that the cross tensors do not exist, the noise polarisation and noise magnetisation, $\hat{\mathbf{P}}_{\mathrm{N}}$ and $\hat{\mathbf{M}}_{\mathrm{N}}$, commute and the duality invariance symmetry is discrete. The difference between isotropic and anisotropic media are that in an anisotropic medium, the electric and displacement fields are not aligned due to a tensorial coupling term, the permittivity. The same is true of the permeability as the coupling between the magnetic and magnetic induction fields. An anisotropic medium can also be inhomogeneous, where the response is dependent on position, and have a non-local spatial response to applied fields. In anisotropic media the commutation relations become

$$\left[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{P}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \frac{\varepsilon_0\hbar}{\pi}\mathcal{I}\mathrm{m}\left[\boldsymbol{\varepsilon}(\mathbf{r},\mathbf{r}',\omega)\right]]\,\delta(\omega-\omega'), \tag{4.44a}$$

$$\left[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{M}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \left[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{P}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \boldsymbol{0}, \tag{4.44b}$$

$$\left[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{M}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = -\frac{\hbar}{\pi\mu_0}\mathcal{I}\mathrm{m}[\boldsymbol{\mu}^{-1}(\mathbf{r},\mathbf{r}',\omega)]\delta(\omega-\omega'). \tag{4.44c}$$

For a local anisotropic medium the substitutions $\boldsymbol{\varepsilon}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{\varepsilon}(\mathbf{r},\omega)\delta(\mathbf{r}-\mathbf{r}')$ and $\boldsymbol{\mu}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{\mu}(\mathbf{r},\omega)\delta(\mathbf{r}-\mathbf{r}')$ are made and for an isotropic medium, $\boldsymbol{\varepsilon}(\mathbf{r},\mathbf{r}',\omega) = \varepsilon(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')$ and $\boldsymbol{\mu}(\mathbf{r},\mathbf{r}',\omega) = \mu(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')$. For a locally responding anisotropic medium, the relations between the noise operators and the dynamical

variables are known and can be constructed from Eqs. (2.24) in the form

$$\begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix} = \sqrt{\frac{\hbar\mu_0}{\pi}} \begin{pmatrix} i\sqrt{\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)]} & 0 \\ 0 & \sqrt{-\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)]} \end{pmatrix} \begin{pmatrix} \hat{\mathbf{f}}_e \\ \hat{\mathbf{f}}_m \end{pmatrix}. \tag{4.45}$$

The duals of the noise current components for anisotropic media can be found by setting $\boldsymbol{\xi} = \mathbf{0}$ in Eq. (4.40) to obtain

$$\begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}^{\circledast} = \begin{pmatrix} \boldsymbol{I}\cos\theta & \boldsymbol{\mu}\sin\theta \\ -\boldsymbol{\varepsilon}^{-1}\sin\theta & \boldsymbol{I}\cos\theta \end{pmatrix} \begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}. \tag{4.46}$$

It can then be shown by substituting Eq. (4.46) into the dual form of Eq. (4.45) that the duals of the matter-field excitations are

$$\begin{pmatrix} \hat{\mathbf{f}}_e \\ \hat{\mathbf{f}}_m \end{pmatrix}^{\circledast} = \begin{pmatrix} \boldsymbol{I}\cos\theta & -i\boldsymbol{\mu}\mathrm{Im}[\boldsymbol{\mu}^{-1}]\sin\theta \\ -i\frac{\mathrm{Im}[\boldsymbol{\varepsilon}]}{\boldsymbol{\varepsilon}}\sin\theta & \boldsymbol{I}\cos\theta \end{pmatrix} \begin{pmatrix} \hat{\mathbf{f}}_e \\ \hat{\mathbf{f}}_m \end{pmatrix}. \tag{4.47}$$

Biisotropic media is a subclass of bianisotropic media where the response functions are all isotropic, i.e.,

$$\boldsymbol{\varepsilon} = \varepsilon\boldsymbol{I},\ \boldsymbol{\mu} = \mu\boldsymbol{I},\ \boldsymbol{\xi} = \xi\boldsymbol{I},\ \boldsymbol{\zeta} = \zeta\boldsymbol{I}. \tag{4.48}$$

The noise current components do not not commute, but simplify from Eq. (4.19) to

$$\left[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{P}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \frac{\varepsilon_0\hbar}{\pi}\mathrm{Im}[\varepsilon(\omega) + \zeta^2(\omega)\mu^{-1}(\omega)]\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega') \tag{4.49a}$$

$$\left[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{M}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = -\frac{\hbar}{2\pi i Z_0}\mathrm{Re}[\zeta(\omega)\mu^{-1}(\omega)]\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega') \tag{4.49b}$$

$$\left[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{P}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \frac{\hbar}{2\pi i Z_0}\mathrm{Re}[\zeta(\omega)\mu^{-1}(\omega)]\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega') \tag{4.49c}$$

$$\left[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega), \hat{\mathbf{M}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = -\frac{\hbar}{\pi\mu_0}\mathrm{Im}[\mu^{-1}(\omega)]\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'). \tag{4.49d}$$

where the reciprocal relation between cross tensors has been used.

The next two chapters discuss two examples of bianisotropic media, chiral media (chapter 5) and moving media (chapter 6). As chiral media are reciprocal, they possess a discrete duality symmetry and the constitutive relations transform as shown in Eq. (4.42), with $\boldsymbol{\zeta} = i\boldsymbol{\kappa}$. Moving media can be described as non-reciprocal bianisotropic media from a laboratory frame, which means that the duality symmetry is continuous.

4.3.1 Duality Relation for Green's Functions

In the description of a physical scenario, a dispersion force between objects for example, the duality invariance of a system can be used to calculate unknown components from known components of the dispersion force. For example, when the dispersion forces between polarisable atoms are calculated, the duality invariance of the system can be used to calculate the dipersion forces between magnetisable atoms by making the necessary duality invariant substitutions. As the calculation of fields, dispersion forces, decay rates, etc., in macroscopic QED uses a Green's function formalism, to effectively use the duality symmetries the behaviour of the Green's function (and atomic or molecular properties) under duality invariant conditions are necessary. The definitions of the electric field, Eq. (3.10), and the noise current, Eq. (2.9), are rewritten as

$$\hat{\mathbf{E}} = i\omega\mu_0\boldsymbol{G}\star\hat{\mathbf{j}}_{\mathrm{N}} = -c\boldsymbol{G}_{ee}\star(Z_0\hat{\mathbf{P}}_{\mathrm{N}}) - c\boldsymbol{G}_{em}\star(\mu_0\hat{\mathbf{M}}_{\mathrm{N}}), \tag{4.50}$$

so the magnetic field in a form suitable for duality applications becomes

$$Z_0\hat{\mathbf{H}} = -c\big[(-\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\boldsymbol{G}_{ee} + \boldsymbol{\mu}^{-1}\star\boldsymbol{G}_{me})\star(Z_0\hat{\mathbf{P}}_{\mathrm{N}}) + (-\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\boldsymbol{G}_{em} + \boldsymbol{\mu}\star\boldsymbol{G}_{mm} + \boldsymbol{I})\star(\mu_0\hat{\mathbf{M}}_{\mathrm{N}})\big], \quad (4.51)$$

where the notational shorthands

$$\boldsymbol{G}_{ee} = (i\frac{\omega}{c})^2\boldsymbol{G}, \quad (4.52a)$$

$$\boldsymbol{G}_{em} = (i\frac{\omega}{c})\boldsymbol{G}\times\overleftarrow{\nabla}', \quad (4.52b)$$

$$\boldsymbol{G}_{me} = \nabla\times\boldsymbol{G}(i\frac{\omega}{c}), \quad (4.52c)$$

$$\boldsymbol{G}_{mm} = \nabla\times\boldsymbol{G}\times\overleftarrow{\nabla}' \quad (4.52d)$$

have been used. The displacement and magnetic induction fields can be written in a similar form

$$\begin{aligned} Z_0\hat{\mathbf{D}} = &-\big((\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\star\boldsymbol{G}_{ee} + \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{G}_{me} - \boldsymbol{I}\big)\star(Z_0\hat{\mathbf{P}}_{\mathrm{N}}) \\ &-\big((\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\star\boldsymbol{G}_{em} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{G}_{mm}\big)\star(\mu_0\hat{\mathbf{M}}_{\mathrm{N}}), \end{aligned} \quad (4.53a)$$

$$\hat{\mathbf{B}} = -\boldsymbol{G}_{me}\star(Z_0\hat{\mathbf{P}}_{\mathrm{N}}) - \boldsymbol{G}_{mm}\star(\mu_0\hat{\mathbf{M}}_{\mathrm{N}}). \quad (4.53b)$$

These can be combined into the duality consistent forms

$$\begin{pmatrix} \hat{\mathbf{E}} \\ Z_0\hat{\mathbf{H}} \end{pmatrix} = -c\mathcal{B}\star\mathcal{G}_{EH}\star\begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}, \quad (4.54a)$$

$$\begin{pmatrix} Z_0\hat{\mathbf{D}} \\ \hat{\mathbf{B}} \end{pmatrix} = -\mathcal{E}\star\mathcal{G}_{DB}\star\begin{pmatrix} Z_0\hat{\mathbf{P}}_{\mathrm{N}} \\ \mu_0\hat{\mathbf{M}}_{\mathrm{N}} \end{pmatrix}, \quad (4.54b)$$

where the matrices $\mathcal{B}$, $\mathcal{E}$, $\mathcal{G}_{EH}$ and $\mathcal{G}_{DB}$ are defined as

$$\mathcal{B} = \begin{pmatrix} \boldsymbol{I} & \boldsymbol{0} \\ -\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} & \boldsymbol{\mu}^{-1} \end{pmatrix}, \quad (4.55a)$$

$$\mathcal{E} = \begin{pmatrix} (\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}) & \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1} \\ \boldsymbol{0} & \boldsymbol{I} \end{pmatrix}, \quad (4.55b)$$

$$\mathcal{G}_{EH} = \begin{pmatrix} \boldsymbol{G}_{ee} & \boldsymbol{G}_{em} \\ \boldsymbol{G}_{me} & \boldsymbol{G}_{mm} + \boldsymbol{\mu} \end{pmatrix}, \quad (4.55c)$$

$$\mathcal{G}_{DB} = \begin{pmatrix} \boldsymbol{G}_{ee} - (\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})^{-1} & \boldsymbol{G}_{em} \\ \boldsymbol{G}_{me} & \boldsymbol{G}_{mm} \end{pmatrix}, \quad (4.55d)$$

respectively. The matrices of the Green tensors, $\mathcal{G}_{EH}$ and $\mathcal{G}_{DB}$, are invariant under the duality transformation if the dual matrices, $\mathcal{G}^{\circledast}_{EH}$ and $\mathcal{G}^{\circledast}_{DB}$ are of the form

$$\mathcal{G}^{\circledast}_{EH} = \mathcal{B}^{\circledast-1}\star D(\theta)\mathcal{B}\star\mathcal{G}_{EH}\star\mathcal{A}^{-1}\star D^{-1}(\theta)\mathcal{A}^{\circledast}, \quad (4.56a)$$

$$\mathcal{G}^{\circledast}_{DB} = \mathcal{E}^{\circledast-1}\star D(\theta)\mathcal{B}\star\mathcal{G}_{DB}\star\mathcal{A}^{-1}\star D^{-1}(\theta)\mathcal{A}^{\circledast}, \quad (4.56b)$$

where

$$\mathcal{B}^{-1} = \begin{pmatrix} \boldsymbol{I} & \boldsymbol{0} \\ \boldsymbol{\zeta} & \boldsymbol{\mu} \end{pmatrix}, \quad (4.57a)$$

$$\mathcal{E}^{-1} = \begin{pmatrix} (\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})^{-1} & -(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})^{-1}\star\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1} \\ \mathbf{0} & \boldsymbol{I} \end{pmatrix}. \tag{4.57b}$$

Although these are the general results for Green's functions describing an absorbing bianisotropic medium, care needs to be taken if the source and observation points are not in the same medium but are across a boundary, briefly discussed in appendix E. However, this does not present an issue when calculating dispersion forces as the source and observation point coincide and are therefore in the same medium. A common example of this is when the source and observation point are in free space near a boundary, as is the case with the Casimir-Polder force when a particle is near a macroscopic body, discussed in section 2.2. In this scenario the linking matrices $\mathcal{A}$, $\mathcal{B}$ and $\mathcal{E}$ become diagonal in $\boldsymbol{I}$, which commutes with the duality operation. This means that to be duality invariant the matrix of the duals of the Green functions, $\mathcal{G}^{\circledast}_{EH}$, is now defined as

$$\mathcal{G}^{\circledast}_{EH} = D(\theta)\mathcal{G}_{EH}\star D^{-1}(\theta). \tag{4.58}$$

This then becomes

$$\begin{pmatrix} \boldsymbol{G}_{ee} \\ \boldsymbol{G}_{em} \\ \boldsymbol{G}_{me} \\ \boldsymbol{G}_{mm}+\boldsymbol{I} \end{pmatrix}^{\circledast} = \mathcal{D}(\theta) \begin{pmatrix} \boldsymbol{G}_{ee} \\ \boldsymbol{G}_{em} \\ \boldsymbol{G}_{me} \\ \boldsymbol{G}_{mm}+\boldsymbol{I} \end{pmatrix}, \tag{4.59}$$

which shows that, in this example, the Green's functions transform as the medium's response functions, seen in Eq. (4.38). To complete the duality invariant description of a dispersion force, the dual of the atomic or molecular properties are needed, which can be found by a duality operation on the atomic and magnetic polarisation.

In this chapter the quantisation of the electromagnetic field in general linear bianisotropic media in terms of electric, magnetic and magnetoelectric response functions has been presented. The duality of electromagnetic systems has been introduced and used to explore the symmetry between electric and magnetic effects and how this can be utilised to theorise the behaviour of novel media. The next chapter will start with the quantisation of the electromagnetic field in a chiral medium, a special class of bianisotropic media. The chiral component of the Casimir-Polder potential will be derived and an example of its use will be detailed.

Chapter 5

Chiral Media and the Chiral Casimir-Polder Potential

A three-dimensional object that cannot be superimposed, through any combination of rotations and translations, onto its mirror image is said to be chiral. When the object in question is a molecule, these mirror images are called enantiomers. Enantiomers have the same chemical composition and molecular structure as each other, this means that the spectroscopic properties are identical and so distinguishing between the enantiomers is not trivial. The characteristic feature of chiral objects is the manner of their interactions with other chiral objects. For example, the refractive indices of left handed and right handed circularly polarised light are not the same in a chiral medium, therefore the two polarisations will propagate at different speeds through the chiral medium. The difference in velocity is related to the phenomenon of circular dichroism, where the wave with the 'slower' polarisation is subject to stronger absorption as it travels through the medium [73]. Many of the processes crucial to life involve chiral molecules whose chiral identity plays a central role in their chemical reactions, the incorrect enantiomer reacting differently and not producing the required result. In nature, these molecules tend to only occur as one enantiomer and are not found as the other, thus the desired reactions will only occur when the correct enantiomer is present. In contrast, artificial production creates both enantiomers in equal proportions. Therefore it is important to be able to distinguish between enantiomers and ultimately to be able to separate a racemic (containing both enantiomers) mixture into an enantiomerically pure sample.

A frequently used method to separate enantiomers in an industrial setting is chiral chromotography. The initial racemic solution is passed through a column packed with a resolving agent, which is usually an enzyme and by necessity is chiral. This enzyme either retards, or stops, the progress of one of the enantiomers passing through the column but crucially does not do this for the other enantiomer and thus allows the solution to be separated. From an optical viewpoint it has been proposed that a racemic sample can be purified by use of coordinated laser pulses, which use a two-step process to initially drive different transitions in the enantiomers before converting one into the other [74, 75]. It has recently been calculated that in the presence of a chiral carbon nanotube the enantiomers of alanine possess different absorption energies and it was theorised that this could lead to a method of discrimination [76]. Furthermore, it has been shown that the Van der Waals dispersion force between molecules can be enantiomer selective [77].

In the introduction to the Casimir-Polder potential given in section 2.2, the Casimir-Polder potential was separated into an electric component, dependent on the electric dipole moments of a particle and the electric response of a medium, and a magnetic contribution, dependent on the magnetic dipole

moments of a particle and the magnetic response of a medium. The terms that contained both electric and magnetic dipole moments were neglected on parity grounds. When the molecule is chiral these terms cannot be neglected and therefore they contribute to the total Casimir-Polder potential. It can reasonably be assumed that the chiral nature of the molecule and the chiral response of the medium will comprise this additional component to the Casimir-Polder potential.

Chiral molecules are common in nature and there has been much work to calculate the relevant transition values either ab initio [78, 79], or through a twisted arc model for simple chiral molecules [80]. Recent technological developments have allowed for the creation of chiral fullerenes such as C_{76} [81, 82], chiral carbon nanotubes [76, 83] and chiral metamaterials [84]. Chiral metamaterials can be made from structures such as a gold helix [85], which shows a broadband electromagnetic response; a woodpile structure [86], a gold bar construction [87], which exhibit a negative refractive index in certain frequency ranges; and gold dots [88], which can be tuned during construction to give a desired response. Recently it has been shown that superchiral electromagnetic fields [89] can arise in planar chiral metamaterials. These fields can generate a much larger dissymmetry between the effective refractive indices for absorbed chiral molecules on left- and right-handed materials than circularly polarised light and a solution of chiral molecules [90]. Chiral metamaterials have also been proposed as a method of producing repulsive Casimir forces [91].

The results of the previous chapter will be applied to chiral media to obtain the quantisation of the electromagnetic field in a chiral medium. The chiral component of the Casimir-Polder potential will be derived using a perturbative approach in section 5.3 and examples will be calculated, including a molecule near a perfect chiral mirror and a chiral metamaterial. Section 5.5 will then present an example of the chiral component of the Casimir-Polder potential used in a cavity set up in order to separate enantiomers. The work in this chapter has been previously published in Ref. [92].

5.1 Field Quantisation in Chiral Media

In chiral media, the response to an applied electric field is described by an electric permittivity and the response to an applied magnetic induction field is described by a magnetic permeability. The chiral response to applied electric and magnetic induction fields is characterised by a tensor, which couples the magnetic field to the displacement field and the electric field to the magnetic induction field. This means that chiral media is a class of bianisotropic media and therefore the description of a general bianisotropic medium given in chapter 4 can be applied to a chiral medium. In fact, the general bianisotropic formalism discussed in section 4.1 ensures that for a chiral medium with a linear causal response, the quantisation procedure and the compliance with the fluctuation dissipation theorem are guaranteed. The following section will detail how the general bianisotropic framework is applied to the specific example of chiral media. When media is reciprocal, the parameter $\boldsymbol{\chi}$ in the Post conditions, Eqs. (4.31a) and (4.31b), is zero and therefore the cross susceptibilities introduced in Eqs. (4.10a) and (4.10b) become

$$\boldsymbol{\zeta} = i\boldsymbol{\kappa}, \quad \boldsymbol{\xi} = -i\boldsymbol{\kappa}^{\mathrm{T}}, \tag{5.1}$$

when the medium is chiral. These are substituted into the constitutive relations for a general bianisotropic medium, Eqs. (4.10a) and (4.10b), and therefore

$$\hat{\mathbf{D}} = \varepsilon_0 \boldsymbol{\varepsilon} \star \hat{\mathbf{E}} - \frac{i}{c}\boldsymbol{\kappa}^{\mathrm{T}} \star \hat{\mathbf{H}} + \hat{\mathbf{P}}_{\mathrm{N}} - \frac{i}{c}\boldsymbol{\kappa}^{\mathrm{T}} \star \hat{\mathbf{M}}_{\mathrm{N}}, \tag{5.2a}$$

$$\hat{\mathbf{B}} = \frac{i}{c}\boldsymbol{\kappa} \star \hat{\mathbf{E}} + \mu_0 \boldsymbol{\mu} \star \hat{\mathbf{H}} + \mu_0 \boldsymbol{\mu} \star \hat{\mathbf{M}}_{\mathrm{N}}. \tag{5.2b}$$

are the constitutive relations for a linear absorbing chiral medium. The Helmholtz vector wave equation for the electric field in a chiral medium is

$$\begin{aligned}&\Big\{\Big[\nabla\times\boldsymbol{\mu}^{-1}(\omega)\star\nabla\times\boldsymbol{I}+\frac{\omega}{c}(\nabla\times\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\kappa}(\omega)+\boldsymbol{\kappa}^{\mathrm{T}}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\nabla\times\boldsymbol{I})\\&-\frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}(\omega)-\boldsymbol{\kappa}^{\mathrm{T}}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\kappa}(\omega))\Big]\star\hat{\mathbf{E}}(\omega)\Big\}(\mathbf{r})=i\omega\mu_0\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega).\end{aligned}\tag{5.3}$$

The electric field is defined as

$$\hat{\mathbf{E}}(\mathbf{r},\omega)=i\omega\mu_0\int\mathrm{d}^3r'\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}',\omega).\tag{5.4}$$

in coordinate space, where the Green's function is the fundamental solution to the inhomogeneous Helmholtz equation

$$\begin{aligned}&\Big\{\Big[\nabla\times\boldsymbol{\mu}^{-1}(\omega)\star\nabla\times\boldsymbol{I}+\frac{\omega}{c}(\nabla\times\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\kappa}(\omega)+\boldsymbol{\kappa}^{\mathrm{T}}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\nabla\times\boldsymbol{I})\\&-\frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}(\omega)-\boldsymbol{\kappa}^{\mathrm{T}}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\kappa})(\omega)\Big]\star\boldsymbol{G}(\omega)\Big\}(\mathbf{r},\mathbf{r}')=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}').\end{aligned}\tag{5.5}$$

The Green's function obeys the Schwarz reflection principle, Eq. (2.15), is causal and it is constrainted such that $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)=\mathbf{0}$ when $|\mathbf{r}-\mathbf{r}'|\to\infty$. As chiral media is reciprocal, the Green's function obeys the reciprocal condition, Eq. (2.16). The substitution of Eq. (5.1) into the commutation relations for the noise polarisation and noise magnetisation fields, Eqs. (4.19), leads to

$$[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{P}}_{\mathrm{N}}^{\dagger}(\mathbf{r}',\omega')]=\frac{\varepsilon_0\hbar}{\pi}\big\{\mathrm{Im}[\boldsymbol{\varepsilon}(\omega)-\boldsymbol{\kappa}^{\mathrm{T}}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\kappa}(\omega)]\big\}(\mathbf{r},\mathbf{r}')\delta(\omega-\omega'),\tag{5.6a}$$

$$[\hat{\mathbf{P}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{M}}_{\mathrm{N}}^{\dagger}(\mathbf{r}',\omega')]=\frac{\hbar}{iZ_0\pi}\big\{\mathrm{Im}[\boldsymbol{\kappa}^{\mathrm{T}}(\omega)\star\boldsymbol{\mu}^{-1}(\omega)]\big\}(\mathbf{r},\mathbf{r}')\delta(\omega-\omega'),\tag{5.6b}$$

$$[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{P}}_{\mathrm{N}}^{\dagger}(\mathbf{r}',\omega')]=-\frac{\hbar}{iZ_0\pi}\big\{\mathrm{Im}[\boldsymbol{\mu}^{-1}(\omega)\star\boldsymbol{\kappa}(\omega)]\big\}(\mathbf{r},\mathbf{r}')\delta(\omega-\omega'),\tag{5.6c}$$

$$[\hat{\mathbf{M}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{\mathbf{M}}_{\mathrm{N}}^{\dagger}(\mathbf{r}',\omega')]=-\frac{\hbar}{\mu_0\pi}\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r},\mathbf{r}',\omega)]\delta(\omega-\omega'),\tag{5.6d}$$

which are the commutation relations for the noise polarisation and noise magnetisation fields in chiral media. The noise currents terms can be connected to electric and magnetic matter-field system excitations following the prescription discussed in section 4.1. The relationship between the noise current components and the system excitations is shown in Eq. (4.20), when applied to chiral media the tensor of response functions, $\mathcal{R}(\omega)$, is the (positive) square root of

$$\mathcal{R}\star\mathcal{R}^{\dagger}=\begin{pmatrix}\varepsilon_0\mathrm{Im}[\boldsymbol{\varepsilon}-\boldsymbol{\kappa}^{\mathrm{T}}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\kappa}] & \frac{\mathrm{Im}[\boldsymbol{\kappa}^{\mathrm{T}}\star\boldsymbol{\mu}^{-1}]}{iZ_0}\\ -\frac{\mathrm{Im}[\boldsymbol{\mu}^{-1}\star\boldsymbol{\kappa}]}{iZ_0} & -\frac{\mathrm{Im}[\boldsymbol{\mu}^{-1}]}{\mu_0}\end{pmatrix},\tag{5.7}$$

and the system excitations must obey the commutation relation given in Eq. (4.22). In Eq. (2.31) the terms $\boldsymbol{G}_e(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{G}_m(\mathbf{r},\mathbf{r}',\omega)$ are introduced, defined for anisotropic media as Eqs. (2.32). For chiral media, these are now be defined as

$$\begin{pmatrix}\boldsymbol{G}_e(\mathbf{r},\mathbf{r}',\omega)\\ \boldsymbol{G}_m(\mathbf{r},\mathbf{r}',\omega)\end{pmatrix}=-i\mu_0\omega\sqrt{\frac{\hbar}{\pi}}\Big[\boldsymbol{G}(\omega)\star\begin{pmatrix}i\omega\\ \times\overleftarrow{\nabla}\end{pmatrix}\star\mathcal{R}(\omega)\Big](\mathbf{r},\mathbf{r}').\tag{5.8}$$

With the definitions Eq. (5.8), the integral relation can be written in the form shown in Eq. (2.34). The Hamiltonian of this system is given by Eq. (4.23), so the quantisation of the electromagnetic field

in chiral media has been completed.

5.2 Propagation in Chiral Media and Chiral Molecules

5.2.1 Propagation in Chiral Media

In terms of the vector wave functions, $\mathbf{M}$ and $\mathbf{N}$, introduced in chapter 2, circularly polarised light is defined as

$$\hat{\mathbf{e}}_L = \frac{1}{\sqrt{2}}\left(\hat{\mathbf{N}} - \hat{\mathbf{M}}\right), \quad \hat{\mathbf{e}}_R = \frac{1}{\sqrt{2}}\left(\hat{\mathbf{N}} + \hat{\mathbf{M}}\right), \tag{5.9}$$

where $\hat{\mathbf{e}}_L$ and $\hat{\mathbf{e}}_R$ are the (normalised) left and right handed circularly polarised wave vectors, respectively. By solving the Booker equation (Eq. (E.12) in Appendix E) it is found that there are two dispersion relations for wave propagation in chiral media, corresponding to a left handed and a right handed dispersion relation. When the media is isotropic, i.e.,

$$\boldsymbol{\varepsilon} = \varepsilon \boldsymbol{I}, \quad \boldsymbol{\mu} = \mu \boldsymbol{I}, \quad \boldsymbol{\kappa} = \kappa \boldsymbol{I}, \tag{5.10}$$

the dispersion relations are given as [93]

$$(k^L)^2 = \left(\frac{\omega}{c}\right)^2 (-\kappa + \sqrt{\varepsilon\mu})^2, \tag{5.11a}$$

$$(k^R)^2 = \left(\frac{\omega}{c}\right)^2 (\kappa + \sqrt{\varepsilon\mu})^2, \tag{5.11b}$$

where the superscripts refer to the left L and right R handed dispersion relations, respectively. This implies that left and right handed circularly polarised light have different refractive indices and therefore propagate at different speeds in chiral media [73]. It should be noted that linearly polarised plane waves can be decomposed into left and right handed circularly polarised light,

$$\hat{\mathbf{N}} = \frac{1}{2}\left(\hat{\mathbf{N}} + \hat{\mathbf{M}}\right) + \frac{1}{2}\left(\hat{\mathbf{N}} - \hat{\mathbf{M}}\right), \tag{5.12a}$$

$$\hat{\mathbf{M}} = \frac{1}{2}\left(\hat{\mathbf{N}} + \hat{\mathbf{M}}\right) - \frac{1}{2}\left(\hat{\mathbf{N}} - \hat{\mathbf{M}}\right). \tag{5.12b}$$

In isotropic media the left and right handed circularly polarised light are constrained by the same dispersion relation so they propagate at the same speed and therefore the left and right handed wave vectors do not become out of phase with each other. In chiral media, the different propagation speeds between the left and right handed wave vectors will result in a phase difference between the two.

The reflection and transmission properties of chiral media are different to the properties of anisotropic media. As an example, consider a linearly polarised wave, propagating in an achiral medium, incident on the boundary with a chiral medium. The transmitted wave will be comprised of a left and a right handed component, which will subsequently become out of phase as they propagate through the chiral medium. The reflected wave from the incident linear polarisation will be circularly polarised, even though the medium it is propagation through is achiral, the handedness of which will depend on the chiral identity of the chiral medium. This means that the reflected wave is comprised of a component that conserves the polarisation of the incident wave and a component of the alternate linear vector wave function. The chiral medium can be said to have rotated the polarisation of the wave. Therefore the standard Fresnel coefficients, shown in Eq. (2.56), are not sufficient to account for the reflection off chiral media as they fail to take into account the mixing of the wave vector functions in the reflected wave.

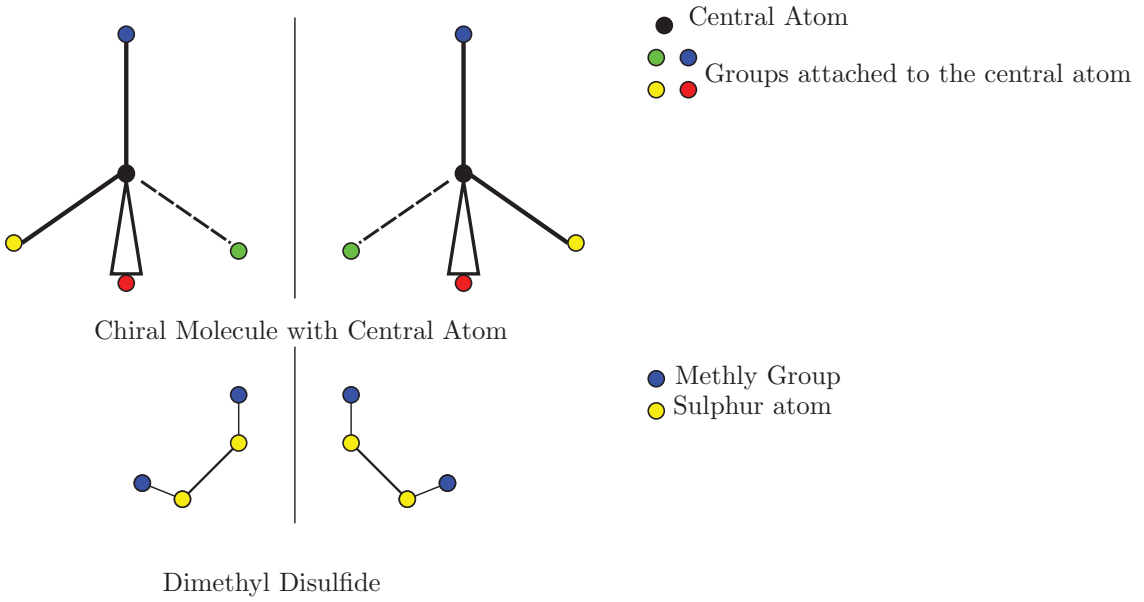


Figure 5.1: The top image shows the configuration of a chiral molecule with one chiral centre, which is typically a carbon atom. The coloured circles represent atomic or molecular groups that all need to be different for the molecule to exhibit chiral behaviour. The dashed line implies a bond directed into the page and the triangle represents an atomic or molecular group coming out of the plane of the page. The second image is a pictorial representation of Dimethly Disulfide, the molecule used as an example later in this chapter. In both structures of chiral molecule it can be seen that no combination of translations or rotations can transform a molecule into its mirror image.

5.2.2 Chiral Molecules

Naturally occuring chiral molecules, such as amino acids and sugars, tend to have a carbon atom as a centre, called a chiral centre. The relative ordering of a molecule's functional groups, the separate collection of atoms attached to the chiral centre, distinguishes the molecule from its enantiomer and as such is important in chiral molecules. In a molecule with 4 functional groups bonded to a carbon atom, the groups must all be different in a chiral molecule. There exist chiral molecules that are not centred on a carbon atom, where the chiral behaviour is induced by the geometry of the molecule. The molecules need to be three dimensional objects to exhibit chiral behaviour. Pictorial examples of these two types of chiral molecule are shown in Fig. 5.1, where the second diagram is a representation of the molecule that will be used as a test molecule in this chapter.

5.3 Chiral Casimir-Polder Potential

In section 2.2 the Casimir-Polder potential was derived from an interaction Hamiltonian in the multipolar coupling regime, reproduced here for convenience

$$\hat{H}_{AF} = -\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A) - \hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A). \tag{5.13}$$

This is substituted into the 2nd order energy perturbation equation, Eq. (2.45), to obtain the expression Eq. (2.47). In chapter 2, terms that included both the electric and magnetic atomic (or molecular) dipoles were excluded on the grounds of parity. However, when considering chiral molecules and chiral media, the cross terms cannot be neglected as they are responsible for the chiral interaction, therefore

$$\langle N|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|N\rangle,\ \langle N|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|N\rangle \neq 0. \tag{5.14}$$

The derivation of the electric and magnetic components of the Casimir-Polder potential in section 2.2 is not altered by the inclusion of the cross terms shown in Eq. (5.14), here these contributions will be neglected to focus on the derivation of the chiral component. Altering Eq. (2.47) to include only the chiral terms and substituting in Eq. (2.49) leads to an energy shift due to the chiral interaction of

$$\begin{aligned}\Delta E_c = \sum_\lambda \sum_k \int \mathrm{d}^3 r \int_0^\infty \mathrm{d}\omega \Big(&\langle N|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|N\rangle \\ &+ \langle N|\hat{\mathbf{m}}\cdot\hat{\mathbf{B}}(\mathbf{r}_A)|I\rangle\langle I|\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}(\mathbf{r}_A)|N\rangle \Big) \Big/ \Big(E_N - E_I \Big).\end{aligned} \tag{5.15}$$

By substituting Eqs. (2.48a) and (2.48b) into Eq. (5.15) and making use of Eq. (2.34) the energy perturbation due to the chiral component of the Casimir-Polder potential is found to be

$$\begin{aligned}\Delta E_c = &\frac{\mu_0}{\pi} \sum_k \mathcal{P} \int_0^\infty \frac{i\ \mathrm{d}\omega\ \omega}{\omega_{kn}+\omega} \Big[\mathbf{d}_{nk}\cdot \mathrm{Im}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega)] \times \overleftarrow{\nabla}' \cdot \mathbf{m}_{kn} \\ &+ \mathbf{m}_{nk}\cdot\nabla\times \mathrm{Im}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega)]\cdot\mathbf{d}_{kn} \Big].\end{aligned} \tag{5.16}$$

It can be seen that Eq. (5.16) is zero when the particle is achiral as there is either an electric dipole transition or a magnetic dipole transition, but not both. The non-analytic imaginary part of the Green's function can be written in terms of the analytic Green's function and its complex conjugate ($\mathrm{Im}[\boldsymbol{G}] = \frac{1}{2i}(\boldsymbol{G} - \boldsymbol{G}^\dagger)$). Following the procedure explained in section 2.2, a contour integral is performed which results in the potential separating into an off-resonant contribution, dependent upon complex frequency, and a resonant contribution. As the bulk part of the Green's function does not contribute to the Casimir-Polder potential, only the scattering part of the Green's function is included. This leads to a Casimir-Polder potential for a chiral molecule initially in state n of

$$\begin{aligned}U_c^n = &-\frac{\mu_0\hbar}{2\pi}\int_0^\infty \mathrm{d}\xi\ \xi \left(\mathrm{tr}[\boldsymbol{\Gamma}^n_{em}(i\xi)\cdot\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)\times\overleftarrow{\nabla}'] + \mathrm{tr}[\boldsymbol{\Gamma}^n_{me}(i\xi)\cdot\nabla\times\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)] \right) \\ &+ i\mu_0 \sum_k \Theta(\omega_{nk})\,\omega_{nk} \Big(\mathbf{d}_{nk}\cdot\mathrm{Re}[\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\times\nabla'\cdot\mathbf{m}_{kn} \\ &+ \mathbf{m}_{nk}\cdot\nabla\times\mathrm{Re}[\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\cdot\mathbf{d}_{kn} \Big),\end{aligned} \tag{5.17}$$

where the chiral susceptibility tensors for a particle initially in state n, $\boldsymbol{\Gamma}^n_{em}(i\xi)$ and $\boldsymbol{\Gamma}^n_{me}(i\xi)$, are defined as

$$\boldsymbol{\Gamma}^n_{em}(i\xi) = \frac{1}{\hbar}\sum_k \left(\frac{\mathbf{m}_{kn}\otimes\mathbf{d}_{nk}}{\omega_{kn}+i\xi} - \frac{\mathbf{m}_{kn}\otimes\mathbf{d}_{nk}}{\omega_{kn}-i\xi} \right), \tag{5.18a}$$

$$\boldsymbol{\Gamma}^n_{me}(i\xi) = \frac{1}{\hbar}\sum_k \left(\frac{\mathbf{d}_{kn}\otimes\mathbf{m}_{nk}}{\omega_{kn}+i\xi} - \frac{\mathbf{d}_{kn}\otimes\mathbf{m}_{nk}}{\omega_{kn}-i\xi} \right). \tag{5.18b}$$

The system under investigation is chiral, therefore the Green's function is reciprocal, this means that

$$\nabla\times\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)] = -\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)\times\overleftarrow{\nabla}'. \tag{5.19}$$

After defining $\boldsymbol{\Gamma}(i\xi)$ such that

$$\boldsymbol{\Gamma}(i\xi) = -\boldsymbol{\Gamma}_{em}(i\xi) + \boldsymbol{\Gamma}_{me}(i\xi), \tag{5.20}$$

and defining a notational shorthand $\boldsymbol{R}$,

$$\boldsymbol{R} = -\mathbf{m}_{nk}\otimes\mathbf{d}_{nk} + \mathbf{d}_{kn}\otimes\mathbf{m}_{nk}, \tag{5.21}$$

the chiral potential can be written as

$$\begin{aligned} U_c^n = &-\frac{\mu_0\hbar}{2\pi}\int_0^\infty \mathrm{d}\xi\ \xi\ \mathrm{tr}[\boldsymbol{\Gamma}^n(i\xi)\cdot\nabla\times\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)] \\ &+ i\mu_0\sum_k \Theta(\omega_{nk})\omega_{nk}\mathrm{tr}\big[\boldsymbol{R}\cdot\mathrm{Re}[\nabla\times\mathrm{Re}[\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\big]. \end{aligned} \tag{5.22}$$

If the molecule is isotropic, the Casimir-Polder potential shown in Eq. (5.22) can be simplified. The dipole matrix elements can be averaged to

$$\mathbf{d}\otimes\mathbf{m} = \frac{\mathbf{d}\cdot\mathbf{m}}{3}\boldsymbol{I}. \tag{5.23}$$

The $\boldsymbol{\Gamma}^n(i\xi)$ term reduces to

$$\boldsymbol{\Gamma}^n(i\xi) = -\frac{2}{3\hbar}\sum_k \frac{\xi\ R_{nk}}{(\omega_{kn})^2+\xi^2}\boldsymbol{I}, \tag{5.24}$$

and $\boldsymbol{R}$ to

$$\boldsymbol{R} = -\frac{2i}{3}R_{nk}\boldsymbol{I}. \tag{5.25}$$

The R_{nk} expression that has been introduced is the optical rotatory strength and it is defined as

$$R_{nk} = \mathrm{Im}(\mathbf{d}_{nk}\cdot\mathbf{m}_{kn}). \tag{5.26}$$

This term captures the interaction between the electric and magnetic dipole moments in a chiral molecule. Crucially, left and right handed enantiomers differ in the sign of R_{nk} for a particular transition. As was discussed in section 2.2, if the particle is achiral there are no intermediate states that can be reached by both an electric dipole transition and a magnetic dipole transition and therefore the optical rotatory strength will be zero.

The chiral component of the Casimir-Polder potential for an isotropic molecule initially in state n is now given as

$$\begin{aligned} U_c^n(\mathbf{r}_A) = &-\frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\xi\ \xi\ \Gamma^n(i\xi)\mathrm{tr}[\nabla\times\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)] \\ &+ \frac{2\mu_0}{3}\sum_k \Theta(\omega_{nk})\omega_{nk}R_{nk}\mathrm{tr}[\nabla\times\mathrm{Re}[\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]]. \end{aligned} \tag{5.27}$$

The structure of the chiral component possesses a similarity with the electric and magentic components in that the potential has a contribution that describes the particles behaviour and a component that describes the mediums response. In the chiral component, it can be seen that if

$$\mathrm{tr}[\nabla\times\boldsymbol{G}^{(1)}(\mathbf{r}_A,\mathbf{r}_A,i\xi)] = 0, \tag{5.28}$$

or

$$R_{nk} = 0, \tag{5.29}$$

then the chiral component of the Casimir-Polder potential vanishes. As R_{nk} only disappears for achiral particles, this means that the chiral component of the Casimir-Polder potential only acts on chiral molecules. The construction of the scattering part of the Green's function, see appendix E, means that Eq. (5.28) does not hold if the medium has magnetoelectric terms, such as the chiral susceptibility. Therefore, the chiral component of the Casimir-Polder potential is only non-zero when both the particle and body are chiral. This can be thought of as a generalisation to the Curie dissymmetry principle, originally formulated for crystal symmetries, which states that in certain interactions between objects

they are required to possess similar properites. In the case above this means that a medium cannot distinguish between enantiomers if it is not chiral itself, and the chiral identity of the medium cannot contribute unless it is interacting with another chiral object.

A few comments are necessary on the validity of the assumption that a chiral molecule can be viewed as isotropic. In a chiral molecule, by definition, the electric and magnetic dipoles can be orientated at any angle (except perpendicular) with respect to each other. Therefore it may seem that an isotropic chiral molecule is impossible. However, the Casimir-Polder interaction time is much faster than the molecules own internal oscillations. This difference in interaction time results in an averaging over the particles orientations and justifies the assumption of an isotropic chiral molecule.

5.4 Chiral Particle near a Chiral Halfspace

As an example of the chiral Casimir-Polder potential, an isotropic molecule in free space near the boundary with an isotropic chiral medium is considered, depicted in Fig. 5.2. To calculate the Casimir-Polder potential in this geometry, the scattering part of the Green's function is required. The form given in Eq. (2.55) cannot be used as it does not take into account the mixing of the polarisation vectors that occurs when linearly polarised light reflects of a chiral medium, see section 5.2.1. Therefore a scattering Green's function of the form

$$\begin{aligned}\boldsymbol{G}^{(1)}(\mathbf{r},\mathbf{r}',\omega) = \frac{i}{8\pi^2}\int \frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\big[&R_{MM}\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}) + R_{MN}\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})\\ &+ R_{NM}\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}) + R_{NN}\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})\Big],\end{aligned} \tag{5.30}$$

is proposed, see appendix E and Ref. [93]. The R_{MN} and R_{NM} reflection coefficients describe the reflection of an incident $\mathbf{M}$ ($\mathbf{N}$) polarised wave to an $\mathbf{N}$ ($\mathbf{M}$) polarised wave. By changing the integration variable to $\tilde{k}_{z1}$ in the off-resonant contribution and performing the curl and trace operations (see section E.2) the chiral potential for this geometry is

$$\begin{aligned}U_c^n(\mathbf{r}_A) = &-\frac{\hbar\mu_0}{4\pi^2 c}\int_0^\infty \mathrm{d}\xi\ \xi^2\ \Gamma^n(i\xi)\int_{\frac{\xi}{c}}^\infty \mathrm{d}\tilde{k}_{z1}\left[R_{MN}\left(1-\frac{2\tilde{k}_{z1}^2c^2}{\xi^2}\right)+R_{NM}\right]e^{-2\tilde{k}_{z1}z_A}\\ &+\frac{\mu_0}{6\pi c}\sum_k \Theta(\omega_{nk})\omega_{nk}^2 R_{nk}\mathrm{Im}\left[\int_0^\infty \mathrm{d}k_s\left[R_{MN}\left(1-\frac{2k_{z1}^2c^2}{\omega_{nk}^2}\right)+R_{NM}\right]e^{i2k_{z1}z_A}\right].\end{aligned} \tag{5.31}$$

The potential shown in Eq. (5.31) requires the cross reflection coefficients, R_{MN} and R_{NM}. In order to compare with the other components of the Casimir-Polder potential, Eqs. (2.53a) and (2.53b), the reflection coefficients R_{MM} and R_{NN} are required, although they will not be of the forms in Eqs. (2.56) due to the inclusion of the magnetoelectric term.

5.4.1 Perfect Chiral Mirror

As an example of the chiral Casimir-Polder potential, the interation between a chiral molecule and a 'perfect chiral mirror' is considered. The reflection coefficients are

$$R_{MN}, R_{NM} = \pm 1 \tag{5.32}$$

and therefore,

$$R_{MM}, R_{NN} = 0. \tag{5.33}$$

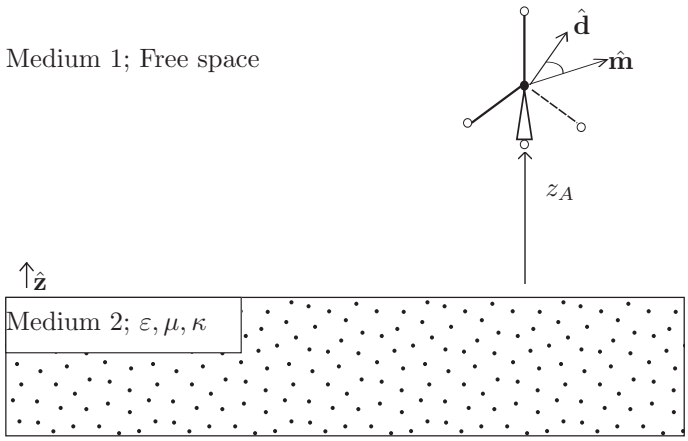


Figure 5.2: This diagram shows the Casimir-Polder force between a chiral particle in free space near the boundary with a chiral medium. The chiral molecule will have a different group, denoted by the circles, surrounding the chiral centre. The electric and magnetic dipole moments of the molecule are not perpendicular in a chiral molecule. The boundary is the $(x-y)$-plane at $z=0$ and free space (region 1) fills $z>0$, whereas the chiral medium (region 2) fills $z<0$.

When the rotation of the polarisation is clockwise, with regard to the incoming wave, the medium is called 'right handed' and the reflection coefficients are

$$R_{MN} = 1,\, R_{NM} = -1. \tag{5.34}$$

For a anticlockwise polarisation rotation, the medium is 'left handed' and the reflection coefficients are

$$R_{MN} = -1,\, R_{NM} = 1. \tag{5.35}$$

This perfect chiral mirror example is taken from Ref. [92]. For simplicity, the particle under consideration is assumed to be isotropic and initially in its ground state. Applying Eq. (5.32) to Eq. (5.31) gives a Casimir-Polder potential of

$$U^0(z_A) = \pm\frac{\hbar Z_0}{8\pi^2 z_A^3}\int_0^\infty \mathrm{d}\xi\; \Gamma^0(i\xi) e^{-2\frac{\xi}{c}z_A}\left(\frac{2\xi z_A}{c}+1\right) \tag{5.36}$$

where the '+' (upper sign) refers to the right-handed medium and '−' (lower sign) refers to the left-handed medium. In the retarded limit this becomes

$$U^0(z_A) = \mp\frac{Z_0 c^2}{16\pi^2 z_A^5}\sum_k \frac{R_{0k}}{(\omega_{k0})^2}, \tag{5.37}$$

where the approximation $\Gamma(i\xi) \approx \Gamma'(0)\xi$ has been used. In the opposite, non-retarded limit, Eq. (5.36) is

$$U^0(z_A) = \pm\frac{Z_0}{12\pi^2 z_A^3}\sum_k R_{0k}\ln\left(\frac{\omega_{k0} z_A}{c}\right). \tag{5.38}$$

To obtain this, the relationship

$$\int_0^\infty \frac{\mathrm{d}x\, x}{x^2+a^2} e^{-2x}\big(1+2x\big) \approx \int_a^b \frac{\mathrm{d}x\, x}{a^2+x^2} \approx -\ln a \tag{5.39}$$

where $a \ll 1$, has been used. The value b is an arbitrary cut-off which does not contribute to the integral.

It can be seen that the sign of Eq. (5.38) will change when the particle body distance becomes

$z_A < \frac{c}{\omega_{k0}}$. Equations (5.37) and (5.38) show a Casimir-Polder potential that can be attractive or repulsive, depending on the medium and particle in question. The sign of the optical rotatory strength R_{nk} depends upon the enantiomer in question and the sign of the medium, explicitly seen as the $\pm$ in Eqs. (5.37) and (5.38), is dependent upon the chiral identity of the medium, i.e., left or right handed. A comparsion with the Casimir-Polder potentials that arise in the same geometric set up with the other two perfect mirrors can be made, where a perfectly conducting (infinitely permeable) plate attracts (repels) polarisable particles and repels (attracts) magnetisable particles [12].

The introduction to this chapter provided a practical motivation for chiral dispersion forces, the separation of enantiomers by a selectively discriminating force. When a chiral molecule is near a perfect chiral mirror, Eq. (5.33) implies that there are no electric and magnetic components of the Casimir-Polder potential. As the chiral component of the Casimir-Polder force between a molecule and a perfect chiral mirror is the only component of the Casimir-Polder force, which can be attractive or repulsive, a perfect chiral mirror can generate a dispersion force that can distinguish between, and ultimately separate, enantiomers. However, it is likely that Casimir-Polder potential that arises due to the perfect chiral mirror represent a theoretical upper bound for the chiral Casimir-Polder potential and is physically unrealisable. This is because it would require a medium that completely rotates the polarisation of the incident waves and perfectly reflects the waves. In section 5.2.1 the example of a linearly polarised wave incident on a chiral medium reflecting as a circularly polarised wave was given. However, in the perfect chiral mirror case the reflected wave would be a complete rotation of the incident waves polarisation to the other linear polarisation. This means that when the incident wave is linearly polarised the reflected wave would still be linearly polarised, not circularly polarised. Although an incident circularly polarised wave will still be reflected as a circularly polarised wave.

A further constraint is the strength of the chiral response with respect to the permittivity and permeability. In general the relationship,

$$\kappa^2 < \varepsilon\mu, \tag{5.40}$$

holds for a typical chiral medium. This would prevent the existence of a perfect chiral mirror where the chiral response needs to be strong. Although it has been shown recently that media with a strong chiral response [94, 95], i.e.,

$$\kappa^2 > \varepsilon\mu, \tag{5.41}$$

could exist, Eq. (5.40) is a bound for most chiral media. An implication of Eq. (5.41) is that one of the waves will be backwards travelling, as can be seen from Eqs. (5.11). The response functions in Eqs. (5.40) and (5.41) must still obey the Kramers - Kronig relations and the dissipative parts must be constrained by the relation [19, 96, 97]

$$|\mathrm{Im}[\boldsymbol{\kappa}]| < \sqrt{\mathrm{Im}[\boldsymbol{\varepsilon}]\mathrm{Im}[\boldsymbol{\mu}]}. \tag{5.42}$$

It is important to evaluate the relative strengths of the different components of the Casimir-Polder potential. As has been noted with a perfect chiral mirror in a simple planar geometry, the reflection coefficients R_{MM} and R_{NN} are necessarily zero. This means that in the planar set up with a perfect chiral mirror, the electric and magnetic components of the Casimir-Polder potential will not exist. However, a natural reference point is the comparison with the perfect mirror, which is either perfectly conducting (perfect dielectric mirror), with reflection coefficients of

$$R_{MM} = -R_{NN} = -1, \tag{5.43}$$

ω_{nk} (s^{-1})	$\lvert\mathbf{d}_{nk}\rvert^2$ $((\mathrm{C\ m})^2)$	R_{nk} $(\mathrm{C^2\ m^3\ s^{-1}})$
9.17x10^{15}	8.264x10^{-60}	3.328x10^{-64}

Table 5.1: This table shows the values of the parameters for a chiral molecule, Dimethyl Disulfide, taken from Ref. [78]. The planes are orientated at 90° to each other.

or infinitely permeable, where the reflection coefficients are

$$R_{MM} = -R_{NN} = 1. \tag{5.44}$$

The electric component Casimir-Polder potential for a perfect dielectric mirror in the non-retarded limit is derived in Ref. [12] and given here for reference,

$$U_e^0(z_A) = -\sum_k \frac{|\mathbf{d}_{0k}|^2}{48\pi\varepsilon_0(z_A)^3}. \tag{5.45}$$

It can be seen that there are different spatial scaling laws for the Casimir-Polder potential between a molecule and a perfect chiral mirror, Eq. (5.38), and a perfect dielectric mirror, Eq. (5.45). In the geometry with the perfect chiral mirror, the scaling law includes an additional logarithmic dependence to the distance from the boundary and the molecule.

In order to compare the Casimir-Polder potential with different media, a chiral molecule is needed. In this chapter, Dimethyl Disulfide $(CH_3)_2S_2$ is to be used, where the dipole and rotatory strengths for each transition have been numerically calculated for various orientations [78]. A single transition is to be considered and the orientation of the two $CH_3 - S - S$ planes is 90°, as shown in Fig. 5.1. The values obtained from Ref. [78] are shown in table 5.1.

A plot of the Casimir-Polder potentials from a molecule near a perfect dielectric mirror and a molecule near a perfect chiral mirror is shown in Fig. 5.3. The same molecule, Dimethly Disulfide, has been used to calculate both potentials. It should be noted that both potentials have been scaled by different values in order to compare the structures. The Casimir-Polder potential between the molecule and the perfect dielectric mirror is 12 orders of magnitude greater than the Casimir-Polder potential between the same molecule and a perfect chiral mirror. An important difference between the two examples is the logarithmic term in the perfect chiral mirror geometry, as shown in Eq. (5.38). However, this does not explain the large discrepancy between the two potentials. The difference arises partly due to the order of magnitude difference in the electric and magnetic dipole moments of the molecule and the physical constants that scale the potentials. The electric dipole moments of the molecule are an order of perturbation larger than the magnetic dipole moments and so $R_{0k} \ll |\mathbf{d}_{0k}|^2$ will always hold.

5.4.2 Chiral Media

To describe the reflection of waves incident on a real medium, the reflection coefficients cannot be unity because the permittivity and permeability are finite valued. For the current geometry, a particle in free space near a planar halfspace, the necessary reflection coefficients are obtained from Ref. [93] and are as follows;

$$R_{MM} = \frac{2k_2^L k_2^R k_{z1}^2 - 2k_1^2 k_{z2}^L k_{z2}^R - \left(\frac{\varepsilon_2-\mu_2}{\sqrt{\varepsilon_2\mu_2}}\right) k_{z1}k_1(k_{z2}^L k_2^R + k_{z2}^R k_2^L)}{D}, \tag{5.46a}$$

$$R_{NN} = \frac{2k_2^L k_2^R k_{z1}^2 - 2k_1^2 k_{z2}^L k_{z2}^R - \left(\frac{\mu_2-\varepsilon_2}{\sqrt{\varepsilon_2\mu_2}}\right) k_{z1}k_1(k_{z2}^L k_2^R + k_{z2}^R k_2^L)}{D}, \tag{5.46b}$$

$$R_{MN} = -R_{NM} = \frac{2ik_1(k_{z2}^L k_2^R - k_{z2}^R k_2^L)}{D}, \tag{5.46c}$$

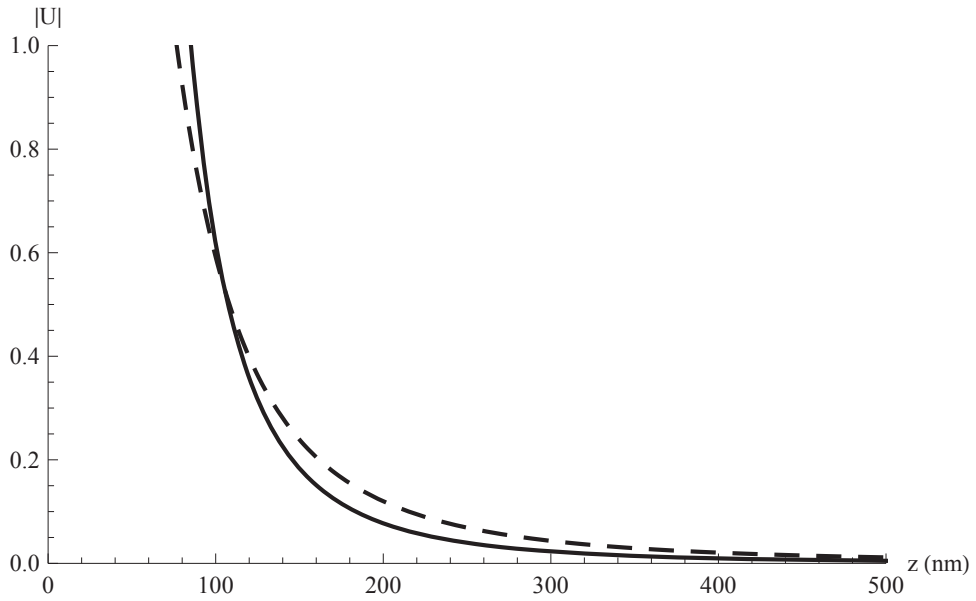


Figure 5.3: This graph shows the Casimir-Polder potential for the perfect dielectric mirror (black line) and the perfect chiral mirror (dashed line), in both cases the molecule near the halfspace is Dimethyl Disulfide. The two potentials have been scaled by different factors so they fit on the same graph. It should be noted that the perfect dielectric mirror potential is larger than the potential between a perfect chiral mirror and the same molecule, by 12 orders of magnitude. The graph highlights that the potential do not have the same structure due to the natural logarithm term in the perfect chiral mirror expression. This provides a slight correction to the potential in the region shown in the graph.

$$D = 2k_2^L k_2^R k_{z1}^2 + 2k_1^2 k_{z2}^L k_{z2}^R + \left(\frac{\varepsilon_2+\mu_2}{\sqrt{\varepsilon_2\mu_2}}\right) k_{z1}k_1(k_{z2}^L k_2^R + k_{z2}^R k_2^L). \tag{5.46d}$$

where k_i and k_{zi} refer to the dispersion relation in medium i and z-component of the dispersion relation in medium i, respectively. The reflection coefficients can be simplified by substituting the definitions for the chiral dispersion relations, shown in Eqs. (5.11), and applying the non-retarded limit ($\tilde{k}_{z1} \approx \tilde{k}_{z2}^L \approx \tilde{k}_{z2}^R$),

$$R_{MM} = \frac{\varepsilon_2\mu_2 - \kappa_2^2 - \varepsilon_2 + \mu_2 - 1}{\varepsilon_2\mu_2 - \kappa_2^2 + \varepsilon_2 + \mu_2 + 1}, \tag{5.47a}$$

$$R_{NN} = \frac{\varepsilon_2\mu_2 - \kappa_2^2 + \varepsilon_2 - \mu_2 - 1}{\varepsilon_2\mu_2 - \kappa_2^2 + \varepsilon_2 + \mu_2 + 1}, \tag{5.47b}$$

$$R_{MN} = -R_{NM} = \frac{2i\kappa_2}{\varepsilon_2\mu_2 - \kappa_2^2 + \varepsilon_2 + \mu_2 + 1}. \tag{5.47c}$$

The response functions are dependent on complex frequency, $\varepsilon_2 = \varepsilon_2(i\xi)$, $\mu_2 = \mu_2(i\xi)$ and $\kappa_2 = \kappa_2(i\xi)$. It can be seen that when $\kappa = 0$, the R_{MM} and R_{NN} reflection coefficients will revert to the standard Fresnel coefficents and the cross reflection coefficients will disappear, i.e., $R_{MN} = R_{NM} = 0$. The chiral component of the Casimir-Polder potential in the non-retarded limit for a realistic medium can be found by substituting the reflection coefficients, Eqs. (5.47), into the full chiral Casimir-Polder potential, shown in Eq. (5.31). The $\tilde{k}_{z1}$ integral is performed in the non-retarded limit and this leads to

$$\begin{aligned} U_c^n(z_A) =& \frac{\hbar Z_0}{4\pi^2 (z_A)^3} \int_0^\infty \mathrm{d}\xi\ \Gamma^n(i\xi) \frac{i\kappa_2}{\varepsilon_2\mu_2 - \kappa_2^2 + \varepsilon_2 + \mu_2 + 1} \\ &+ \frac{Z_0}{6\pi (z_A)^3} \sum_k \Theta(\omega_{nk}) R_{nk} \mathrm{Im}\left[\frac{i\kappa_2}{\varepsilon_2\mu_2 - \kappa_2^2 + \varepsilon_2 + \mu_2 + 1}\right]. \end{aligned} \tag{5.48}$$

	$\varepsilon(\omega)$	$\mu(\omega)$	$\kappa(\omega)$
Oscillator Strengths (ω_p, ω_m, a)	5.47x10^{14}	3.06x10^{14}	$-$3.61x10^{14}
Resonance (ω_E, ω_B, ω_C)	4.96x10^{14}	4.96x10^{14}	4.96x10^{14}
Damping (γ_E, γ_B, γ_C)	2.51x10^{13}	2.51x10^{13}	$-$2.58x10^{13}

Table 5.2: This table shows the values of the parameters for a chiral metamaterial, taken from Ref. [86]. The medium is constructed in a woodpile fashion by laser writing and silver evaporation. The first and second columns contains the values for the permittivity and permeability respectively, with the assumption that these are described by the Drude Lorentz model. The third column lists the values for the chiral response of the metamaterial under the assumption that this is modelled by the Condon model.

The spatial scaling law for both the resonant and off-resonant contributions to the chiral Casimir-Polder potential is $1/(z_A)^3$. This can be contrasted with the spatial scaling from the scenario with a perfect chiral mirror, $1/(z_A)^3 \ln(\omega_{k0} z_A/c)$. As the distance between the molecule and the medium decreases, the potential that arises from the perfect chiral mirror will increase by a greater amount than the case from more realistic media.

In a causal medium the permittivity and permeability response functions can both be described by a Drude-Lorentz model

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 - \omega_E^2 + i\gamma_E\omega}, \tag{5.49a}$$

$$\mu(\omega) = 1 - \frac{\omega_m^2}{\omega^2 - \omega_B^2 + i\gamma_B\omega}, \tag{5.49b}$$

respectively and the chiral susceptibility can be described by the Condon model [73, 98]

$$\kappa(\omega) = \frac{a\omega}{\omega^2 - \omega_C^2 + i\gamma_C\omega}. \tag{5.50}$$

The response function models have similar features and their constants can be grouped. The constants ω_p, ω_m and a represent the oscillator strengths for the various dipole transitions in the medium and they correspond to the dielectric, magnetic and rotatory strengths of the medium, respectively. The remaining constants are the resonant frequencies and damping factors for the permittivity (ω_E, γ_E), permeability (ω_B, γ_B) and the chirality (ω_C, γ_C) of the medium. It has been assumed that the medium has a single transition and therefore a single resonant term.

To model the chiral component of the Casimir-Polder potential with a more realistic medium, a chiral metamaterial with a woodpile structure is chosen, it's properties are given in Ref. [86] and are shown in table 5.2. Using the values in table 5.2 a comparison between the chiral Casimir-Polder potential between a molecule and a chiral metamaterial and between a molecule and the perfect chiral mirror can be made, this is shown in Fig. 5.4, where the molecule is initially in its ground state. As can be seen, a molecule near a perfect chiral mirror induces a stronger Casimir-Polder potential than a molecule near a chiral metamaterial. This is due to the extra logarithmic term in the perfect chiral mirror case and the greater value of the reflection coefficients in the perfect chiral mirror geometry.

The only component of the Casimir-Polder potential that can distinguish between enantiomers is the chiral component, which in a planar geometry with dispersive media will not be the only contribution of the Casimir-Polder potential acting on a molecule. Therefore it is important to know the electric component as well as the chiral component of the Casimir-Polder potential.

The electric component of the Casimir-Polder potential between a molecule and a chiral medium can be found by substituting the adapted reflection coefficients (Eqs. (5.47a) and (5.47b)) into Eq. (5.31),

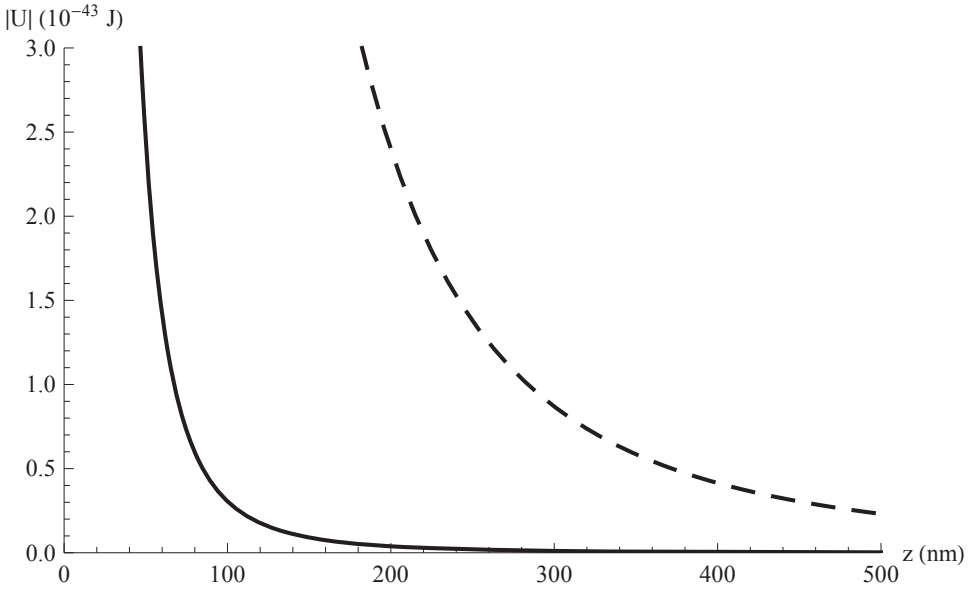


Figure 5.4: This graph shows the Casimir-Polder potential between Dimethyl Disulfide and a chiral metamaterial (black line), parameters from Ref. [86], and the Casimir-Polder potential between Dimethyl Disulfide and a perfect chiral mirror (dashed line). The potential between the molecule and the perfect chiral mirror is greater than the potential between the molecule and the chiral metamaterial. This is due to the contribution from the logarithmic term in addition to the greater value of the reflection coefficients in the perfect chiral mirror geometry.

which is then substituted into Eq. (2.53a) and taken to the non-retarded limit to obtain

$$\begin{aligned} U_e^n(z_A) = &- \frac{\hbar}{16\pi^2\varepsilon_0(z_A)^3}\int_0^\infty \mathrm{d}\xi\ \alpha^n(i\xi)\left[\frac{\varepsilon_2\mu_2-\kappa_2^2+\varepsilon_2-\mu_2-1}{\varepsilon_2\mu_2-\kappa_2^2+\varepsilon_2+\mu_2+1}\right] \\ &- \frac{1}{24\pi\varepsilon_0(z_A)^3}\sum_k \Theta(\omega_{nk})|d_{nk}|^2\mathrm{Re}\left[\frac{\varepsilon_2\mu_2-\kappa_2^2+\varepsilon_2-\mu_2-1}{\varepsilon_2\mu_2-\kappa_2^2+\varepsilon_2+\mu_2+1}\right]. \end{aligned} \tag{5.51}$$

The comparison between the electric and chiral potentials, when the molecule is initially in its ground state, is given in Fig. 5.5. It can be seen that both potentials have the same spatial scaling law, however, there is a large discrepancy in the size of the different components of the Casimir-Polder potential in this geometry. In Fig. 5.5 the potentials have been scaled by different factors, which vary by 13 orders of magnitude.

5.5 Chiral Cavity

5.5.1 Ground State Force

The Casimir-Polder potential between a chiral molecule and a chiral medium can be attractive or it can repel, depending upon the chiral identity of the molecule and the medium. This means that, for example, by replacing a molecule with it's enantiomer, the direction that the Casimir-Polder force acts will change and an attractive force will become a repelling force, and vice versa. Hence there is a method to distinguish between enantiomers, which can be placed in a geometry that enhances this ability to discriminate between enantiomers. A cavity is constructed between two chiral media of opposite chirality. This means that an enantiomer will be repelled by one side of the cavity and attracted by the other, as shown in Fig 5.6. In a racemic mixture the left handed enantiomers will be separated from the right handed enantiomers and the solution can be purified.

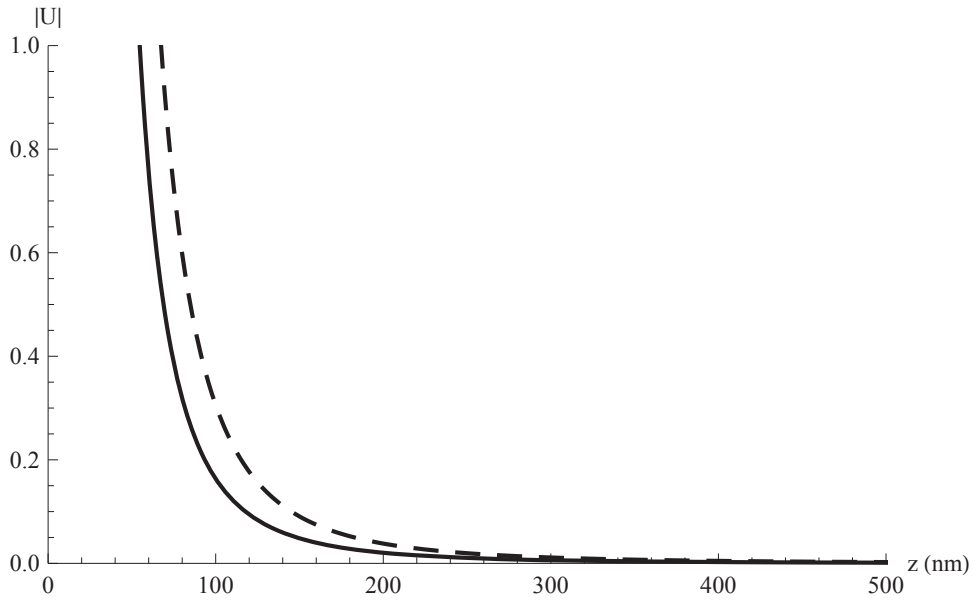


Figure 5.5: This graph shows the electric component of the Casimir-Polder potential (black line) and the chiral component of the Casimir-Polder potential (dashed line) between Dimethyl Disulfide and a chiral metamaterial with the parameters given in table 5.2. Each potential has been individually scaled to fit on the same graph, with the electric component 13 orders of magnitude larger than the chiral component. The structure of each potential is the same, with a $1/(z_A)^3$ spatial scaling. The disparity in the strength of the potential arises due to the difference in the electric and magnetic molecular dipole moments, the difference between the electric and chiral properties of the medium and the physical constants.

In this geometry, the complete scattering Green's function would take into consideration; multiple reflections in the cavity, backscattering off the far boundary (of the chiral plates), multiple backscattering events, etc. For this simulation only chiral halfspaces will be considered, this eliminates the scattering off the far boundaries and all subsequent contributions derived from this. To further simplify, the cavity is considered in the non-retarded limit. This means that the multiple reflections between the halfspaces are neglected and the total forces on the molecule from each halfspace are simply added. The forces acting on the molecule are calculated by the gradient of the potential, Eq. (2.43). The molecule within the cavity will be affected by the full Casimir-Polder force, this includes the electric, magnetic and chiral components. In this planar geometry the electric component of the Casimir-Polder force is attractive and tends to dominate over the magnetic component, which is not considered here. The electric component of the Casimir-Polder force provides a major contribution to the total force. The properties of the two halfspaces that border the cavity are identical except for their chirality, characterised by a in Eq. (5.50), which is negative for one of the halfspaces and positive for the other. As with the examples in the previous section the molecule, Dimethyl Disulfide, is assumed to be two level and therefore has just one transition.

The separation between the halfspaces is 100nm and the results are shown in Fig. 5.7 for the electric component of the force and Fig. 5.8 for the chiral component of the Casimir-Polder force. The electric component of the Casimir-Polder force, Fig. 5.7, is always attractive towards both halfspaces. In this scenario the components of the force that derives from each halfspace are equal and opposite. This means that in the electric component of the total Casimir-Polder force on the molecule, the halfspace closest to the molecule will provide the dominant contribution and the molecule will be preferentially attracted to this halfspace. An implication of this is that when the molecule is in the centre (i.e. equal distance from each halfspace), the electric components of the Casimir-Polder force from each halfspace cancel and the net contribution to the total Casimir-Polder force is zero. Although the chirality of the halfspaces

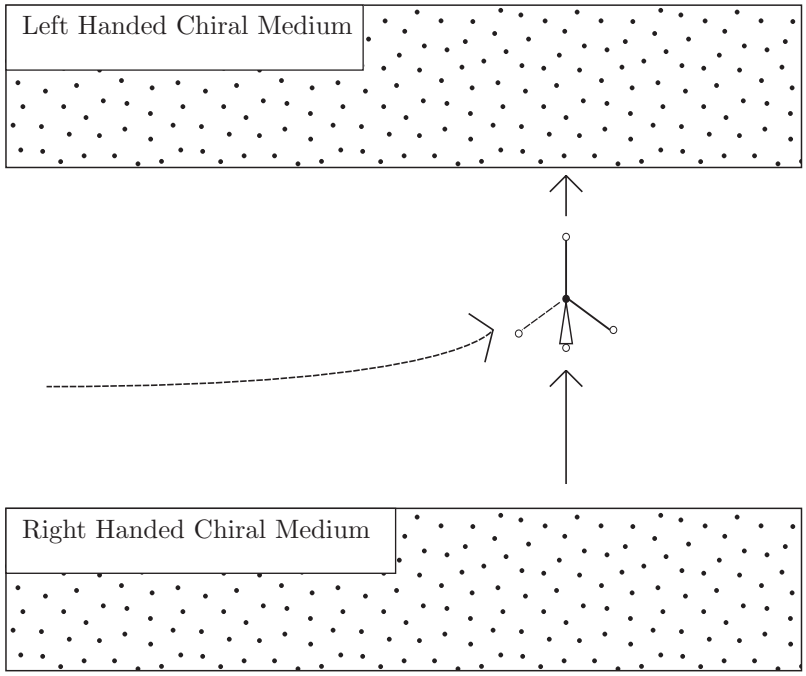


Figure 5.6: This diagram shows the Casimir-Polder force between a chiral particle in a cavity of free space between two chiral media with opposite handedness. If only the chiral component of the Casimir-Polder force is considered, the chiral molecule will be attracted to one of the halfspaces, in this case the medium labelled as left handed, and will be repelled from the other chiral medium, labelled right handed. This means that enantiomers will be separated by this force. When the electric component of the Casimir-Polder force is considered, this dominates over the chiral component and when the molecule is initially in its ground state, there can be no enantiomer separation.

has introduced a correction to the electric component of the Casimir-Polder force, this correction is not dependent on the handedness of the halfspace. Therefore the difference in the chirality of the halfspaces does not effect direction of the electric force from either halfspace and hence the combined electric component of the total force.

The chiral component of the Casimir-Polder force, see Fig. 5.8, shows an attractive force between the chiral molecule and one of the halfspaces (to the left hand side in Fig. 5.8) and a repulsive force between the chiral molecule and the other halfspace (to the right hand side). Furthermore, as the chiral forces from each halfspace are equal and acting in the same direction, the total chiral component of the Casimir-Polder force does not disappear at the midpoint between the halfspaces.

In the comparsion between the electric and chiral components of the Casimir-Polder force, it can been seen in Figs. 5.7 and 5.8 that for most of the cavity the electric component is many orders of magnitude larger than the chiral component. This means that in any interaction the electric component will be the dominant contibution to the Casimir-Polder force and as such will, almost exclusively, dictate the molecules behaviour. The exception to this is the central region between the halfspaces where the overall electric component is reduced sufficiently to allow the chiral component to provide the greatest contribution to the force. The width of this central region can be found by calculating the distances either side of the centre of the cavity where the electric and chiral components of the Casimir-Polder force are equal.

However, due to the large difference in magnitude between the components of the Casimir-Polder force, the width of this central region is smaller than the size of the molecule. This means that for a particle initially in the ground state, the Casimir-Polder force in the geometry as shown in Fig. 5.6 would not be able to distinguish between enantiomers and subsequently separate them.

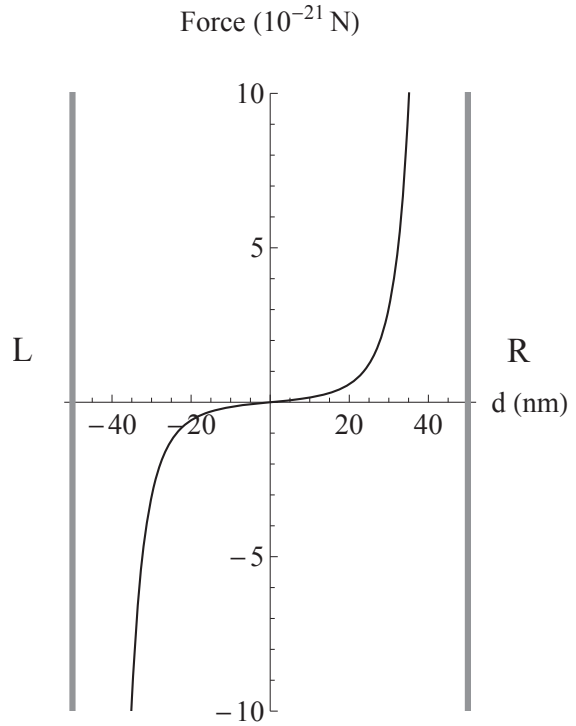


Figure 5.7: This graph shows the electric component of the Casimir-Polder force on a molecule in a cavity between two halfspaces of equal but opposite chirality. The molecule is attracted to both halfspaces with the stronger attraction coming from the halfspace the molecule is closest to. The vertical lines denote the boundaries of the metamaterials, the 100nm gap between is free space where the molecule is located. The parameters for the metamaterial were obtained from Ref. [86] as shown in table 5.2 and the molecular properties for Dimethly Disulfide are from Ref. [78].

The difference, by orders of magnitude, in the size of the force components arises from the disparity between the electric and chiral properties of both the molecule and the medium. By comparing the values of $|\mathbf{d}_{nk}|^2$ and R_{nk} in table 5.1, a difference of orders of magnitude can be found, $R_{nk}/c \lesssim 10^{-11}|\mathbf{d}_{nk}|^2$. This is because the electric dipole transition matrix elements are an order of perturbation larger than the magnetic dipole transition matrix elements [99],

$$\hat{\mathbf{d}} \gg \hat{\mathbf{m}}, \tag{5.52}$$

therefore the optical rotatory strength will be smaller than the electric dipole transition matrix elements squared. The chirality is only slightly smaller than the permittivity for the chiral metamaterial used in the above simulation, but this difference is enhanced by the structure of the reflection coefficients which favour the permittivity and thus produce a stronger component of the Casimir-Polder force.

5.5.2 Excited-State Force

If the initial state of the molecule is it's ground state, there is no resonant contribution to the electric or chiral components of the Casimir-Polder force. Sometimes these resonant parts of the force can act against the off-resonant contributions and thus suppress the overall force. In the same planar cavity geometry as discussed above, the off-resonant part of the electric component of the Casimir-Polder force is attractive whereas the direction of the chiral off-resonant component is dependent on the chiral identity of the molecule and medium in question.

Due to their versatility it can be theorised with a high degree of confidence that a metamaterial can

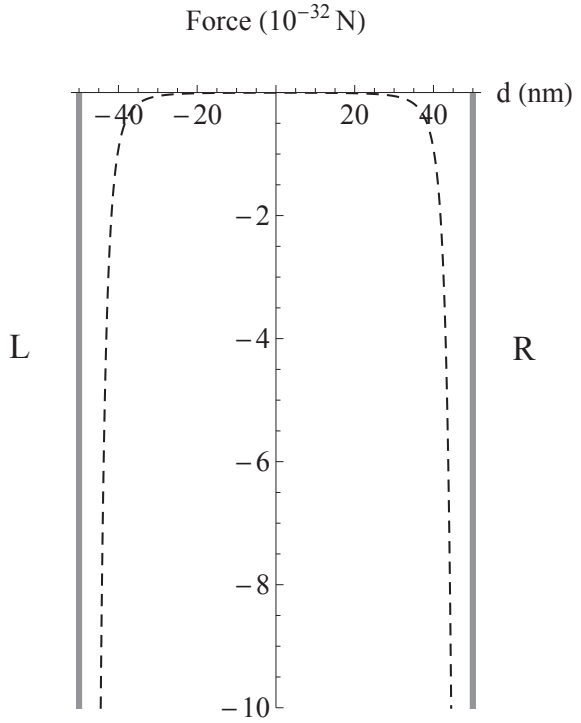


Figure 5.8: This graph shows the chiral component of the Casimir-Polder force on a molecule in a cavity between two halfspaces of equal but opposite chirality, with the same molecular and medium parameters as the electric component, shown in Fig. 5.7. The individual contributions to the force from each halfspace are an attractive force between the molecule and the left hand side halfspace and a repulsive force between the molecule and the right hand side halfspace. This means that the total force is always attractive to the left halfspace and the molecule is repelled from the right hand halfspace. This is in contrast to the electric component, which is an attractive force to both halfspaces with the larger contribution deriving from the closest halfspace.

be constructed such that for a given transition frequency of a chiral molecule, the resonant contribution to the electric component of the Casimir-Polder force will counteract the off-resonant contribution. By creating a cavity between two such metamaterials, with identical properties except for their chirality, a chiral molecule initially in an excited state can be separated from its enantiomer. This is due to the suppression of the electric component of the Casimir-Polder force which allows the chiral component of the force to attract an enantiomer to the corresponding halfspace while it is being repelled from the opposite halfspace.

As a proof of principle the theoretical metamaterial's parameters are chosen to be suitable multiples of the plasma frequency and the values for the optical rotatory strength and dipole strength are as given above for Dimethly Disulfide. The cavity between the metamaterials is 100nm of free space and the results for the electric and chiral components of the Casimir-Polder force are shown in Fig. 5.9. As can be seen, for a particular transition frequency there is a suppression of the electric component of the Casimir-Polder force that is sufficient to allow enantiomer separation. The chiral component is the largest contribution in the central region ($\approx$ 10nm) of the cavity (see the inset to Fig. 5.9 and a magnified version in Fig. 5.10), meaning that the direction of the force acting on a chiral molecule in this region will be dependent on its chirality. Therefore enantiomers that pass at low speeds through the centre of the cavity will be attracted or repelled in opposite directions and will be separated based on their chirality. This initial separation will then be amplified when the molecule is drawn out of the central region as the total electric component of the Casimir-Polder force is attractive and so the divergent paths of the enantiomers will continue to diverge. As an aside it is important to note that although the chirality of

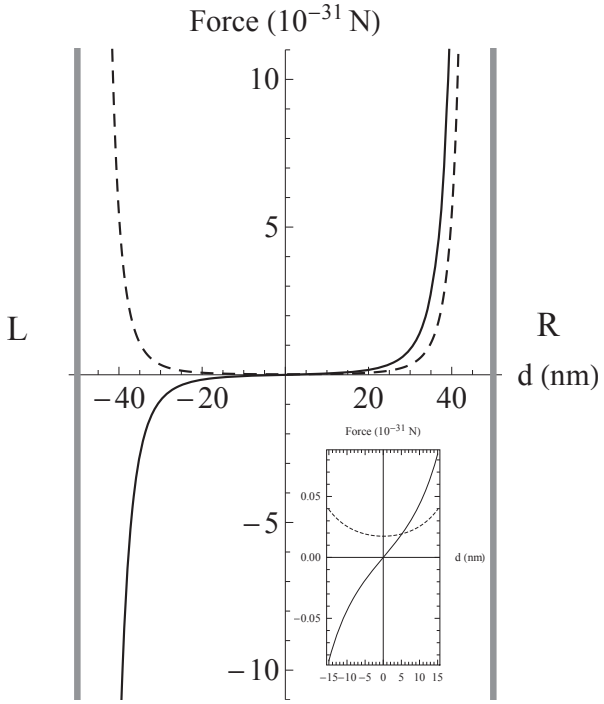


Figure 5.9: This graphs shows the total electric component of the Casimir-Polder force (black line) and the total chiral component (dashed line). The components of the force are of equal magnitude, with the chiral contribution larger in the centre and the electric contribution larger towards the halfspaces. The parameters are chosen as $\omega_E = \omega_B = \omega_C = \frac{\omega_p}{2}$, $\gamma_E = \gamma_B = -\gamma_C = \frac{\omega_p}{10^3}$, $a = -\frac{\omega_p}{3}$ and $\omega_m = \frac{\omega_p}{5}$. The inset in the bottom right shows a magnification of the centre region between -15 nm and 15 nm.

the metamaterials has not changed, the chiral component of the Casimir-Polder force is now acting in the opposite direction. This is due to the resonant part of the chiral force, which in this case is larger than the off-resonant part and acts against it. However, as this force still discriminates between the enantiomers, the necessary separation will still occur.

It should be noted that the above is just a theoretical proof of principle of the use of a dispersion force to distinguish between chiral enantiomers. Due to the large discrepancy in size between the electric and chiral components of the Casimir-Polder force, approximately 12 orders of magnitude, the resonant and off-resonant contributions to the electric component need to exactly match to a high level of accuracy to allow an adequate supression of the total electric contribution to the Casimir-Polder force. This is achieved by selecting a medium to construct the cavity which possesses a resonant frequency that allows the cancellation of the electric components of the Casimir-Polder force. If the resonant and off-resonant electric contributions are not of equal magnitude, the total electric force will still be larger than the total chiral force, therefore the electric component of the Casimir-Polder force will dictate the behaviour of the molecules and no chiral selection can occur. To increase the strength of the chiral component of the Casimir-Polder force, strongly chiral media could be used.

In this chapter the quantisation of the electromagnetic field in a chiral medium, based on the results for general bianisotropic media, has been presented. The chiral component of the Casimir-Polder potential has been derived through a perturbative approach, this was found to be dependent on the chiral identity of the molecule and the medium. As a proof of principle, the Casimir-Polder force in a chiral cavity geometry was shown to separate enantiomers under specific circumstances. The next chapter will discuss the quantisation of the electromagnetic field in a moving medium and will explore how motion affects the Casimir-Polder potential between an isotropic dielectric in motion and a stationary atom.

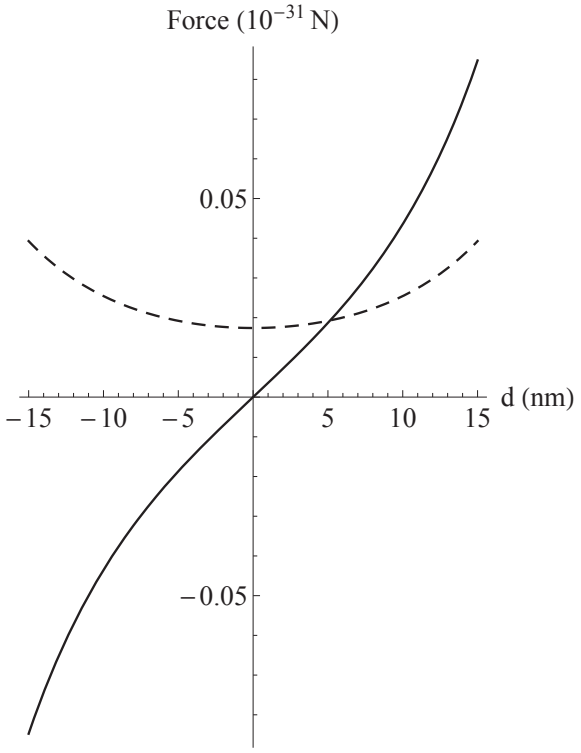


Figure 5.10: This graph shows a magnified version of the inset from Fig. 5.9. As can be seen, the chiral component of the Casimir-Polder force is greater than the electric component and thus the direction of the total force will depend on the relative chirality of the molecule and the halfspaces.

Chapter 6

Electromagnetic Field in Moving Media

It has been known that motion has an effect on physical quantities since the dawn of the scientific revolution, with notable work by Galileo and Newton. This resultant theory of the effect of motion introduced some important ideas, such as inertial reference frames. This concept can be illustrated by considering a 3 dimensional coordinate system in which an object is free to move around at a constant velocity. This coordinate system, or inertial reference frame, is labelled as frame 1 and is itself moving at a constant velocity with respect to a different reference frame, frame 2, which has its own coordinate system. As frame 1 moves with respect to frame 2, observers in each frame will not agree on the position of the object in frame 1 due to the relative motion of the frames. Each of these frames is an inertial reference frame, this means that there are no net forces that act on either of the frames therefore the laws of physics are the same in each frame. In fact, the laws of physics are the same in any inertial reference frame that can be defined. Although observers in each frame do not, in general, agree on positions, in Newtonian mechanics the observers will agree on the time duration of events. This is because time is considered as a universal property that is independent of the reference frames. Comparing the physical quantities as they appeared in each reference frame can be done by performing a Galilean transformation on the quantity. For position, this is of the form

$$\mathbf{r}' = \mathbf{r} - \mathbf{v}t. \tag{6.1}$$

However, it was found that this model of the effects of motion only applies when the speeds are slow compared to the speed of light, $v \ll c$, and so a theory that could account for greater speeds was needed. In 1905 this theory was published by A. Einstein and was called the theory of special relativity [100]. The key differences between the Einstein and Newton theories of motion are the presence of a universal speed limit and the removal of the concept of universal time in special relativity. Einstein found that the speed of light in a vacuum, c, is the fastest speed at which a body can theoretically travel. In Newtonian mechanics there is a universal time measurement, whereas Einstein showed that each frame has its own time measurement, as well as its own coordinate system. This means that observers in different inertial reference frames will not agree on the time duration of events. Although there are invariant properties that observers in different reference frames will agree on, such as the space-time interval of events, shown in Appendix F. In special relativity the calculations of properties in different reference frames were found by the Lorentz transformations, which replaced the Galilean transformations.

The results of special relativity imply that the electromagnetic fields as seen from an observer in one

inertial reference frame will not agree with the observed values in an alternative inertial reference frame. This effect of motion on the electromagnetic field was initially formulated by Minkowski and has since become standard [62]. Subsequent work has focussed on the propagation of waves through moving media [101, 102]. It was found that relative motion produces a mixing effect on the observed electromagnetic fields, with electric effects causing additional magnetic responses and vice versa. In general, this is not dissimilar to bianisotropic media with a continuous duality symmetry. With the knowledge of the electrodynamics in moving media, the next step was to consider the quantised electromagnetic field in moving absorbing media. The approaches include a relativistic formulation [103] and a fully covariant framework [104] that can be subject to Lorentz transformations and therefore can describe the quantised electromagnetic field in absorbing media that is in motion with respect to an arbitrary observer. Further theories include a description of the quantised electromagnetic field in moving media in terms of macroscopic variables [105] and a canonical formulation [19, 106].

Recent interest in the quantisation of the electromagnetic field in moving absorbing media has been driven by the debate over resistive forces between objects in relative motion, otherwise known as Quantum friction [107–110]. As was been discussed in section 2.2 the correlations of quantum fluctuations of the electromagnetic field can generate dispersion forces between polarisable and magnetisable objects. The dispersion forces are dependent on the properties of the objects from the perspective of the source and observation point. This means that if the objects are moving relative to each other, additional effects such as the Doppler shift and the mixing of electric and magnetic phenomena can become important in the dispersion force and can produce velocity dependent components as well as velocity dependent corrections. Some of the geometries studied include dielectric slabs moving relative to each other [111], a moving atom near a stationary dielectric [112, 113] and a moving medium and near a stationary two level detector [106].

In this chapter the quantisation of the electromagnetic field in moving media will be achieved by treating moving media as linear non-reciprocal bianisotropic media. The properties of the Green's function in moving media will be discussed. The Casimir-Polder potential between a stationary atom and a moving medium will be calculated in the low velocity and non-retarded regimes. The work in the chapter is taken from the manuscript Ref. [114].

6.1 Field Quantisation in Moving Media

The constitutive relations for a locally responding isotropic magnetodielectric are

$$\hat{\mathbf{D}}' = \varepsilon_0 \boldsymbol{\varepsilon}' \cdot \hat{\mathbf{E}}' + \hat{\mathbf{P}}'_{\mathrm{N}}, \tag{6.2a}$$

$$\hat{\mathbf{B}}' = \mu_0 \boldsymbol{\mu}' \cdot (\hat{\mathbf{H}}' + \hat{\mathbf{M}}'_{\mathrm{N}}), \tag{6.2b}$$

where

$$\boldsymbol{\varepsilon}' = \varepsilon'(\omega')\boldsymbol{I}, \tag{6.3a}$$

$$\boldsymbol{\mu}' = \mu'(\omega')\boldsymbol{I}. \tag{6.3b}$$

The constitutive relations, Eqs. (6.2), describe the medium's response to an applied electromagnetic field from the perspective of an observer that is not in motion with respect to the medium. This defines an inertial reference frame, in which the medium appears stationary, called the rest frame. From the perspective of other inertial reference frames the medium will be in motion and Eqs. (6.2) will not describe the medium's response to an applied electromagnetic field. This means that the relative velocity of the medium with respect to an arbitrary reference frame changes the observed behaviour of

the electromagnetic field in the medium.

To investigate the effect of motion on the electromagnetic fields properties in media, an isotropic magnetodielectric medium as described by Eqs. (6.2) is considered to be moving at a constant velocity, $\mathbf{v}$, with respect to a laboratory frame. To distinguish between values observed in the laboratory frame and values observed in the rest frame, the quantities observed in the mediums rest frame are denoted by primes, for example, $\mathbf{E}'$, etc. It is assumed that the origins of the coordinate systems are initially aligned, i.e.,

$$x' = x,\, y' = y,\, z' = z \quad \text{at} \quad t' = t = 0. \tag{6.4}$$

The constitutive relations of the medium from the perspective of the laboratory frame can now be obtained by applying the Lorentz transformations to the fields, this is shown in Appendix F. The result is a set of constitutive relations of the form,

$$\hat{\mathbf{D}} = \varepsilon_0 \boldsymbol{\varepsilon} \cdot \hat{\mathbf{E}} + \hat{\mathbf{P}}_{\mathrm{N}} + \frac{1}{c}\boldsymbol{\xi} \cdot \hat{\mathbf{H}} + \frac{1}{c}\boldsymbol{\xi} \cdot \hat{\mathbf{M}}_{\mathrm{N}}, \tag{6.5a}$$

$$\hat{\mathbf{B}} = \mu_0 \boldsymbol{\mu} \cdot \hat{\mathbf{H}} + \frac{1}{c}\boldsymbol{\zeta} \cdot \hat{\mathbf{E}} + \mu_0 \boldsymbol{\mu} \cdot \hat{\mathbf{M}}_{\mathrm{N}}, \tag{6.5b}$$

where the effective response functions $\boldsymbol{\varepsilon}, \boldsymbol{\zeta}, \boldsymbol{\xi}$ and $\boldsymbol{\mu}$ are defined as

$$\boldsymbol{\varepsilon} = \varepsilon'(\omega')\left(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1 - \frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}}\right), \tag{6.6a}$$

$$\boldsymbol{\zeta} = \frac{\gamma'^2}{c}\left(1 - \varepsilon'(\omega')\mu'(\omega')\right)\mathbf{v} \times \boldsymbol{I}, \tag{6.6b}$$

$$\boldsymbol{\xi} = \frac{\gamma'^2}{c}\left(\varepsilon'(\omega')\mu'(\omega') - 1\right)\mathbf{v} \times \boldsymbol{I}, \tag{6.6c}$$

$$\boldsymbol{\mu} = \mu'(\omega')\left(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1 - \frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}}\right). \tag{6.6d}$$

The Lorentz factor, γ, and medium assisted Lorentz factor, γ' are defined as

$$\gamma = \frac{1}{\sqrt{1 - \frac{v^2}{c^2}}}, \tag{6.7a}$$

$$\gamma' = \frac{1}{\sqrt{1 - \frac{v^2}{(c')^2}}}, \quad (c')^2 = \frac{1}{\mu'\varepsilon'\mu_0\epsilon_0}, \tag{6.7b}$$

respectively. It should be noted that the response functions in the constitutive relations that serve as magnetoelectric coupling terms, $\boldsymbol{\xi}$ and $\boldsymbol{\zeta}$, are related by

$$\boldsymbol{\xi} = -\boldsymbol{\zeta}. \tag{6.8}$$

The transpose of the cross response functions reveals further relationships,

$$\boldsymbol{\xi} = \boldsymbol{\zeta}^{\mathrm{T}},\ \boldsymbol{\xi}^{\mathrm{T}} = \boldsymbol{\zeta}. \tag{6.9}$$

This can be contrasted with the relationship between the magnetoelectric response functions in reciprocal media, Eq. (4.29). As Eq. (6.9) and Eq. (4.29) are not the same, this confirms that moving media have a non-reciprocal response in reference frames other than their rest frame. The constitutive relations for moving media from the perspective of a laboratory frame, Eqs. (6.5), are of the same form as the

constitutive relations for general linear bianisotropic media, Eqs. (4.10a) and (4.10b). This means that moving media can be considered as a subclass of non-reciprocal, linear bianisotropic media. Chapter 4 detailed the quantisation of the electromagnetic field in non-reciprocal linear bianisotropic media, in terms of explicit response functions. Therefore the quantisation of the electromagnetic field in moving media has already been achieved. However in the Hamiltonian as shown in Eq. (4.23), the lower bound of the frequency is in fact $-\gamma\mathbf{v}\cdot\mathbf{k}$ and not 0, i.e. there can be negative frequencies

$$\hat{H} = \int \mathrm{d}^3 r \int_{-\gamma\mathbf{v}\mathbf{k}}^{\infty} \mathrm{d}\omega\, \hbar\omega\, \hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega). \tag{6.10}$$

The form of Eqs. (6.5) shows that media can only be considered as isotropic in one reference frame, the frame in which it is stationary.

The response functions as shown in Eqs. (6.6) are dependent on the frequency in the mediums rest frame, ω', through the scalar permittivity and permeability from the moving mediums rest frame, $\varepsilon'(\omega')$ and $\mu'(\omega')$. The laboratory frame frequency, ω, is linked to the moving mediums rest frame frequency by the relativistic equation

$$\omega' = \gamma(\omega - \mathbf{k}\cdot\mathbf{v}) \tag{6.11}$$

in k - space, as is shown in Appendix F. By making the assumption that in the partially Fourier transformed domain $(\mathbf{r},\omega)$, the Fourier transform $\nabla \rightarrow i\mathbf{k}$ holds, this leads to an alternative relationship between the frequencies

$$\omega' = \gamma(\omega + i\mathbf{v}\cdot\nabla). \tag{6.12}$$

This difference between the frequency in the rest frame and the frequency in the laboratory frame is the origin of the Doppler effect. The medium in motion was assumed to be a homogeneous, locally responding magnetodielectric in its rest frame. This means that the response functions in the rest frame were not dependent on the position, $\mathbf{r}'$. By applying a Lorentz transformation to the position vector, the relationship between the position vectors in the laboratory frame and the mediums rest frame is

$$\mathbf{r}' = \mathbf{r}_{\perp} + \gamma(\mathbf{r}_{\parallel} - \mathbf{v}t), \tag{6.13}$$

which is shown in Appendix F. The subscripts, $\perp$ and $\parallel$ denote the components of the position vector that are perpendicular or parallel to the velocity vector, respectively. This can be contrasted with the Galilean transformation, shown in Eq. (6.1). As Eq. (6.13) shows, there is an additional time dependence that can generate non-local effects in an inhomogeneous medium in motion.

6.2 Properties of the Green's Function for Moving Media

A fundamental principle of special relativity is that the laws of physics are the same in all inertial reference frames, this means that the electromagnetic fields are governed by Maxwell's equations in all inertial reference frames. A plane wave propagating through an inertial frame will appear as a plane wave in all inertial frames. This can be shown by considering a plane wave propagating in the rest frame of the moving medium, modelled by the expression

$$e^{i\mathbf{k}'\cdot\mathbf{r}'-i\omega' t'} = e^{ik^{\mu'}x_{\mu'}} \tag{6.14}$$

which is in terms of the fourvectors defined in Appendix F. The Lorentz transformation of the contraction $k^{\mu'}x_{\mu'}$ is

$$k^{\mu'}x_{\mu'} = \Lambda_{\mu}^{\mu'}\Lambda_{\mu'}^{\mu}k^{\mu}x_{\mu} = k^{\mu}x_{\mu}, \quad \Lambda_{\mu}^{\eta}\Lambda_{\sigma}^{\mu} = \delta_{\sigma}^{\eta}, \tag{6.15}$$

which shows that $k^{\mu'}x_{\mu'}$ is invariant under a Lorentz transformation and therefore a plane wave structure is preserved across different inertial frames,

$$e^{ik^{\mu'}x_{\mu'}} = e^{ik^{\mu}x_{\mu}} = e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t}. \tag{6.16}$$

However, this does not mean that the fields are necessarily time harmonic in all frames. Here, it is assumed that the fields are time harmonic in the laboratory frame, this means that

$$\frac{\partial}{\partial t}\mathbf{E} = -i\omega\mathbf{E}, \tag{6.17}$$

holds, although

$$\frac{\partial}{\partial t'}\mathbf{E}' = -i\omega'\mathbf{E}' \tag{6.18}$$

is not necessarily true.

The moving medium is treated as a non-reciprocal linear bianisotropic medium with time harmonic fields in the laboratory frame, this means that the Helmholtz vector wave equation for the electric field is

$$\left[\nabla\times\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I} - \frac{i\omega}{c}\left(\nabla\times\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta} - \boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}\right) - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta})\right]\cdot\hat{\mathbf{E}}(\mathbf{r},\omega) = i\omega\mu_0\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega). \tag{6.19}$$

In the definition of the response functions, Eqs. (6.6), the scalar permittivity and permeability are functions of the frequency in the laboratory frame, i.e.,

$$\varepsilon' = \varepsilon'(\gamma(\omega + i\mathbf{v}\cdot\nabla)), \tag{6.20a}$$

$$\mu' = \mu'(\gamma(\omega + i\mathbf{v}\cdot\nabla)). \tag{6.20b}$$

The electric field is defined as

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = i\omega\mu_0\int \mathrm{d}^3r\ \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}',\omega) \tag{6.21}$$

and therefore the Green's function is the unique solution to

$$\left[\nabla\times\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I} - \frac{i\omega}{c}\left(\nabla\times\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta} - \boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}\right) - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta})\right]\cdot\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'). \tag{6.22}$$

The Green's function is causal, obeys the Schwarz reflection principle, Eq. (2.15), and is constrained by $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \mathbf{0}$ in the limit $|\mathbf{r}-\mathbf{r}'| \to \infty$. A medium in motion is not reciprocal in a laboratory frame, therefore the Green's function defined above will not be reciprocal, i.e., Eq. (2.16) does not necessarily hold. This is because the velocity must also be reversed in order for the system to be time invariant. With this in mind, an adapted reciprocity condition for moving media can be derived. As discussed in Appendix D, the Green's function can be thought of as the inverse to the Helmholtz matrix operator and a left inverse as well as a right inverse exist. This means that Eq. (6.22) can alternatively be written as

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\left[\boldsymbol{I}\times\overleftarrow{\nabla}'\cdot\boldsymbol{\mu}^{-1}\times\overleftarrow{\nabla}' + \frac{i\omega}{c}\left(\boldsymbol{I}\times\overleftarrow{\nabla}'\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta} - \boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\times\overleftarrow{\nabla}'\right) - \frac{\omega^2}{c^2}\left(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta}\right)\right] = \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'). \tag{6.23}$$

The curl operators are now acting on the second spatial variable, this adjustment changes the sign of the curl terms, which means that the central terms, dependent on a single curl expression, change sign.

Taking the transpose of Eq. (6.23) leads to

$$\begin{aligned}&\Big[\nabla'\times\boldsymbol{\mu}^{-1\mathrm{T}}\cdot\nabla'\times\boldsymbol{I}+\frac{i\omega}{c}\big(-\boldsymbol{\zeta}^{\mathrm{T}}\cdot\boldsymbol{\mu}^{-1\mathrm{T}}\cdot\nabla'\times\boldsymbol{I}+\nabla'\times\boldsymbol{\mu}^{-1\mathrm{T}}\cdot\boldsymbol{\xi}^{\mathrm{T}}\big)\\&-\frac{\omega^2}{c^2}\big(\boldsymbol{\varepsilon}^{\mathrm{T}}-\boldsymbol{\zeta}^{\mathrm{T}}\cdot\boldsymbol{\mu}^{-1\mathrm{T}}\cdot\boldsymbol{\xi}^{\mathrm{T}}\big)\Big]\cdot\boldsymbol{G}^{\mathrm{T}}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}').\end{aligned}\tag{6.24}$$

The transpose of the response functions as defined in Eq. (6.6) are

$$\boldsymbol{\varepsilon}=\boldsymbol{\varepsilon}^{\mathrm{T}},\tag{6.25a}$$

$$\boldsymbol{\zeta}=\boldsymbol{\xi}^{\mathrm{T}},\tag{6.25b}$$

$$\boldsymbol{\xi}=\boldsymbol{\zeta}^{\mathrm{T}},\tag{6.25c}$$

$$\boldsymbol{\mu}=\boldsymbol{\mu}^{\mathrm{T}}.\tag{6.25d}$$

Substituting Eqs. (6.25) into Eq. (6.24) and swapping the spatial variables leads to

$$\Big[\nabla\times\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}+\frac{i\omega}{c}\big(\nabla\times\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}\big)-\frac{\omega^2}{c^2}\big(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta}\big)\Big]\cdot\boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}').\tag{6.26}$$

It can be seen that Eq. (6.26) is not the same as Eq. (6.22), due to the different sign in front of the second term on the left hand sides. This confirms that for moving media,

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\neq\boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega).\tag{6.27}$$

The effect of a reversal of the velocity vector, i.e., $\mathbf{v}\to-\mathbf{v}$, on the response functions defined above is as follows,

$$\boldsymbol{\varepsilon}(\mathbf{v})=\boldsymbol{\varepsilon}(-\mathbf{v}),\tag{6.28a}$$

$$\boldsymbol{\mu}(\mathbf{v})=\boldsymbol{\mu}(-\mathbf{v}),\tag{6.28b}$$

$$\boldsymbol{\zeta}(\mathbf{v})=-\boldsymbol{\zeta}(-\mathbf{v}),\tag{6.28c}$$

$$\boldsymbol{\xi}(\mathbf{v})=-\boldsymbol{\xi}(-\mathbf{v}),\tag{6.28d}$$

$$\boldsymbol{\zeta}(\mathbf{v})=\boldsymbol{\xi}(-\mathbf{v}).\tag{6.28e}$$

Although the expressions for $\boldsymbol{\varepsilon}$ and $\boldsymbol{\mu}$ are not altered by a reversal of velocity, $\boldsymbol{\zeta}$ and $\boldsymbol{\xi}$ are affected, as is seen by the sign change.

The reversed velocity response functions, Eqs. (6.28), are substituted into Eq. (6.26), to give

$$\Big[\nabla\times\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}-\frac{i\omega}{c}\big(\nabla\times\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}\big)-\frac{\omega^2}{c^2}\big(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta}\big)\Big]\cdot\boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega,-\mathbf{v})=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}').\tag{6.29}$$

The Helmholtz matrix operator in Eq. (6.29) is now the same as the Helmholtz matrix operator in Eq. (6.22). This means that the Green's functions defined by each Helmholtz matrix must be equivalent. Therefore an adapted reciprocal condition for moving media of the form,

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega,\mathbf{v})=\boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega,-\mathbf{v}),\tag{6.30}$$

holds for constant velocity, $\mathbf{v}$.

6.3 Properties of Moving Media

To describe the propagation of the electromagnetic field through a medium, the dispersion relation and the vector wave functions are required.

6.3.1 Dispersion Relation for Moving Media

As shown in Appendix E, the dispersion relation can be found by solving

$$\mathrm{Det}[\boldsymbol{H}(k)] = 0, \tag{6.31}$$

for k. For moving media, the Helmholtz matrix operator $\boldsymbol{H}$ is

$$\boldsymbol{H} = -\mathbf{k}\times\boldsymbol{\mu}^{-1}\cdot\mathbf{k}\times\boldsymbol{I} + \frac{\omega}{c}\left(\mathbf{k}\times\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta} - \boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\mathbf{k}\times\boldsymbol{I}\right) - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta}), \tag{6.32}$$

in k- space. The dispersion relation is usually solved for k_z, the full dispersion relation, k, can be found by use of

$$k_z^2 + k_y^2 + k_x^2 = k^2. \tag{6.33}$$

The dispersion relation of the medium found by solving Eq. (6.31) with Eq. (6.32) will be from the perspective of the laboratory frame, the dispersion relation seen in the mediums rest frame will be different.

6.3.2 Vector Wave Functions for Moving Media

As is discussed in Appendix E, the vector wave functions of a wave propagating through a medium can be found using

$$\boldsymbol{H}_i\cdot\mathbf{P}(k_i) = \mathbf{0}, \tag{6.34}$$

where k_i are the solutions of the dispersion relation for the medium described by the Helmholtz matrix operator, $\boldsymbol{H}_i$. The vector wave functions introduced earlier, Eqs. (2.57), are not solutions to Eq. (6.34) and therefore additional vector wave functions are required. The vector wave functions for moving media are labelled as $\mathbf{P}$ and $\mathbf{Q}$ and they must solve the homogeneous Helmholtz equation

$$\begin{gathered}\left[\nabla\times\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I} - \frac{i\omega}{c}\left(\nabla\times\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta} - \boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\nabla\times\boldsymbol{I}\right) - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}-\boldsymbol{\xi}\cdot\boldsymbol{\mu}^{-1}\cdot\boldsymbol{\zeta})\right]_i\cdot\sigma(k_i) = \mathbf{0}, \\ \sigma = \mathbf{P}, \mathbf{Q}.\end{gathered} \tag{6.35}$$

The two vector wave functions must also be perpendicular to each other, i.e.,

$$\mathbf{P}\cdot\mathbf{Q} = 0. \tag{6.36}$$

In Appendix E, the initial trial solution to the Helmholtz matrix operator is of the form

$$\nabla\times\mathbf{c}e^{i\mathbf{k}\cdot\mathbf{r}}. \tag{6.37}$$

Letting the arbitrary pilot vector, $\mathbf{c}$, equal the velocity, $\mathbf{v}$, gives a vector wave function of the form

$$\mathbf{P} = \nabla\times\mathbf{v}e^{i\mathbf{k}\cdot\mathbf{r}} = i\begin{pmatrix} k_y v_z - k_z v_y \\ k_z v_x - k_x v_z \\ k_x v_y - k_y v_x \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}}. \tag{6.38}$$

Substitution of Eq. (6.38) into Eq. (6.35) confirms that it is indeed a solution to Eq. (6.35). The vector wave functions defined in Eqs. (2.57) are related to each other by a curl operation. Taking the curl of Eq. (6.38) leads to an expression of the form

$$\mathbf{Q} \propto \nabla \times \mathbf{P}, \tag{6.39}$$

which is not a solution to Eq. (6.35). However, a second solution to Eq. (6.35) that is perpendicular to Eq. (6.38) can be found, it is of the form

$$\mathbf{Q} = \frac{1}{i\omega\mu_0}\boldsymbol{\mu}^{-1}\cdot\left((\nabla - \frac{i\omega}{c}\boldsymbol{\zeta})\times\mathbf{P}\right). \tag{6.40}$$

It should be noted that the vector wave functions in Eqs. (6.38) and (6.40) are poorly defined when $\mathbf{k} \parallel \mathbf{v}$. Although in the limit of $v \to 0$, the vector wave function does not necessarily become undefined. If the velocity vector is kept as a unit vector while the velocity magnitude tends to zero, the vector wave functions can still exist.

6.4 Low Velocity Approximation

When the velocity is small, i.e., $v \lll c$, approximations can be made which simplify the response functions and the dispersion relation. The small size of the velocity means that terms of order v^2 or higher can be neglected. The gamma factors, γ, γ', shown in Eqs. (6.7a) and (6.7b), become

$$\gamma, \gamma' \to 1, \tag{6.41}$$

for slowly moving media. This alters the response functions defined in Eqs. (6.6) to the following

$$\boldsymbol{\varepsilon} = \varepsilon'\boldsymbol{I}, \tag{6.42a}$$

$$\boldsymbol{\zeta} = \frac{1}{c}(1 - \varepsilon'\mu')\mathbf{v}\times\boldsymbol{I}, \tag{6.42b}$$

$$\boldsymbol{\xi} = \frac{1}{c}(\varepsilon'\mu' - 1)\mathbf{v}\times\boldsymbol{I}, \tag{6.42c}$$

$$\boldsymbol{\mu} = \mu'\boldsymbol{I}, \tag{6.42d}$$

where $\varepsilon' = \varepsilon'(\omega + i\mathbf{v}\cdot\nabla)$ and $\mu' = \mu'(\omega + i\mathbf{v}\cdot\nabla)$. The small value of v means that ε' and μ' can be Taylor expanded around $\mathbf{v} = 0$ to obtain

$$\varepsilon'(\omega') = \varepsilon'(\omega + i\mathbf{v}\cdot\nabla) \approx \varepsilon'(\omega) + i(\mathbf{v}\cdot\nabla)\frac{\partial}{\partial\omega}(\varepsilon'(\omega)) + \ldots \tag{6.43a}$$

$$\mu'(\omega') = \mu'(\omega + i\mathbf{v}\cdot\nabla) \approx \mu'(\omega) + i(\mathbf{v}\cdot\nabla)\frac{\partial}{\partial\omega}(\mu'(\omega)) + \ldots \tag{6.43b}$$

These are substituted into Eqs. (6.42) to obtain

$$\boldsymbol{\varepsilon} = (\varepsilon'(\omega) + i(\mathbf{v}\cdot\nabla)\frac{\partial}{\partial\omega}(\varepsilon'(\omega)))\boldsymbol{I}, \tag{6.44a}$$

$$\boldsymbol{\zeta} = \frac{1}{c}(1 - \varepsilon'(\omega)\mu'(\omega))\mathbf{v}\times\boldsymbol{I}, \tag{6.44b}$$

$$\boldsymbol{\xi} = \frac{1}{c}(\varepsilon'(\omega)\mu'(\omega) - 1)\mathbf{v}\times\boldsymbol{I}, \tag{6.44c}$$

$$\boldsymbol{\mu} = (\mu'(\omega) + i(\mathbf{v}\cdot\nabla)\frac{\partial}{\partial\omega}(\mu'(\omega)))\boldsymbol{I}, \tag{6.44d}$$

which have been truncated to $\mathcal{O}(v)$. In k - space it can be seen that this expansion is only valid when k is small.

The dispersion relation for k_z can be found by solving Eq. (6.31) as

$$k_z = \frac{v_z(\zeta-\xi)\omega \pm 2c\sqrt{-c^2k_s^2 + \omega((\mathbf{k}_s\cdot\mathbf{v})(\zeta-\xi) + (\varepsilon\mu - \mathbf{k}\cdot\mathbf{v}\partial_\omega(\varepsilon\mu))\omega)c^2}}{2c^2}, \tag{6.45}$$

where the notational shorthands

$$\zeta = 1 - \varepsilon\mu, \tag{6.46a}$$

$$\xi = \varepsilon\mu - 1, \tag{6.46b}$$

have been introduced for convenience.

6.5 Casimir-Polder Potential between Moving Medium and Particle

The Casimir-Polder potential between a particle and macroscopic bodies which included bianisotropic effects was shown in chapter 5. The same derivation can be used to obtain the Casimir-Polder potential in a non-reciprocal system and the off-resonant contribution to the potential is

$$\begin{aligned} U_O^n(\mathbf{r}_A) = \frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi \mathrm{tr}\Big[&\xi^2\boldsymbol{\alpha}^n(i\xi)\cdot\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,i\xi) + \xi\boldsymbol{\Gamma}_{em}^n(i\xi)\cdot\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,i\xi)\times\overleftarrow{\nabla} \\ &+ \xi\boldsymbol{\Gamma}_{me}^n(i\xi)\cdot\nabla\times\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,i\xi) + \boldsymbol{\beta}^n(i\xi)\cdot\nabla\times\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,i\xi)\times\overleftarrow{\nabla}\Big] \end{aligned} \tag{6.47}$$

where the polarisabilities are defined as

$$\boldsymbol{\alpha}^n(i\xi) = \frac{1}{\hbar}\sum_k\left(\frac{\hat{\mathbf{d}}_{kn}\otimes\hat{\mathbf{d}}_{nk}}{\omega_{kn}+i\xi} + \frac{\hat{\mathbf{d}}_{nk}\otimes\hat{\mathbf{d}}_{kn}}{\omega_{kn}-i\xi}\right) \tag{6.48a}$$

$$\boldsymbol{\Gamma}_{em}^n(i\xi) = \frac{1}{\hbar}\sum_k\left(\frac{\hat{\mathbf{m}}_{kn}\otimes\hat{\mathbf{d}}_{nk}}{\omega_{kn}+i\xi} - \frac{\hat{\mathbf{m}}_{nk}\otimes\hat{\mathbf{d}}_{kn}}{\omega_{kn}-i\xi}\right) \tag{6.48b}$$

$$\boldsymbol{\Gamma}_{me}^n(i\xi) = \frac{1}{\hbar}\sum_k\left(\frac{\hat{\mathbf{d}}_{kn}\otimes\hat{\mathbf{m}}_{nk}}{\omega_{kn}+i\xi} - \frac{\hat{\mathbf{d}}_{nk}\otimes\hat{\mathbf{m}}_{kn}}{\omega_{kn}-i\xi}\right) \tag{6.48c}$$

$$\boldsymbol{\beta}^n(i\xi) = \frac{1}{\hbar}\sum_k\left(\frac{\hat{\mathbf{m}}_{kn}\otimes\hat{\mathbf{m}}_{nk}}{\omega_{kn}+i\xi} + \frac{\hat{\mathbf{m}}_{nk}\otimes\hat{\mathbf{m}}_{kn}}{\omega_{kn}-i\xi}\right) \tag{6.48d}$$

and the resonant component is

$$\begin{aligned} U_R^n(\mathbf{r}_A) = -\mu_0\sum_k\Theta(\omega_{nk})\Big[&\omega_{nk}^2\hat{\mathbf{d}}_{nk}\cdot\mathcal{R}\mathrm{e}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\cdot\hat{\mathbf{d}}_{kn} \\ &+ i\mu_0\omega_{nk}\Big(\hat{\mathbf{d}}_{nk}\cdot\mathcal{R}\mathrm{e}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\times\nabla\cdot\hat{\mathbf{m}}_{kn} + \hat{\mathbf{m}}_{nk}\cdot\nabla\times\mathcal{R}\mathrm{e}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})\cdot\hat{\mathbf{d}}_{kn}]\Big) \\ &+ \mu_0\hat{\mathbf{m}}_{nk}\cdot(\nabla\times\mathcal{R}\mathrm{e}[\boldsymbol{G}(\mathbf{r}_A,\mathbf{r}_A,\omega_{nk})]\times\nabla)\cdot\hat{\mathbf{m}}_{kn}\Big]. \end{aligned} \tag{6.49}$$

As an example of the effect of the motion of a medium on the Casimir-Polder potential, an atom initially in its ground state situated near the boundary of a moving medium is envisioned. The moving

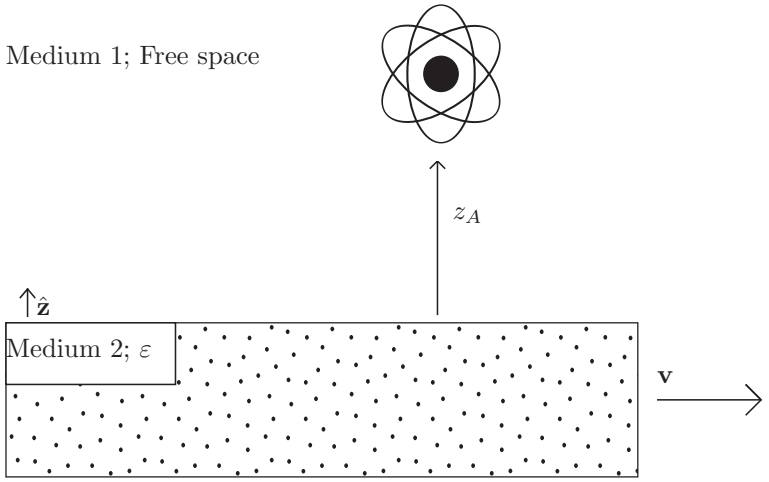


Figure 6.1: Shows the Casimir-Polder force between a stationary particle in free space near the boundary with a moving medium. In its rest frame, the medium is an isotropic dielectric, which is seen as a bianisotropic medium from the rest frame of the particle.

medium is an isotropic dielectric in its rest frame and is considered to be a halfspace. The boundary with free space is the $(x-y)$-plane at $z=0$, where $z>0$ is free space and $z<0$ is the medium. The motion is assumed to take place parallel to the interface in the $(x-y)$-plane, i.e.,

$$\mathbf{v} = (v_x, v_y, 0), \tag{6.50}$$

as is pictorially represented in Fig. 6.1. It will also be assumed that the velocity is sufficiently slow that the low velocity approximations can be made. In the free space region, the Green's function for this geometry is

$$\begin{aligned}\boldsymbol{G}_1(\mathbf{r},\mathbf{r}',\omega) = \frac{i}{8\pi^2}\iint_{-\infty}^{\infty}\frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\Big[&\mathbf{M}(\mathbf{k}_s,-k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\\ &+\mathbf{N}(\mathbf{k}_s,\pm k_z,\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,\mp k_z,\mathbf{r}')+r_{MM}\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\\ &+r_{MN}\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r}')+r_{NM}\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\\ &+r_{NN}\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\Big].\end{aligned} \tag{6.51}$$

This is the same structure for the scattering part of the Green's function as was used in chapter 5. This form is applicable due to the similarities between the two scenarios. The scattering problem in section 5.4 can be thought of as an electromagnetic source in free space near a biisotropic medium. Here, the scattering problem can be considered as an electromagnetic source in free space near a bianisotropic medium. This means that the fundamental scattering scenario is quite similar, the differences due to the different forms of media will appear in the reflection coefficients. In region 2, it is proposed that the Green's function is of the form

$$\begin{aligned}\boldsymbol{G}_2(\mathbf{r},\mathbf{r}',\omega) =\frac{i}{8\pi^2}\iint_{-\infty}^{\infty}\frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\Big[&t_{PM}\mathbf{P}(\mathbf{k}_s,-k_{z2},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\\ &+t_{QM}\mathbf{Q}(\mathbf{k}_s,-k_{z2},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r}')+t_{PN}\mathbf{P}(\mathbf{k}_s,-k_{z2},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\\ &+t_{QN}\mathbf{Q}(\mathbf{k}_s,-k_{z2},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r}')\Big],\end{aligned} \tag{6.52}$$

where t_{PM}, t_{QM}, t_{PN} and t_{QN} are the transmission coefficients. In the geometry shown in Fig. 6.1 and

at low velocity, the dispersion relation for the moving medium is

$$k_{z2} = \pm\sqrt{-k_s^2 + \frac{\omega}{c^2}((\mathbf{k}_s\cdot\mathbf{v})(\zeta-\xi) + (\varepsilon - (\mathbf{k}_s\cdot\mathbf{v})\partial_\omega(\varepsilon))\omega)}, \tag{6.53}$$

where the prime notation has been dropped for convenience. The full dispersion relation is now

$$k_2 = \pm\sqrt{\frac{\omega}{c^2}((\mathbf{k}_s\cdot\mathbf{v})(\zeta-\xi) + (\varepsilon - (\mathbf{k}_s\cdot\mathbf{v})\partial_\omega(\varepsilon))\omega)}. \tag{6.54}$$

6.5.1 Reflection and Transmission Coefficients

Applying Eq. (6.30) to Eq. (6.51) reveals the constraints on the cross term reflection coefficients. To be compliant with the modified reciprocity condition, the reflection coefficients behave as

$$r_{MM}(k_x,k_y,v_x,v_y) = r_{MM}(-k_x,-k_y,-v_x,-v_y), \tag{6.55a}$$

$$r_{NN}(k_x,k_y,v_x,v_y) = r_{NN}(-k_x,-k_y,-v_x,-v_y), \tag{6.55b}$$

$$r_{MN}(k_x,k_y,v_x,v_y) = r_{NM}(-k_x,-k_y,-v_x,-v_y), \tag{6.55c}$$

$$r_{NM}(k_x,k_y,v_x,v_y) = r_{MN}(-k_x,-k_y,-v_x,-v_y). \tag{6.55d}$$

These are derived by noting that the transpose of the Green's function can be found by switching the order of the dyads in the dyadic products, as shown in Eq. (E.31). The other transformations are straightforward, swapping the source and observation points and changing the sign of the velocity and $\mathbf{k}_s$ vector.

6.5.2 Bianisotropic Boundary Conditions

The reflection coefficients in Eq. (6.51) and the transmission coefficients in Eq. (6.52) are required. To obtain them, the boundary conditions for general linear bianisotropic media are needed. The derivation is shown in Appendix G and the boundary conditions are as follows,

$$\hat{\mathbf{e}}_\parallel\cdot\Big[\boldsymbol{G}_i - \boldsymbol{G}_j\Big] = \mathbf{0}, \tag{6.56a}$$

$$\hat{\mathbf{e}}_\parallel\cdot\big([\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \frac{i\omega}{c}\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i\star\boldsymbol{G}_i] - [\boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j - \frac{i\omega}{c}\boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j\star\boldsymbol{G}_j]\big) = \lim_{h\to0}\int \mathrm{d}\mathbf{h}\cdot\boldsymbol{\delta}. \tag{6.56b}$$

$$\hat{\mathbf{e}}_\perp\cdot[\nabla\times\boldsymbol{G}_i - \nabla\times\boldsymbol{G}_j] = \mathbf{0}, \tag{6.56c}$$

$$\begin{aligned}\hat{\mathbf{e}}_\perp\cdot\Bigg[\bigg(\frac{i\omega}{c}\boldsymbol{\xi}_i\star\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}_i - \boldsymbol{\xi}_i\star\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i)\star\boldsymbol{G}_i\bigg) - \bigg(\frac{i\omega}{c}\boldsymbol{\xi}_j\star\boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j \\ - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}_j - \boldsymbol{\xi}_j\star\boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j)\star\boldsymbol{G}_j\bigg)\Bigg] = \lim_{h\to0}\int_{h'}\mathrm{d}h'\nabla\boldsymbol{\delta},\end{aligned} \tag{6.56d}$$

where the i and j subscripts refer to different bianisotropic media that meet at a boundary, the expression $\boldsymbol{G}_i = \boldsymbol{G}(\mathbf{r}_i,\mathbf{r},\omega)$ is used for notational convenience. In Eqs. (6.56b) and (6.56d) the terms on the right hand side of the equality are zero if there is no surface charge density. The unit vectors $\hat{\mathbf{e}}_\parallel$ and $\hat{\mathbf{e}}_\perp$ refer to directions that are parallel and perpendicular to the boundary surface, respectiveley. As shown in Fig. 6.1, in the geometry used here, the $(x-y)$-plane is parallel to the boundary and therefore there are two parallel unit vectors. These are chosen to be $\hat{\mathbf{e}}_\parallel = \hat{\mathbf{e}}_M, \hat{\mathbf{e}}_{\mathbf{k}_s}$ [25] where

$$\hat{\mathbf{e}}_M = \frac{1}{k_s}(k_y,-k_x,0), \tag{6.57a}$$

$$\hat{\mathbf{e}}_{\mathbf{k}_s} = \frac{1}{k_s}(k_x, k_y, 0). \tag{6.57b}$$

The unit vector perpendicular to the boundary of the medium is therefore

$$\hat{\mathbf{e}}_\perp = \hat{\mathbf{e}}_z = (0, 0, 1). \tag{6.58}$$

The Green's function for the free space region, Eq. (6.51), requires 4 reflection coefficients and the Green's function for the moving medium, Eq. (6.52), needs 4 transmission coefficients. The linear independence of the vector wave functions $\mathbf{M}$ and $\mathbf{N}$ means that Eqs. (6.56a) and (6.56b) each contribute 4 expressions, two from each parallel unit vector. This means that Eqs. (6.56c) and (6.56d) are not required to obtain the reflection and transmission coefficients in this geometry. However, Eq. (6.56c) is equivalent to Eq. (6.56a) when $\hat{\mathbf{e}}_\parallel = \hat{\mathbf{e}}_M$ and Eq. (6.56d) is equivalent to Eq. (6.56b) when $\hat{\mathbf{e}}_\parallel = \hat{\mathbf{e}}_M$, this provides a consistency check. The equivalency derives from the fact that the boundary conditions can be thought of in terms of the electromagnetic fields, as is shown in Eqs. (G.14) to (G.17). The relationships between the fields and therefore the boundary condition equivalence are

$$B_z \propto k_x E_y - k_y E_x \propto \hat{\mathbf{e}}_M \cdot \hat{\mathbf{E}}, \tag{6.59a}$$

$$D_z \propto k_x H_y - k_y H_x \propto \hat{\mathbf{e}}_M \cdot \hat{\mathbf{H}}, \tag{6.59b}$$

where Maxwell's equations, Eqs. (2.1), have been used.

Applying Eqs. (6.51) and (6.52) to Eqs. (6.56a) and (6.56b) and solving for the reflection and transmission coefficients leads to reflection coefficients of the form

$$r_{MM} = \frac{k_{z1} - k_{z2}}{k_{z1} + k_{z2}} \tag{6.60a}$$

$$r_{MN} = \frac{2ik_{z1}(k_y v_x - k_x v_y)\zeta}{c(k_{z1} + k_{z2})(-k_{z2} + k_{z1}(k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon - \varepsilon))} \tag{6.60b}$$

$$r_{NM} = \frac{2ik_{z1}(k_y v_x - k_x v_y)\zeta}{c(k_{z1} + k_{z2})(-k_{z2} + k_{z1}(k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon - \varepsilon))} \tag{6.60c}$$

$$r_{NN} = \frac{k_{z1}(\varepsilon - k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon) - k_{z2}}{k_{z2} + k_{z1}(\varepsilon - k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon)} \tag{6.60d}$$

after the low velocity approximations have been made. The reflection coefficients obey the conditions Eqs. (6.55). In the non-retarded limit to first order, as discussed in section 2.2, the reflection coefficients become

$$r_{MM} = 0, \tag{6.61a}$$

$$r_{MN} = \frac{-i(k_y v_x - k_x v_y)(1 - \varepsilon)}{ck_{z1}(\varepsilon - k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon + 1)}, \tag{6.61b}$$

$$r_{NM} = \frac{-i(k_y v_x - k_x v_y)(1 - \varepsilon)}{ck_{z1}(\varepsilon - k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon + 1)}, \tag{6.61c}$$

$$r_{NN} = \frac{(\varepsilon - k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial_\omega\varepsilon - 1)}{(\varepsilon - k_s(\hat{\mathbf{k}}_s \cdot \mathbf{v})\partial\varepsilon + 1)}, \tag{6.61d}$$

where the substitution

$$\zeta = 1 - \varepsilon + k_s\hat{\mathbf{k}}_s \cdot \mathbf{v}\partial\varepsilon, \tag{6.62}$$

has been made.

An alternative method of obtaining the reflection and transmission coefficients is through transforming

the scattering matrix [102].

6.5.3 Casimir-Polder Potential

The reflection coefficients are substituted into the scattering part of the Green's function shown in Eq. (6.51). The trace of the Green's function is taken, as described in section E.2 and this is substituted into Eq. (6.47). The off-resonant part of the Casimir-Polder potential is

$$U_e^0(z_A) = \frac{\hbar\mu_0}{16\pi^3}\int_0^\infty \mathrm{d}\xi\xi^2 \int_0^{2\pi}\mathrm{d}\theta\int_0^\infty \frac{\mathrm{d}k_s k_s}{\tilde{k}_{z1}}\alpha^0(i\xi)\left(1-\frac{2\tilde{k}_{z1}^2}{\tilde{k}_1^2}\right)\left[\frac{(\varepsilon - k_s v\cos\theta\partial\varepsilon - 1)}{(\varepsilon - k_s v\cos\theta\partial\varepsilon + 1)}\right]e^{-2\tilde{k}_{z1}z_A} \quad (6.63)$$

for the electric component and

$$\begin{aligned} U_c^0(z_A) = \frac{i\hbar\mu_0}{8\pi^3 c}\int_0^\infty \mathrm{d}\xi\xi^2\int_0^{2\pi}\mathrm{d}\theta\int_0^\infty \frac{\mathrm{d}k_s k_s}{\tilde{k}_{z1}}(\Gamma^0_{em}(i\xi)+\Gamma^0_{me}(i\xi))\left(1-\frac{\tilde{k}_{z1}^2}{\tilde{k}_1^2}\right) \\ \times\left[\frac{(k_s v\sin\theta)(1-\varepsilon)}{c\tilde{k}_{z1}(\varepsilon - k_s v\cos\theta\partial\varepsilon+1)}\right]e^{-2\tilde{k}_{z1}z_A} \end{aligned} \quad (6.64)$$

for the cross terms. Although the atom is initially in its ground state, the negative frequencies in the Hamiltonian mean that there can be a contribution from the resonant component of the Casimir-Polder potential. The electric permittivity is a functions of complex frequency, i.e., $\varepsilon = \varepsilon(i\xi)$ and is therefore a real quantity.

A resonant contribution to the Casimir-Polder potential in this scenario will only occur if

$$|\omega_{kn}| < |\mathbf{v}\cdot\mathbf{k}|. \quad (6.65)$$

The electric part of the resonant component will be proportional to the real part of the Green's function evaluated at $\omega = -\mathbf{v}\cdot\mathbf{k}$. However, the resonant components will not be included in this calculation of the Casimir-Polder potential. The Taylor expansions of the response functions, Eqs. (6.43a) and Eq. (6.43b), are only valid for small v and k. Although the integral over $\tilde{k}_{z1}$ has an upper bound of ∞, the exponential function acts as a cut off and therefore k is restrained to small values and the expansion of the response functions can be used in this scenario. The integrals have been altered by

$$\int \mathrm{d}\mathbf{k}_s \to \int_0^{2\pi}\mathrm{d}\theta\int_0^\infty \mathrm{d}k_s k_s, \quad (6.66)$$

where it should be noted that the angle integral has not been performed yet. In Eqs. (6.63) and (6.64) the relationship

$$k_s\hat{\mathbf{k}}_s\cdot\mathbf{v} = k_s v\cos\theta, \quad (6.67)$$

has been used. In Eq. (6.64), the numerator of the reflection coefficients Eqs. (6.61b) and (6.61c) has been rewritten as

$$k_y v_x - k_x v_y = (\mathbf{v}\times\mathbf{k}_s)\cdot\hat{\mathbf{z}} = v k_s\sin\theta\hat{\mathbf{n}}\cdot\hat{\mathbf{z}}. \quad (6.68)$$

The velocity vector is constrained to the $(x-y)$-plane, as is the $\mathbf{k}_s$ vector, this means that $\hat{\mathbf{n}} = \hat{\mathbf{z}}$ and therefore $\hat{\mathbf{n}}\cdot\hat{\mathbf{z}} = 1$. In the non-retarded limit, $k_s \approx \tilde{k}_{z1}$, this means that Eqs. (6.63) and (6.64) become

$$U_e^0(z_A) = \frac{\hbar\mu_0}{16\pi^3}\int_0^\infty \mathrm{d}\xi\xi^2\int_0^{2\pi}\mathrm{d}\theta\int_{\frac{\xi}{c}}^\infty \mathrm{d}\tilde{k}_{z1}\alpha^0(i\xi)\left(1-\frac{2\tilde{k}_{z1}^2}{\tilde{k}_1^2}\right)\left[\frac{(\varepsilon-\tilde{k}_{z1}v\cos\theta\partial\varepsilon-1)}{(\varepsilon-\tilde{k}_{z1}v\cos\theta\partial\varepsilon+1)}\right]e^{-2\tilde{k}_{z1}z_A}, \quad (6.69a)$$

$$U_c^0(z_A) = \frac{i\hbar\mu_0}{8\pi^3 c}\int_0^\infty \mathrm{d}\xi\xi^2\int_0^{2\pi}\mathrm{d}\theta\int_0^\infty \mathrm{d}k_{z1}(\Gamma_{em}^0(i\xi)+\Gamma_{me}^0(i\xi))\left(1-\frac{\tilde{k}_{z1}^2}{\tilde{k}_1^2}\right)$$
$$\times\left[\frac{(\tilde{k}_{z1}v\sin\theta)(1-\varepsilon)}{c\tilde{k}_{z1}(\varepsilon-\tilde{k}_{z1}v\cos\theta\partial\varepsilon+1)}\right]e^{-2\tilde{k}_{z1}z_A}, \tag{6.69b}$$

respectively. It should be noted that the quantity $\partial[\varepsilon(i\xi)]$ is imaginary, whereas $\varepsilon(i\xi)$, v and $\tilde{k}_{z1}$ (in the non-retarded limit) are real valued. This means that there will not be any singularites and therefore poles when performing the contour integral in complex frequency space. Although the velocity is constant, as the integral is over all k_s, the angle between $\mathbf{v}$ and $\mathbf{k}_s$ changes with the angle of $\mathbf{k}_s$ in polar coordinates (denoted as θ). Therefore, it is assumed that the angle between $\mathbf{v}$ and $\mathbf{k}_s$ is aligned with the polar coordinate θ. In Eqs. (6.69a) and (6.69b) the angle integrals are performed,

$$\int_0^{2\pi}\mathrm{d}\theta\left(\frac{\varepsilon-1-\tilde{k}_{z1}v\cos\theta\partial\varepsilon}{\varepsilon+1-\tilde{k}_{z1}v\cos\theta\partial\varepsilon}\right)=2\pi\left[\frac{\varepsilon+1+\tilde{k}_{z1}v\partial\varepsilon-2\sqrt{\frac{\varepsilon+1+\tilde{k}_{z1}v\partial\varepsilon}{\varepsilon+1-\tilde{k}_{z1}v\partial\varepsilon}}}{\varepsilon+1+\tilde{k}_{z1}v\partial\varepsilon}\right], \tag{6.70a}$$

$$\int_0^{2\pi}\mathrm{d}\theta\left(\frac{(\tilde{k}_{z1}v\sin\theta)(1-\varepsilon)}{(\varepsilon+1-\tilde{k}_{z1}v\cos\theta\partial\varepsilon)}\right)=0. \tag{6.70b}$$

The result in Eq. (6.70b) has the implication that there is no contribution to the cross term in the Casimir-Polder potential to linear order in velocity. Substituting Eq. (6.70a) into Eq. (6.69a) leads to

$$U_e^0(z_A)=\frac{\hbar\mu_0}{8\pi^2}\int_0^\infty \mathrm{d}\xi\xi^2\alpha^0(i\xi)\int_{\frac{\xi}{c}}^\infty \mathrm{d}\tilde{k}_{z1}\left(1-\frac{2\tilde{k}_{z1}^2}{\tilde{k}_1^2}\right)\left[\frac{(\varepsilon+1+\tilde{k}_{z1}v\partial_\omega\varepsilon-2\sqrt{\frac{\varepsilon+1+\tilde{k}_{z1}v\partial_\omega\varepsilon}{\varepsilon+1-\tilde{k}_{z1}v\partial_\omega\varepsilon}})}{(\varepsilon+1+\tilde{k}_{z1}v\partial_\omega\varepsilon)}\right]e^{-2\tilde{k}_{z1}z_A}, \tag{6.71}$$

which can be written as

$$U_e^0(z_A)=\frac{\hbar\mu_0}{8\pi^2}\int_0^\infty \mathrm{d}\xi\xi^2\alpha^0(i\xi)\int_{\frac{\xi}{c}}^\infty \mathrm{d}\tilde{k}_{z1}\left(1-\frac{2\tilde{k}_{z1}^2}{\tilde{k}_1^2}\right)\left[\frac{\sqrt{(\varepsilon+1)^2-(\tilde{k}_{z1}v\partial\varepsilon)^2}-2}{\sqrt{(\varepsilon+1)^2-(\tilde{k}_{z1}v\partial\varepsilon)^2}}\right]e^{-2\tilde{k}_{z1}z_A}. \tag{6.72}$$

It can be seen that the only extra contribution to the Casimir-Polder potential comes from the effect that the Doppler shift in frequency has on the dielectrics permittivity, even in the low velocity limit. This cannot be solved analytically and therefore a numerical solution is required. Similar to Refs. [102, 108], it was found that to first order in velocity there is no additional contribution to the Casimir-Polder potential deriving from magnetoelectric mixing effects.

In this chapter the quantisation of the electromagnetic field in moving media has been presented. A medium that is isotropic in its rest frame will be observed as bianisotropic in an arbitrary inertial frame. From the perspective of a laboratory frame moving media is treated as equivalent to stationary non-reciprocal bianisotropic media. This ensures the quantisation of the electromagnetic fields in the medium and it ensures compliance with the fluctuation dissipation theorem. At low velocities, and to first order in velocity, the motion of the medium only affects the Casimir-Polder potential through a Doppler shift in frequency. In this regime there are no mixed components of the Casimir-Polder potential generated due to the motion.

Chapter 7

Conclusion and Outlook

The recent advances in nanotechnology and metamaterial research have opened up the possibility that media with specific desired properties can be constructed. The response of these media to an applied electromagnetic field can then be used to generate unusual properties of dispersion forces. For example, dispersion forces that can selectively discriminate between particles based on atomic or molecular properties can be used as a method to separate a mixed solution.

In order to understand the properties of the media and therefore the dispersion forces they can generate, it is necessary to extend macroscopic QED to include the description of all linear absorbing media. This thesis has shown that it is possible to encorporate all linear media within the macroscopic QED framework, this includes media with a non-reciprocal response, spatially non-local response and media described by bianisotropic response functions.

In chapter 3 the quantisation of the electromagnetic field in general linear absorbing media was presented. This can be applied to all linear absorbing media, including non-reciprocal and spatially non-local media and only prior knowledge of the mediums conductivity tensor is required. Chapter 4 showed the quantisation of the electromagnetic field in media described by bianisotropic response functions. It was found that the duality invariance is a continuous symmetry for non-reciprocal media, whereas it is a discrete symmetry for reciprocal bianisotropic media.

In chapter 5 the chiral component of the Casimir-Polder potential was derived and was found to exist only if the molecule and the medium are chiral. As an example, the Casimir-Polder potential between Dimethyl Disulfide and a chiral metamaterial was calculated and it was found that the electric component of the Casimir-Polder potential was larger than the chiral contribution to the Casimir-Polder potential by 12 orders of magnitude. It was found that in a cavity between chiral media of opposite handedness, a molecule initially in an excited state could be separated from its enantiomer if the electric component is suppressed sufficiently by a suitable choice of the mediums resonant frequency.

In chapter 6 the quantisation of the electromagnetic field in moving media was achieved by noting that from the viewpoint of an inertial laboratory frame, moving media can be considered as equivalent to stationary non-reciprocal bianisotropic media. This means that the results of chapters 3 and 4 ensure the quantisation of the electromagnetic field and compliance with the fluctuation dissipation theorem in moving media. A modified reciprocity condition for moving media was derived, which takes into account the need for velocity to be reversed in order to preserve time symmetry. It was found that, to first order in velocity, additional components of the Casimir-Polder potential are not generated between a stationary atom and a moving medium. In fact, in this regime the only effect of the motion is seen as a Doppler shift in the frequency which affects the dielectric permittivity of the medium in motion.

The work in this thesis has provided the framework for the quantisation of the electromagnetic field in general absorbing linear media. This can be applied to any media in the linear response regime, the

media can be described by a conductivity tensor or by magnetoelectric response functions. A potential application could be to use non-reciprocal media to detect CP (Charge Parity) violating particles through a dispersion force. The derivation of the chiral component of the Casimir-Polder potential is applicable to a general arrangement of chiral media near a chiral molecule and therefore further exploration with different geometries and strongly chiral media is the next step. The inclusion of higher order velocity terms in the Casimir-Polder force between a stationary atom and a moving medium could contribute to a consistent theory of the dispersion forces between objects in relative motion, quantum friction.

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Appendix A

Kramers-Kronig Relations

The electric susceptibility, $\boldsymbol{\chi}_{ee}(\mathbf{r},t)$, is a linear order response function that relates the polarisation field, $\mathbf{P}(\mathbf{r},t)$, to an applied electric field $\mathbf{E}(\mathbf{r},t)$ through

$$\mathbf{P}(\mathbf{r},t) = \varepsilon_0 \int_0^\infty \mathrm{d}\tau \boldsymbol{\chi}_{ee}(\mathbf{r},\tau)\cdot\mathbf{E}(\mathbf{r},t-\tau). \tag{A.1}$$

To comply with the causality requirement that for times $t<0$, $\boldsymbol{\chi}_{ee}(\mathbf{r},t)=\mathbf{0}$, the susceptibility can be rewritten as

$$\boldsymbol{\chi}_{ee}(\mathbf{r},t) = \Theta(t)\boldsymbol{\chi}_{ee}(\mathbf{r},t), \tag{A.2}$$

where $\Theta(t)$ is the step function. Following the procedure of Ref. [34], the step function is equivalent to $\Theta(t)=\frac{1}{2}(1+S(t))$ where $S(t)=\frac{t}{|t|}$ is the sign function. The Fourier transform of the step function is

$$\Theta(\omega) = \frac{i}{\omega - i\eta} = i\mathcal{P}\frac{1}{\omega} - \pi\delta(\omega), \tag{A.3}$$

where η is a small value that tends to zero from above. The right hand side of Eq. (A.3) is a statement of Sochotzki's formula, where $\mathcal{P}$ refers to the Cauchy principle value. This means that the Fourier transform of Eq. (A.2) is

$$\boldsymbol{\chi}_{ee}(\mathbf{r},\omega) = \frac{\mathcal{P}}{i\pi}\int_{-\infty}^{\infty}\mathrm{d}\omega' \frac{\boldsymbol{\chi}_{ee}(\mathbf{r},\omega')}{\omega-\omega'}. \tag{A.4}$$

Substituting $\boldsymbol{\chi}_{ee}(\mathbf{r},\omega) = \mathrm{Re}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega)] + i\mathrm{Im}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega)]$ into the above leads to the relations

$$\mathrm{Re}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega)] = \frac{\mathcal{P}}{\pi}\int_{-\infty}^{\infty}\mathrm{d}\omega' \frac{\mathrm{Im}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega')]}{\omega-\omega'}, \tag{A.5a}$$

$$\mathrm{Im}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega)] = -\frac{\mathcal{P}}{\pi}\int_{-\infty}^{\infty}\mathrm{d}\omega' \frac{\mathrm{Re}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega')]}{\omega-\omega'}. \tag{A.5b}$$

These are the Kramers - Kronig relations for the electric susceptibility response function. It can be seen that the causality condition on the susceptibility means that a real part of the susceptibility necessarily implies the existence of a non-zero imaginary part. However, it should be noted that the response function must go to zero as $\omega\to\infty$ in order for the Kramers - Kronig relations to exist. An equivalent set of relations can be found for the magnetic susceptibility, which links the magnetisation field and the magnetic induction field. In this thesis the permittivity, $\boldsymbol{\varepsilon}$, and permeability, $\boldsymbol{\mu}$, will be used in preference to the susceptibilities and the simple links between them are,

$$\mathrm{Re}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega)] + \boldsymbol{I} = \mathrm{Re}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)], \tag{A.6a}$$

$$\mathrm{Im}[\boldsymbol{\chi}_{ee}(\mathbf{r},\omega)] = \mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)], \tag{A.6b}$$

$$\mathrm{Re}[\boldsymbol{\chi}_{mm}(\mathbf{r},\omega)] - \boldsymbol{I} = \mathrm{Re}[\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)], \tag{A.6c}$$

$$\mathrm{Im}[\boldsymbol{\chi}_{mm}(\mathbf{r},\omega)] = \mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)]. \tag{A.6d}$$

Appendix B

Schwarz Reflection Principle

The Schwarz reflection principle, sometimes called the reality conditon, applies to real functions, where $f(t) = f^*(t)$. The Fourier transformation and subsequent inverse Fourier transformation of a function, $f(t)$, are as follows,

$$f(\mathrm{t}) = \int \frac{\mathrm{d}\omega}{2\pi} e^{-i\omega t} f(\omega), \tag{B.1a}$$

$$f(\omega) = \int \mathrm{d}t e^{i\omega t} f(t). \tag{B.1b}$$

Taking the complex conjugate of Eq. (B.1b) results in

$$f^*(\omega) = \int \mathrm{d}t e^{-i\omega^* t} f^*(t) = \int \mathrm{d}t e^{-i\omega^* t} f(t), \tag{B.2}$$

where in the far right equality the reality of the function has been used, i.e., $f(t) = f^*(t)$ has been substituted. This means that Eq. (B.2) now shows the Fourier transform of $f(-\omega^*)$. Therefore it has been shown that,

$$f^*(\omega) = f(-\omega^*), \tag{B.3}$$

the Schwarz reflection principle.

Appendix C

Reciprocity of the Green's Function

The Green's function is defined as the unique solution to the Helmholtz equation shown in Eq. (2.13)

$$\left[\nabla\times\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)\cdot\nabla\times\boldsymbol{I}-\frac{\omega^2}{c^2}\boldsymbol{\varepsilon}(\mathbf{r},\omega)\right]\cdot\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'). \tag{C.1}$$

This is the right inverse of the Helmholtz operator, which means that a left inverse must exist,

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\left[\boldsymbol{I}\times\nabla'\cdot\boldsymbol{\mu}^{-1}(\mathbf{r}',\omega)\times\nabla-\frac{\omega^2}{c^2}\boldsymbol{\varepsilon}(\mathbf{r}',\omega)\right]=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'). \tag{C.2}$$

By taking the transpose and swapping the spatial variables, $\mathbf{r}$ and $\mathbf{r}'$, Eq. (C.2) can be rewritten as

$$\left[\nabla\times\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)\cdot\nabla\times\boldsymbol{I}-\frac{\omega^2}{c^2}\boldsymbol{\varepsilon}(\mathbf{r},\omega)\right]\cdot\boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega)=\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'), \tag{C.3}$$

where the reciprocity of the of the permittivity and permeability tensors has been assumed. It can be seen that the Helmholtz operators in Eqs. (C.1) and (C.3) are the same so the Green's function defined must be equal,

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{G}^{\mathrm{T}}(\mathbf{r}',\mathbf{r},\omega). \tag{C.4}$$

Therefore the reciprocity condition, which follows from the reciprocity of the permittivity and permeability, has been shown.

Appendix D

Integral Relation for the Green's Function

D.1 Derivation of the Integral Relation

The integral relation connects the fluctuations of the system with the dissipation into the medium. In this appendix the general form of the integral relation is derived, followed by its application to various linear media. As is shown in Refs. [12, 40], the integral relation can be constructed as follows. In the most general form the solution to the Green's function is given by the equation

$$\int \mathrm{d}^3 s \boldsymbol{H}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'), \tag{D.1}$$

or

$$\boldsymbol{H}\star\boldsymbol{G} = \boldsymbol{\delta}, \tag{D.2}$$

in the convolution notation detailed in chapter 4. The term, $\boldsymbol{H}$, is the Helmholtz matrix operator and it is dependent upon the medium in question. This matrix can be seen as the left inverse of the Green's function, but it should be noted that the right inverse is equally valid, i.e.,

$$\boldsymbol{G}\star\boldsymbol{H} = \boldsymbol{\delta}, \tag{D.3}$$

holds. Taking the Hermitian conjugates of Eqs. (D.2) and (D.3), where Hermitian conjugation is denoted by a † superscript, are

$$\boldsymbol{G}^\dagger\star\boldsymbol{H}^\dagger = \boldsymbol{\delta}, \tag{D.4a}$$

$$\boldsymbol{H}^\dagger\star\boldsymbol{G}^\dagger = \boldsymbol{\delta}, \tag{D.4b}$$

respectively. By multiplying Eq. (D.3) by $\boldsymbol{G}^\dagger$ on the right, Eq. (D.4b) by $\boldsymbol{G}$ on the left and substracting the results, the most general result for the integral relation

$$\begin{aligned} &\boldsymbol{G}\star(\boldsymbol{H}-\boldsymbol{H}^\dagger)\star\boldsymbol{G}^\dagger = \boldsymbol{G}^\dagger - \boldsymbol{G} \\ &\boldsymbol{G}\star\mathcal{I}\mathrm{m}[\boldsymbol{H}]\star\boldsymbol{G}^\dagger = -\mathcal{I}\mathrm{m}[\boldsymbol{G}], \end{aligned} \tag{D.5}$$

is obtained. $\mathcal{I}$m denotes the generalised imaginary part of the tensor, as introduced in Eq. (3.6b), section 3.1. In coordinate space this is written as

$$\int \mathrm{d}^3 s \int \mathrm{d}^3 s' \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\mathcal{I}\mathrm{m}[\boldsymbol{H}(\mathbf{s},\mathbf{s}',\omega)]\cdot\boldsymbol{G}^\dagger(\mathbf{r}',\mathbf{s}',\omega) = -\mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)]. \tag{D.6}$$

D.2 Linear Media

D.2.1 Reciprocal Anisotropic Media

For linear anisotropic media described by the response functions, $\boldsymbol{\varepsilon}(\mathbf{r},\omega)$ and $\boldsymbol{\mu}(\mathbf{r},\omega)$, the Helmholtz matrix operator that defines the Green's function is

$$\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega) = \nabla\times\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)\cdot\nabla\times\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}') - \frac{\omega^2}{c^2}\boldsymbol{\varepsilon}(\mathbf{r},\omega)\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'), \tag{D.7}$$

which can be read off from Eq. (2.13). The imaginary part is

$$\mathrm{Im}[\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega)] = -\frac{\omega^2}{c^2}\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{r},\omega)]\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}') + \nabla\times\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{r},\omega)]\cdot\nabla\times\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'), \tag{D.8}$$

where it has been assumed that the frequencies are real, i.e, $\omega = \omega^*$. The integral relation can then be written as

$$\begin{aligned}\int \mathrm{d}^3 s \int \mathrm{d}^3 s' \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\Big[&\frac{\omega^2}{c^2}\mathrm{Im}[\boldsymbol{\varepsilon}(\mathbf{s},\omega)]\boldsymbol{\delta}(\mathbf{s}-\mathbf{s}') \\ &- \nabla\times\mathrm{Im}[\boldsymbol{\mu}^{-1}(\mathbf{s},\omega)]\cdot\nabla\times\boldsymbol{\delta}(\mathbf{s}-\mathbf{s}')\Big]\cdot\boldsymbol{G}^\dagger(\mathbf{r}',\mathbf{s}',\omega) = \mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)],\end{aligned} \tag{D.9}$$

When multiplied by $\frac{\hbar\mu_0\omega^2}{\pi}$, the definitions Eqs. (2.32a) and (2.32b) can be used to write the integral relation in the form

$$\sum_{\lambda=e,m}\int \mathrm{d}^3 s \boldsymbol{G}_\lambda(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_\lambda^\dagger(\mathbf{r}',\mathbf{s},\omega) = \frac{\hbar\mu_0\omega^2}{\pi}\mathrm{Im}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)]. \tag{D.10}$$

D.2.2 General Linear Media

A general linear medium can be described in terms of a complex conductivity tensor as shown in chapter 3. In this case the Helmholtz matrix operator can be read off Eq. (3.11)

$$\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega) = \left[\nabla\times\nabla\times\boldsymbol{I} - \frac{\omega^2}{c^2}\boldsymbol{I}\right]\cdot\boldsymbol{\delta}(\mathbf{r}-\mathbf{r}') - i\mu_0\omega\int \mathrm{d}^3 s \boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\delta(\mathbf{s}-\mathbf{r}'). \tag{D.11}$$

The generalised imaginary part of this Helmholtz matrix operator is

$$\mathcal{I}\mathrm{m}[\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega)] = -\mu_0\omega\mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)], \tag{D.12}$$

where it has been assumed that the frequency is real. This leads to an integral relation of

$$\mu_0\omega\int \mathrm{d}^3 s \int \mathrm{d}^3 s' \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\mathcal{R}\mathrm{e}[\boldsymbol{Q}(\mathbf{s},\mathbf{s}',\omega)]\cdot\boldsymbol{G}^\dagger(\mathbf{r}',\mathbf{s}',\omega) = \mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)]. \tag{D.13}$$

D.2.3 Bianisotropic Linear Media

Chapter 4 contains the quantisation of the electromagnetic field in linear bianisotropic media. The Helmholtz matrix operator can be read off from Eq. (4.13),

$$\boldsymbol{H} = \nabla\times\boldsymbol{\mu}^{-1}\star\nabla\times - \frac{i\omega}{c}\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} \; + \frac{i\omega}{c}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}), \tag{D.14}$$

in the convolution notation. The generalised imaginary part of this Helmholtz matrix operator is

$$\begin{aligned}\mathcal{I}\mathrm{m}[\boldsymbol{H}] = &-\frac{\omega^2}{c^2}\mathcal{I}\mathrm{m}[\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}] - \frac{\omega}{2c}\nabla\times(\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} - \boldsymbol{\mu}^{-1\dagger}\star\boldsymbol{\xi}^{\dagger}) \\ &+ \frac{\omega}{2c}(\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1} - \boldsymbol{\zeta}^{\dagger}\star\boldsymbol{\mu}^{-1\dagger})\star\nabla\times\boldsymbol{I} + \nabla\times\mathcal{I}\mathrm{m}[\boldsymbol{\mu}^{-1}]\star\nabla\times\boldsymbol{I},\end{aligned} \tag{D.15}$$

where it has been assumed that the frequencies are real. The integral relation can then be written as

$$\begin{aligned}&\Bigg[\boldsymbol{G}(\omega)\star\Bigg[\frac{\omega^2}{c^2}\mathcal{I}\mathrm{m}[\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}] + \frac{\omega}{2c}\nabla\times(\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} - \boldsymbol{\mu}^{-1\dagger}\star\boldsymbol{\xi}^{\dagger}) \\ &- \frac{\omega}{2c}(\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1} - \boldsymbol{\zeta}^{\dagger}\star\boldsymbol{\mu}^{-1\dagger})\star\nabla\times\boldsymbol{I} - \nabla\times\mathcal{I}\mathrm{m}[\boldsymbol{\mu}^{-1}]\star\nabla\times\boldsymbol{I}\Bigg]\star\boldsymbol{G}^{\dagger}(\omega)\Bigg](\mathbf{r},\mathbf{r}') = \mathcal{I}\mathrm{m}[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)].\end{aligned} \tag{D.16}$$

For a chiral medium, the Post substitutions described in section 4.2 can be made to obtain the integral relation. It can also be seen that Eq. D.16 is equivalent to Eq. (D.13) when $\boldsymbol{Q}$ is

$$\boldsymbol{Q} = (i\mu_0\omega)^{-1}\nabla\times(\boldsymbol{\mu}^{-1} - \boldsymbol{I})\times\overleftarrow{\nabla} + Z_0^{-1}\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} + Z_0^{-1}\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\times\overleftarrow{\nabla} - i\varepsilon_0\omega(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta} - \boldsymbol{I}), \tag{D.17}$$

as is shown in chapter 4.

Appendix E

Scattering Part of the Green's Function

E.1 Derivation of the Scattering Part of the Green's Function

The constitutive relations can be used with Maxwell's equations to obtain a wave (or Helmholtz) equation for the electric field,

$$\int \mathrm{d}^3 r' \boldsymbol{H}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{E}(\mathbf{r}', \omega) = i\omega\mu_0 \mathbf{j}_{\mathrm{N}}(\mathbf{r}, \omega), \tag{E.1}$$

where $\boldsymbol{H}$ is a Helmholtz matrix operator that is dependent on the medium in which the electric field is propagating. Equation (E.1) is solved by

$$\mathbf{E}(\mathbf{r}, \omega) = i\omega\mu_0 \int \mathrm{d}^3 r' \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{j}_{\mathrm{N}}(\mathbf{r}', \omega) \tag{E.2}$$

where the Green's function, $\boldsymbol{G}$, is defined as the solution to

$$\int \mathrm{d}^3 s \boldsymbol{H}(\mathbf{r}, \mathbf{s}, \omega) \cdot \boldsymbol{G}(\mathbf{s}, \mathbf{r}', \omega) = \boldsymbol{\delta}(\mathbf{r} - \mathbf{r}'). \tag{E.3}$$

When there are other media near the electromagnetic source, the picture is more complicated as the electromagnetic fields can reflect off (or transmit through) the boundaries of the other media. These scattering effects influence the propagation of the electromagnetic fields and therefore need to be part of the Green's function solution. In this case it is common to write the Green's function in terms of a bulk part, $\boldsymbol{G}^{(0)}$, and a scattering part, $\boldsymbol{G}^{(1)}$,

$$\boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{G}^{(0)}(\mathbf{r}, \mathbf{r}', \omega) + \boldsymbol{G}^{(1)}(\mathbf{r}, \mathbf{r}', \omega). \tag{E.4}$$

The bulk part is position independent and is part of the solution to the electric field in a region with an electromagnetic source term. The scattering part of the Green's function is position dependent and describes the propagation through media that arises due to the reflection or transmission from the boundaries between one media and another, different media. Both parts of the Green's function inherit the properties of the total Green's function, these include the Schwarz reflection condition, the behaviour in the spatial limits and the compliance with causality, as is discussed at the end of section 2.1.1. The reciprocity condition, Eq. (2.16), is only inherited if the full Green's function is reciprocal.

The two spatial variables in the Green's function refer to a source point, $\mathbf{r}'$, where the atom or molecule (electromagnetic source term) is situated and an observation point, $\mathbf{r}$. If the observation point

is not in the same medium as the source position, i.e. across the boundary between media, there is no bulk contribution to the Green's function in this medium. This can be expressed by noting that the bulk part solves the inhomogeneous equation

$$\int \mathrm{d}^3 s \boldsymbol{H}_i(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}^{(0)}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{\delta}(\mathbf{r}-\mathbf{r}'), \tag{E.5}$$

only when $\mathbf{r}$ and $\mathbf{r}'$ are in the volume, V_i, otherwise $\boldsymbol{G}^{(0)}(\mathbf{s},\mathbf{r}',\omega) = \mathbf{0}$. The scattering part of the Green's function solves the homogeneous equation

$$\int \mathrm{d}^3 s \boldsymbol{H}_i(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}^{(1)}(\mathbf{s},\mathbf{r}',\omega) = \mathbf{0}, \tag{E.6}$$

for $\mathbf{r},\mathbf{r}' \in V_i$ and

$$\int \mathrm{d}^3 s \boldsymbol{H}_j(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}^{(1)}(\mathbf{s},\mathbf{r}',\omega) = \mathbf{0}, \tag{E.7}$$

for $\mathbf{r}' \in V_i$ and $\mathbf{r} \in V_j$ where $i \neq j$. As per the procedure in [25], the scattering part of the Green's function can be written as

$$\boldsymbol{G}^{(1)}(\mathbf{r},\mathbf{r}',\omega) = \sum_\lambda \mathbf{a}_\lambda(\mathbf{r})\mathbf{c}_\lambda(\mathbf{r}'), \tag{E.8}$$

this means that the terms $\mathbf{a}_\lambda(\mathbf{r})$ need to solve

$$\int \mathrm{d}^3 s \boldsymbol{H}(\mathbf{r},\mathbf{s},\omega)\cdot\mathbf{a}_\lambda(\mathbf{s}) = \mathbf{0}. \tag{E.9}$$

Using an isotropic magnetodielectric medium as an example, the Helmholtz matrix operator is

$$\boldsymbol{H} = \nabla\times\nabla\times\boldsymbol{I} - \frac{\omega^2}{c^2}\varepsilon(\omega)\mu(\omega)\boldsymbol{I}. \tag{E.10}$$

which is

$$\boldsymbol{H} = -\mathbf{k}\times\mathbf{k}\times\boldsymbol{I} - \frac{\omega^2}{c^2}\varepsilon(\omega)\mu(\omega)\boldsymbol{I}, \tag{E.11}$$

in k-space. The dispersion relation is obtained by solving Booker's equation for k [115],

$$\mathrm{Det}[\boldsymbol{H}(k)] = 0, \tag{E.12}$$

which in the case of an isotropic magnetodielectric is

$$k = \frac{\omega}{c}\sqrt{\varepsilon(\omega)\mu(\omega)}. \tag{E.13}$$

This can be compared to the dispersion relation in free space

$$k = \frac{\omega}{c}, \tag{E.14}$$

which is found by solving Eq. (E.12) for

$$\boldsymbol{H} = \nabla\times\nabla\times\boldsymbol{I} - \frac{\omega^2}{c^2}\boldsymbol{I}. \tag{E.15}$$

The solutions to Eq. (E.9) are well known and are usually referred to as the s and p polarisation vectors of plane waves. In this thesis a slightly different form, given in Ref. [116], will be used. The polarisation

vectors can be found by considering a vector wave equation of the form

$$\nabla\times\nabla\times\mathbf{F}(\mathbf{r}) - k^2\mathbf{F}(\mathbf{r}) = \mathbf{0}. \tag{E.16}$$

If the scalar potential, $\psi(\mathbf{r})$, solves the scalar Helmholtz equation

$$(\nabla^2 + k^2)\psi(\mathbf{r}) = \mathbf{0}, \tag{E.17}$$

then a solution of the form

$$\mathbf{M}(\mathbf{k},\mathbf{r}) = \frac{1}{k_s}\nabla\times\mathbf{c}\psi(\mathbf{r}), \tag{E.18}$$

solves Eq. (E.16), where $\mathbf{c}$ is an arbitrary pilot vector. A futher solution of the form

$$\mathbf{N}(\mathbf{k},\mathbf{r}) = \frac{1}{k}\nabla\times\mathbf{M}(\mathbf{k},\mathbf{r}), \tag{E.19}$$

is also found. To derive the polarisation vectors to be used in this thesis, the solution to the scalar Helmholtz equation, $\psi(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}$, and choice of the unit vector in the z-direction as the pilot vector, $\mathbf{c} = \hat{\mathbf{z}}$, are used to construct

$$\mathbf{M}(\mathbf{k},\mathbf{r}) = \frac{1}{k_s}\nabla\times\hat{\mathbf{z}}e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{E.20a}$$

$$\mathbf{N}(\mathbf{k},\mathbf{r}) = \frac{1}{kk_s}\nabla\times\nabla\times\hat{\mathbf{z}}e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{E.20b}$$

where $k_s^2 = k_x^2 + k_y^2$. It should be noted that although these polarisation vectors are sufficient for the construction of a scattering Green's function in many cases, in some bulk media the curl - free polarisation vector is needed. This is defined as

$$\mathbf{L}(\mathbf{k},\mathbf{r}) = \nabla\psi(\mathbf{r}), \tag{E.21}$$

and, with $\mathbf{M}(\mathbf{k},\mathbf{r})$ and $\mathbf{N}(\mathbf{k},\mathbf{r})$, it completes an orthogonal set of polarisation vectors.

An incident plane wave will not change polarisation when it reflects off the boundary of an isotropic magnetodielectric medium. This means that an incident wave with an $\mathbf{M}$ polarisation is reflected and transmitted into plane waves which are also $\mathbf{M}$ polarised, the same is true for $\mathbf{N}$ polarised incident waves. Consider a source point, $\mathbf{r}'$, in free space (denoted by a subscript 1) near the boundary with an isotropic magnetodielectric halfspace (denoted by a subscript 2), the boundary is the $(x-y)$-plane at $z = 0$ and the medium fills out all space where $z < 0$. The scattering part of the Green's function when the observation point is in the same halfspace as the electromagnetic source, region 1, is dependent on the reflection coefficients, R_{MM} and R_{NN} and the scattering Green's function is [25]

$$\begin{aligned}\boldsymbol{G}_1^{(1)}(\mathbf{r},\mathbf{r}',\omega) = \frac{i}{8\pi^2}\int\frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\big[&R_{MM}\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r})\\ &+ R_{NN}\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})\big],\end{aligned} \tag{E.22}$$

where the definitions $\mathbf{k}_s = (k_x,k_y,0)$ and $\mathrm{d}\mathbf{k}_s = \mathrm{d}k_x\mathrm{d}k_y$ have been used. If the observation point is instead in the magnetodielectric medium, the scattering Green's function for medium 2 is

$$\begin{aligned}\boldsymbol{G}_2^{(1)}(\mathbf{r},\mathbf{r}',\omega) = \frac{i}{8\pi^2}\int\frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\big[&t_{MM}\mathbf{M}(\mathbf{k}_s,-k_{z2},\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r})\\ &+ t_{NN}\mathbf{N}(\mathbf{k}_s,-k_{z2},\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})\big],\end{aligned} \tag{E.23}$$

where t_{MM} and t_{NN} are transmission coefficients. The reflection and transmission coefficients can be

obtained by the boundary conditions [25]

$$\hat{\mathbf{e}}_{\parallel}\cdot\Big(\boldsymbol{G}_1(\mathbf{r}_1,\mathbf{r}',\omega)-\boldsymbol{G}_2(\mathbf{r}_2,\mathbf{r}',\omega)\Big)=0, \tag{E.24a}$$

$$\hat{\mathbf{e}}_{\parallel}\cdot\left(\frac{1}{\mu_1(\omega)}\nabla\times\boldsymbol{G}_1(\mathbf{r}_1,\mathbf{r}',\omega)-\frac{1}{\mu_2(\omega)}\nabla\times\boldsymbol{G}_2(\mathbf{r}_2,\mathbf{r}',\omega)\right)=0, \tag{E.24b}$$

which apply when there is no surface current. The term $\hat{\mathbf{e}}_{\parallel}$ refers to two unit vectors that are perpendicular to each other, and to the normal of the boundary surface. The bulk part of the Green's function in free space, the region with the electromagnetic source, is required to complete the total Green's function in region 1, $\boldsymbol{G}_1(\mathbf{r}_1,\mathbf{r}',\omega)$. This is a well known result [116]

$$\boldsymbol{G}_1^{(0)}(\mathbf{r},\mathbf{r}',\omega)=\frac{i}{8\pi^2}\int\frac{\mathrm{d}\mathbf{k}_s}{k_{z1}}\Big[\mathbf{M}(\mathbf{k}_s,\pm k_z,\mathbf{r})\otimes\mathbf{M}(-\mathbf{k}_s,\mp k_z,\mathbf{r}')+\mathbf{N}(\mathbf{k}_s,\pm k_z,\mathbf{r})\otimes\mathbf{N}(-\mathbf{k}_s,\mp k_z,\mathbf{r}'), \tag{E.25}$$

where $\pm$ for $z\gtrless z'$. Setting

$$\hat{\mathbf{e}}_{\parallel}=\hat{\mathbf{e}}_{\mathbf{k}_s},\,\hat{\mathbf{e}}_M, \tag{E.26a}$$

$$\hat{\mathbf{e}}_{\mathbf{k}_s}=\frac{1}{k_s}(k_x,k_y,0), \tag{E.26b}$$

$$\hat{\mathbf{e}}_M=\frac{1}{k_s}(k_y,-k_x,0), \tag{E.26c}$$

and solving for the reflection and transmission coefficients leads to

$$R_{MM}=\frac{\mu_2(\omega)k_{z1}-k_{z2}}{\mu_2(\omega)k_{z1}+k_{z2}}, \tag{E.27a}$$

$$R_{NN}=\frac{\varepsilon_2(\omega)k_{z1}-k_{z2}}{\varepsilon_2(\omega)k_{z1}+k_{z2}}, \tag{E.27b}$$

$$t_{MM}=\frac{2\mu_2(\omega)k_{z1}}{\mu_2(\omega)k_{z1}+k_{z2}}, \tag{E.27c}$$

$$t_{NN}=\frac{k_2}{k_1}\frac{\varepsilon_2(\omega)k_{z1}}{\varepsilon_2(\omega)k_{z1}+k_{z2}}, \tag{E.27d}$$

which are the Fresnel reflection and transmission coefficients. The scattering part of the Green's function in free space, region 1, and the isotropic magnetodielectric medium, region 2, can now be fully expressed.

E.2 Dyadic Properties of the Green's Function

To calculate the dispersion forces discussed in this thesis, the trace of the Green's function is needed. In the dyadic construction of the Green's function this involves taking the scalar product of the dyads, which are the polarisation vectors. These are

$$\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\cdot\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r})=e^{i\mathbf{k}_s\cdot(\mathbf{r}-\mathbf{r}')+k_{z1}(z+z')}, \tag{E.28a}$$

$$\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\cdot\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})=\left(1-\frac{2k_{z1}^2}{k_1^2}\right)e^{i\mathbf{k}_s\cdot(\mathbf{r}-\mathbf{r}')+k_{z1}(z+z')}, \tag{E.28b}$$

$$\mathbf{M}(\mathbf{k}_s,k_{z1},\mathbf{r})\cdot\mathbf{N}(-\mathbf{k}_s,k_{z1},\mathbf{r})=\mathbf{N}(\mathbf{k}_s,k_{z1},\mathbf{r})\cdot\mathbf{M}(-\mathbf{k}_s,k_{z1},\mathbf{r})=\mathbf{0}. \tag{E.28c}$$

The vector product of dyads is known from dyadic algebra [117]

$$\mathbf{u}\times\mathbf{a}\otimes\mathbf{b}=(\mathbf{u}\times\mathbf{a})\otimes\mathbf{b}, \tag{E.29}$$

where $\mathbf{a}\otimes\mathbf{b}$ is a dyadic product and subsequently $(\mathbf{u}\times\mathbf{a})\otimes\mathbf{b}$ is also a dyadic product, this allows the curl of the Green's function to be calculated. In terms of the polarisation vectors defined above the vector products are

$$\nabla\times\mathbf{M}(\mathbf{k}_s, k_z, \mathbf{r}) = k\mathbf{N}(\mathbf{k}_s, k_z, \mathbf{r}), \tag{E.30a}$$

$$\nabla\times\mathbf{N}(\mathbf{k}_s, k_z, \mathbf{r}) = k\mathbf{M}(\mathbf{k}_s, k_z, \mathbf{r}). \tag{E.30b}$$

The transpose of a dyadic product results in the dyads switching position, i.e., taking the transpose of the dyadic product $\mathbf{a}\otimes\mathbf{b}$ is

$$(\mathbf{a}\otimes\mathbf{b})^{\mathrm{T}} = \mathbf{b}\otimes\mathbf{a}. \tag{E.31}$$

It can be convenient to change the integration variables to a form that allows analytical solutions. The integral over $\mathrm{d}\mathbf{k}_s$ can be changed to

$$\int \mathrm{d}\mathbf{k}_s \to \int_0^{2\pi} \mathrm{d}\theta \int_0^{\infty} \mathrm{d}k_s k_s \to -2\pi \int_{\frac{\omega}{c}}^{\infty} \mathrm{d}k_z k_z \to 2\pi \int_{\frac{\xi}{c}}^{\infty} \mathrm{d}\tilde{k}_z \tilde{k}_z, \tag{E.32}$$

and in complex frequency ($\omega \to i\xi$) the wave vectors become

$$k_1 = i\frac{\xi}{c} = i\tilde{k}_1, \quad k_{1z} = i\tilde{k}_{1z}, \tag{E.33}$$

with

$$\tilde{k}_{1z} = \sqrt{\left(\frac{\xi}{c}\right)^2 + k_s^2}, \tag{E.34}$$

in free space and

$$k_2 = i\left(\frac{\xi}{c}\right)\left(\sqrt{\varepsilon_2(i\xi)\mu_2(i\xi)}\right) = i\tilde{k}_2, \quad k_{z2} = i\sqrt{\left(\tilde{k}_2\right) + k_s^2} = i\tilde{k}_{z2}, \tag{E.35}$$

in the magnetodielectric halfspace.

Appendix F

Transformation of Electromagnetic Fields in Moving Media

In this appendix the transformation of the electromagnetic fields propagating through a medium in motion with respect to an arbitrary laboratory frame will be shown. The rest frame of the medium is the inertial reference frame in which the medium appears stationary. In all other inertial frames the medium will appear to be in motion, which will affect the observed electromagnetic field. Although the observed values differ between inertial reference frames it should be noted that the laws of physics are the same and the speed of light, c, is a constant in every inertial reference frame.

In order to transform the electromagnetic field from one inertial reference frame to an arbitrary inertial reference frame, some new notation needs to be introduced. Fourvectors are a mathematical construct that combine the space like and the time like properties of quantities. For example, the position vector $\mathbf{r} = (x, y, z)$ and the time component, t, are combined into the event fourvector, which is defined as

$$x^{\mu} = [ct, x, y, z], \quad x_{\mu} = [ct, -x, -y, -z], \tag{F.1}$$

where the free index runs over $\mu = 0, 1, 2, 3$. The two forms of the event fourvector, labelled as contravariant and covariant respectively, are connected by a metric, $g^{\mu\nu}$

$$x^{\mu} = g^{\mu\nu} x_{\nu}, \tag{F.2}$$

where a repeated index implies a summation. For the purposes of this thesis, a useful fourvector is the wave fourvector

$$k^{\mu} = \left[\frac{\omega}{c}, k_x, k_y, k_z\right]. \tag{F.3}$$

In the same notation, the electric and magnetic induction fields comprise the Faraday tensor, $F^{\mu\nu}$,

$$F^{\mu\nu} = \begin{pmatrix} 0 & -\frac{E_x}{c} & -\frac{E_y}{c} & -\frac{E_z}{c} \\ \frac{E_x}{c} & 0 & -B_z & B_y \\ \frac{E_y}{c} & B_z & 0 & -B_x \\ \frac{E_z}{c} & -B_y & B_x & 0 \end{pmatrix}, \tag{F.4}$$

and tensors that comprises of the medium assisted fields and the noise current components can be

constructed

$$D^{\mu\nu} = \begin{pmatrix} 0 & -cD_x & -cD_y & -cD_z \\ cD_x & 0 & -H_z & H_y \\ cD_y & H_z & 0 & -H_x \\ cD_z & -H_y & H_x & 0 \end{pmatrix}, \tag{F.5a}$$

$$M_{\mathrm{N}}^{\mu\nu} = \begin{pmatrix} 0 & -cP_{\mathrm{N}x} & -cP_{\mathrm{N}y} & -cP_{\mathrm{N}z} \\ cP_{\mathrm{N}x} & 0 & M_{\mathrm{N}z} & -M_{\mathrm{N}y} \\ cP_{\mathrm{N}y} & -M_{\mathrm{N}z} & 0 & M_{\mathrm{N}x} \\ cP_{\mathrm{N}z} & M_{\mathrm{N}y} & -M_{\mathrm{N}x} & 0 \end{pmatrix}. \tag{F.5b}$$

The effect of motion on these fields can be found by applying Lorentz transformations. These provide a connection between quantities as observed in an inertial reference frame and the same quantities as observed from the perspective of an arbitrary reference frame moving at velocity $\mathbf{v}$ with respect to the first reference frame. These can be written in tensor form, Λ^{μ}_{ν}, where each element is defined as $\Lambda^{\mu}_{\nu} = \frac{\partial x^{\mu}}{\partial x^{\nu}}$ [34]. In a Lorentz transformation it is assumed that the origins of the two inertial reference frames, S and S', coincide at $t = t' = 0$. The Lorentz tensor, Λ^{μ}_{ν}, is the inverse of the Lorentz tensor Λ^{ν}_{μ} and therefore

$$\Lambda^{\mu}_{\nu}\Lambda^{\nu}_{\eta} = \delta^{\mu}_{\eta}, \tag{F.6}$$

where

$$\delta^{\mu}_{\eta} = \begin{cases} 1, & \text{if } \mu = \eta \\ 0, & \text{otherwise.} \end{cases} \tag{F.7}$$

To Lorentz transform a quantity such as a fourvector or tensor, each index needs to be transformed, for example, an event fourvector transforms as

$$x^{\mu'} = \Lambda^{\mu'}_{\mu} x^{\mu}, \tag{F.8}$$

whereas a tensor, such as the Faraday tensor, transforms as

$$F^{\mu'\nu'} = \Lambda^{\mu'}_{\mu}\Lambda^{\nu'}_{\nu} F^{\mu\nu}. \tag{F.9}$$

There are some quantities that are called invariant because they do no change after Lorentz transformations have been applied. An example is the space-time interval of an event, defined as

$$s^2 = x^{\mu}x_{\mu} = (ct)^2 - x^2 - y^2 - z^2. \tag{F.10}$$

As can be seen, applying the Lorentz transformations

$$s'^2 = x^{\mu'}x_{\mu'} = \Lambda^{\mu'}_{\eta} x^{\eta}\Lambda^{\sigma}_{\mu'}x_{\sigma} = \Lambda^{\mu'}_{\eta}\Lambda^{\sigma}_{\mu'}x^{\eta}x_{\sigma} = x^{\eta}x_{\eta} = s^2, \tag{F.11}$$

does not change the quantity.

Applying the Lorentz transformation to the event fourvector and the wavevector fourvector and writing in coordinate space leads to

$$\left[ct', \mathbf{r}'\right] = \left[c(\gamma(t - \frac{\mathbf{r}\cdot\mathbf{v}}{c^2})), \mathbf{r}_{\perp} + \gamma(\mathbf{r}_{\parallel} - \mathbf{v}t)\right], \tag{F.12a}$$

$$\left[\frac{\omega'}{c}, \mathbf{k}'\right] = \left[\frac{(\gamma(\omega - \mathbf{k}\cdot\mathbf{v}))}{c}, \mathbf{k}_{\perp} + \gamma(\mathbf{k}_{\parallel} - \frac{\mathbf{v}\omega}{c^2})\right], \tag{F.12b}$$

where the subscripts $\perp$ and $\parallel$ refer to the components that are perpendicular and parallel to the direction of the velocity, respectively. The superscript primes, $'$, denote quantities observed in the inertial reference frame S'. The electromagnetic fields can now be Lorentz transformed. These can be written in tensor notation, but the vector notation shown in Ref. [117] is prefered. The transformed fields are

$$\mathbf{E}' = \mathbf{E}_\parallel + \gamma(\mathbf{E}_\perp + \mathbf{v}\times\mathbf{B}), \tag{F.13a}$$

$$\mathbf{B}' = \mathbf{B}_\parallel + \gamma(\mathbf{B}_\perp - \frac{1}{c^2}\mathbf{v}\times\mathbf{E}), \tag{F.13b}$$

$$\mathbf{H}' = \mathbf{H}_\parallel + \gamma(\mathbf{H}_\perp - \mathbf{v}\times\mathbf{D}), \tag{F.13c}$$

$$\mathbf{D}' = \mathbf{D}_\parallel + \gamma(\mathbf{D}_\perp + \frac{1}{c^2}\mathbf{v}\times\mathbf{H}). \tag{F.13d}$$

The noise current components transform as

$$\mathbf{P}'_\mathrm{N} = \mathbf{P}_{\mathrm{N}\parallel} + \gamma(\mathbf{P}_{\mathrm{N}\perp} - \frac{\mathbf{v}\times\mathbf{M}_\mathrm{N}}{c^2}), \tag{F.14a}$$

$$\mathbf{M}'_\mathrm{N} = \mathbf{M}_{\mathrm{N}\parallel} + \gamma(\mathbf{M}_{\mathrm{N}\perp} + \mathbf{v}\times\mathbf{P}_\mathrm{N}). \tag{F.14b}$$

The term, γ, is the Lorentz factor and is defined as

$$\gamma = \frac{1}{\sqrt{1-\frac{v^2}{c^2}}}. \tag{F.15}$$

A fundamental feature of special relativity is that the laws of physics, including Maxwell's equations, are the same in every inertial reference frame. This can be used to obtain the constitutive relations for the moving medium from the perspective of an arbitrary reference frame.

Consider a medium that is a locally responding isotropic magnetoelectric in its rest frame. The constitutive relations are

$$\mathbf{D}' = \varepsilon_0\boldsymbol{\varepsilon}'\cdot\mathbf{E}' + \mathbf{P}'_\mathrm{N}, \tag{F.16a}$$

$$\mathbf{B}' = \mu_0\boldsymbol{\mu}'\cdot(\mathbf{H}' + \mathbf{M}'_\mathrm{N}), \tag{F.16b}$$

where $\boldsymbol{\varepsilon}' = \varepsilon'(\omega')\boldsymbol{I}$ and $\boldsymbol{\mu}' = \mu'(\omega')\boldsymbol{I}$.

A laboratory frame is defined such that the medium is moving at constant velocity $\mathbf{v}$ with respect to an observer at rest in the laboratory frame. The constitutive relations of the medium, as viewed from the laboratory frame will not appear as Eqs. (F.16a) and (F.16b).

The constitutive relations can be found by substituting Eqs. (F.13a), (F.13b), (F.13c), (F.13d), (F.14a) and (F.14b) into Eqs. (F.16a) and (F.16b) and expressing every perpendicular field in terms of the full field and the parallel field, i.e., $\mathbf{F}_\perp = \mathbf{F} - \mathbf{F}_\parallel$. This leads to

$$\mathbf{D}_\parallel + \gamma(\mathbf{D} - \mathbf{D}_\parallel + \frac{1}{c^2}\mathbf{v}\times\mathbf{H}) = \varepsilon_0\varepsilon'\mathbf{E}_\parallel + \gamma(\mathbf{E} - \mathbf{E}_\parallel + \mathbf{v}\times\mathbf{B}) + \mathbf{P}_{\mathrm{N}\parallel} + \gamma(\mathbf{P}_\mathrm{N} - \mathbf{P}_{\mathrm{N}\parallel} - \frac{\mathbf{v}\times\mathbf{M}_\mathrm{N}}{c^2}), \tag{F.17a}$$

$$\mathbf{B}_\parallel + \gamma(\mathbf{B} - \mathbf{B}_\parallel - \frac{1}{c^2}\mathbf{v}\times\mathbf{E}) = \mu_0\mu'(\mathbf{H}_\parallel + \gamma(\mathbf{H} - \mathbf{H}_\parallel - \mathbf{v}\times\mathbf{D}) + \mathbf{M}_{\mathrm{N}\parallel} + \gamma(\mathbf{M}_\mathrm{N} - \mathbf{M}_{\mathrm{N}\parallel} + \mathbf{v}\times\mathbf{P}_\mathrm{N})). \tag{F.17b}$$

Taking the vector product of Eqs. (F.17a) and (F.17a) with respect to velocity gives

$$\mathbf{v}\times\mathbf{D} = \varepsilon_0\varepsilon'(\mathbf{v}\times\mathbf{E} + \mathbf{v}\times\mathbf{v}\times\mathbf{B}) + \mathbf{v}\times\mathbf{P}_\mathrm{N} - \frac{\mathbf{v}\times\mathbf{v}\times\mathbf{M}_\mathrm{N}}{c^2} - \frac{\mathbf{v}\times\mathbf{v}\times\mathbf{H}}{c^2}, \tag{F.18a}$$

$$\mathbf{v}\times\mathbf{B} = \mu_0\mu'(\mathbf{v}\times\mathbf{H} - \mathbf{v}\times\mathbf{v}\times\mathbf{D} + \mathbf{v}\times\mathbf{M}_\mathrm{N} + \mathbf{v}\times\mathbf{v}\times\mathbf{P}_\mathrm{N}) + \frac{\mathbf{v}\times\mathbf{v}\times\mathbf{E}}{c^2}. \tag{F.18b}$$

Placing Eq. (F.18a) into Eq. (F.17b) and Eq. (F.18b) into Eq. (F.17a) and making use of the identity

$$\mathbf{v}\times\mathbf{v}\times\boldsymbol{I} = \mathbf{v}\mathbf{v} - v^2\boldsymbol{I}, \tag{F.19}$$

leads to

$$\begin{aligned}\frac{\gamma}{\gamma'^2}\mathbf{D} + (1-\frac{\gamma}{\gamma'^2})\mathbf{D}_\parallel = \varepsilon_0\varepsilon'(\frac{1}{\gamma}\mathbf{E} + (\frac{\gamma}{\gamma'^2}-\frac{1}{\gamma})\mathbf{E}_\parallel) + \varepsilon_0\varepsilon'(1-\frac{\gamma}{\gamma'^2})\mathbf{E}_\parallel + \frac{\gamma}{\gamma'^2}\mathbf{P}_\mathrm{N} + (1-\frac{\gamma}{\gamma'^2})\mathbf{P}_{\mathrm{N}\parallel} \\ + \frac{\gamma}{c^2}(\varepsilon'\mu'-1)[\mathbf{v}\times\mathbf{H} + \mathbf{v}\times\mathbf{M}_\mathrm{N}],\end{aligned} \tag{F.20a}$$

$$\begin{aligned}\frac{\gamma}{\gamma'^2}\mathbf{B} + (1-\frac{\gamma}{\gamma'^2})\mathbf{B}_\parallel = \mu_0\mu'(\frac{1}{\gamma}\mathbf{H} + (\frac{\gamma}{\gamma'^2}-\frac{1}{\gamma})\mathbf{H}_\parallel) + \mu_0\mu'(1-\frac{\gamma}{\gamma'^2})\mathbf{H}_\parallel + \mu_0\mu'(\frac{1}{\gamma}\mathbf{H} + (\frac{\gamma}{\gamma'^2}-\frac{1}{\gamma})\mathbf{H}_\parallel) \\ + \mu_0\mu'(1-\frac{\gamma}{\gamma'^2})\mathbf{M}_{\mathrm{N}\parallel} + \frac{\gamma}{c^2}(1-\varepsilon'\mu')\mathbf{v}\times\mathbf{E},\end{aligned} \tag{F.20b}$$

where the definitions

$$\gamma' = \frac{1}{\sqrt{1-\frac{v^2}{(c')^2}}}, \quad (c')^2 = \frac{1}{\mu'\varepsilon'\mu_0\epsilon_0}, \tag{F.21}$$

have been used. As can be seen from the properties of the fields as they transform, if Eqs. (6.2a) and (6.2b) hold, then

$$\mathbf{D}_\parallel = \varepsilon_0\boldsymbol{\varepsilon}'\cdot\mathbf{E}_\parallel + \mathbf{P}_{\mathrm{N}_\parallel}, \tag{F.22a}$$

$$\mathbf{B}_\parallel = \mu_0\boldsymbol{\mu}'\cdot(\mathbf{H}_\parallel + \mathbf{M}_{\mathrm{N}_\parallel}), \tag{F.22b}$$

which means these terms can be removed from the respective equations above to give the transformed constitutive relations as

$$\mathbf{D} = \varepsilon_0\varepsilon'(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1-\frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}})\cdot\mathbf{E} + \mathbf{P}_\mathrm{N} + \frac{\gamma'^2}{c^2}(\varepsilon'\mu'-1)\mathbf{v}\times\mathbf{H} + \frac{\gamma'^2}{c^2}(\varepsilon'\mu'-1)\mathbf{v}\times\mathbf{M}_\mathrm{N}, \tag{F.23a}$$

$$\mathbf{B} = \mu_0\mu'(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1-\frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}})\cdot\mathbf{H} + \frac{\gamma'^2}{c^2}(1-\varepsilon'\mu')\mathbf{v}\times\mathbf{E} + \mu_0\mu'(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1-\frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}})\cdot\mathbf{M}_\mathrm{N}. \tag{F.23b}$$

For simplicity these can now be written as

$$\mathbf{D} = \varepsilon_0\boldsymbol{\varepsilon}\cdot\mathbf{E} + \mathbf{P}_\mathrm{N} + \frac{1}{c}\boldsymbol{\xi}\cdot\mathbf{H} + \frac{1}{c}\boldsymbol{\xi}\cdot\mathbf{M}_\mathrm{N}, \tag{F.24a}$$

$$\mathbf{B} = \mu_0\boldsymbol{\mu}\cdot\mathbf{H} + \frac{1}{c}\boldsymbol{\zeta}\cdot\mathbf{E} + \mu_0\boldsymbol{\mu}\cdot\mathbf{M}_\mathrm{N}, \tag{F.24b}$$

where

$$\boldsymbol{\varepsilon} = \varepsilon'(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1-\frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}}), \tag{F.25a}$$

$$\boldsymbol{\zeta} = \frac{\gamma'^2}{c}(1-\varepsilon'\mu')\mathbf{v}\times\boldsymbol{I}, \tag{F.25b}$$

$$\boldsymbol{\xi} = \frac{\gamma'^2}{c}(\varepsilon'\mu'-1)\mathbf{v}\times\boldsymbol{I}, \tag{F.25c}$$

$$\boldsymbol{\mu} = \mu'(\frac{\gamma'^2}{\gamma^2}\boldsymbol{I} + (1-\frac{\gamma'^2}{\gamma^2})\hat{\mathbf{v}}\hat{\mathbf{v}}). \tag{F.25d}$$

Appendix G

Bianisotropic Boundary Conditions

The reflection and transmission from the boundary of bianisotropic media differs from the reflection and transmission off anisotropic media. This is due to the magnetoelectric terms, which partially rotate the polarisation of the incident wave in the reflected and transmitted wave and therefore generates additional terms. A well known example of this is the reflection off a chiral medium when an incident linearly polarised wave is reflected as a circularly polarised wave.

This means that boundary conditions which apply to bianisotropic media are required.

G.1 Boundary Conditions

An arbitrary point, $\mathbf{r}$, is on the boundary between two bianisotropic media, labelled as i and j respectively, the boundary can be considered as a plane in a small neighbourhood around $\mathbf{r}$. The normal vector of this plane, $\hat{\mathbf{e}}_\perp$, is directed into region i.

The first boundary condition is found by considering a rectangle, A, of height h in the $\hat{\mathbf{e}}_\perp$ direction and a length l in a parallel $\hat{\mathbf{e}}_\parallel$, centred on the point $\mathbf{r}$ in the interface plane, where $h \ll l$. The term $\nabla \times \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega)$ is then integrated around the rectangle

$$\int_A \mathrm{d}\mathbf{A} \cdot \nabla \times \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) = \oint_{\partial \mathbf{A}} \mathrm{d}\mathbf{l} \cdot \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega), \tag{G.1}$$

where Stokes theorem has been used. In the limit of $h \to 0$, $A \to 0$ and so the left hand side of Eq. (G.1) tends to zero and the term on the right hand side becomes

$$l\hat{\mathbf{e}}_\parallel \cdot \Big[\boldsymbol{G}(\mathbf{r}_i, \mathbf{r}', \omega) - \boldsymbol{G}(\mathbf{r}_j, \mathbf{r}', \omega) \Big], \tag{G.2}$$

where $\mathbf{r}_{i/j} = \lim_{h \to 0} \mathbf{r} \pm h\hat{\mathbf{e}}_\perp$. This leads to a condition on the tangential component of the Green's function at the boundary, it needs to be continuous and so the first boundary condition is

$$\hat{\mathbf{e}}_\parallel \cdot \Big[\boldsymbol{G}_i - \boldsymbol{G}_j \Big] = \mathbf{0}, \tag{G.3}$$

where the definition $\boldsymbol{G}_i = \boldsymbol{G}(\mathbf{r}_i, \mathbf{r}', \omega)$ has been introduced. For the next boundary condition the starting point is the wave equation for a general linear bianisotropic medium, Eq. (4.13) in section 4.1. This

wave equation is integrated over the rectangle, A, to obtain

$$\begin{aligned}&\int_A \mathrm{d}\mathbf{A}\cdot\nabla\times\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G} - \frac{i\omega}{c}\int_A \mathrm{d}\mathbf{A}\cdot\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\boldsymbol{G}\\ &= \int_A \mathrm{d}\mathbf{A}\cdot\boldsymbol{\delta} + \frac{\omega^2}{c^2}\int_A \mathrm{d}\mathbf{A}\cdot(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}) - \frac{i\omega}{c}\int_A \mathrm{d}\mathbf{A}\cdot\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G}.\end{aligned} \tag{G.4}$$

The terms on the left hand side of Eq. (G.4) are rewritten using Stoke's theorem

$$\int_A \mathrm{d}\mathbf{A}\cdot\nabla\times\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G} = \oint_{\partial\mathbf{A}} \mathrm{d}\mathbf{l}\cdot\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G} \simeq l\hat{\mathbf{e}}_\parallel\cdot[\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j], \tag{G.5a}$$

$$\int_A \mathrm{d}\mathbf{A}\cdot\nabla\times\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\boldsymbol{G} = \oint_{\partial\mathbf{A}} \mathrm{d}\mathbf{l}\cdot\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta}\star\boldsymbol{G} \simeq l\hat{\mathbf{e}}_\parallel\cdot[\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i\star\boldsymbol{G}_i - \boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j\star\boldsymbol{G}_j]. \tag{G.5b}$$

In the limit $h\to 0$ the area disappears and the $\frac{\omega^2}{c^2}\int_A \mathrm{d}\mathbf{A}\cdot(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})$ and $\frac{i\omega}{c}\int_A \mathrm{d}\mathbf{A}\cdot\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G}$ terms in Eq. (G.4) are zero. This leads to

$$l\hat{\mathbf{e}}_\parallel\cdot\left([\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j] - \frac{i\omega}{c}[\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i\star\boldsymbol{G}_i - \boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j\star\boldsymbol{G}_j]\right) = \lim_{h\to 0}\int_A \mathrm{d}\mathbf{A}\cdot\boldsymbol{\delta}. \tag{G.6}$$

Dividing this by l and rearranging leads to the second boundary condition

$$\hat{\mathbf{e}}_\parallel\cdot\left([\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \frac{i\omega}{c}\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i\star\boldsymbol{G}_i] - [\boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j - \frac{i\omega}{c}\boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j\star\boldsymbol{G}_j]\right) = \lim_{h\to 0}\int \mathrm{d}\mathbf{h}\cdot\boldsymbol{\delta}. \tag{G.7}$$

The right hand side is zero unless the source and observation points are the same. For the next two boundary conditions, a pillbox, instead of a rectangle is considered. The pillbox is centred on $\mathbf{r}$, with a height h, and areas A that have normals perpendicular and anti-perpendicular to the interface plane. Integrating the divergence of $\nabla\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ over the volume of the pillbox

$$\begin{aligned}\int_V \mathrm{d}V\nabla\cdot\nabla\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) &= \int_{\partial V}\mathrm{d}\mathbf{A}\cdot\nabla\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\\ &\simeq A\hat{\mathbf{e}}_\perp\cdot\Big[\nabla\times\boldsymbol{G}(\mathbf{r}_i,\mathbf{r}',\omega) - \nabla\times\boldsymbol{G}(\mathbf{r}_j,\mathbf{r}',\omega)\Big].\end{aligned} \tag{G.8}$$

Taking the limit $h\to 0$ leads to

$$\hat{\mathbf{e}}_\perp\cdot[\nabla\times\boldsymbol{G}_i - \nabla\times\boldsymbol{G}_j] = \mathbf{0}, \tag{G.9}$$

the third boundary condition. The derivation of the fourth condition starts by taking the divergence of Eq. (4.13),

$$\frac{i\omega}{c}\nabla\cdot\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G} - \frac{\omega^2}{c^2}\nabla\cdot(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\star\boldsymbol{G} = \nabla\cdot\boldsymbol{\delta}. \tag{G.10}$$

Integrating Eq. (G.10) over the volume of the pillbox

$$\frac{i\omega}{c}\int_V \mathrm{d}V\nabla\cdot\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G} - \frac{\omega^2}{c^2}\int_V \mathrm{d}V\nabla\cdot(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\star\boldsymbol{G} = \int_V \mathrm{d}V\nabla\cdot\boldsymbol{\delta}. \tag{G.11}$$

and applying divergence theorem to the terms on the left hand side of the equation, leads to

$$\begin{aligned}\int_V \mathrm{d}V\nabla\cdot\boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G} &= \oint_A \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\nabla\times\boldsymbol{G}\cdot\hat{\mathbf{n}}\mathrm{d}A\\ &\simeq A\hat{\mathbf{e}}_\perp\cdot[\boldsymbol{\xi}_i\star\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \boldsymbol{\xi}_j\star\boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j],\end{aligned} \tag{G.12a}$$

$$\int_V \mathrm{d}V \nabla\cdot(\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\star\boldsymbol{G} = \oint_A (\boldsymbol{\varepsilon} - \boldsymbol{\xi}\star\boldsymbol{\mu}^{-1}\star\boldsymbol{\zeta})\star\boldsymbol{G}\cdot\hat{\mathbf{n}}\mathrm{d}A \\ \simeq A\hat{\mathbf{e}}_\perp\cdot[(\boldsymbol{\varepsilon}_i - \boldsymbol{\xi}_i\star\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i)\star\boldsymbol{G}_i - (\boldsymbol{\varepsilon}_j - \boldsymbol{\xi}_j\star\boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j)\star\boldsymbol{G}_j]. \tag{G.12b}$$

Substituting these into Eq. (G.11) and dividing by A and taking the limit $h \to 0$ gives

$$\hat{\mathbf{e}}_\perp\cdot\left[\left(\frac{i\omega}{c}\boldsymbol{\xi}_i\star\boldsymbol{\mu}_i^{-1}\star\nabla\times\boldsymbol{G}_i - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}_i - \boldsymbol{\xi}_i\star\boldsymbol{\mu}_i^{-1}\star\boldsymbol{\zeta}_i)\star\boldsymbol{G}_i\right) - \left(\frac{i\omega}{c}\boldsymbol{\xi}_j\star\boldsymbol{\mu}_j^{-1}\star\nabla\times\boldsymbol{G}_j \right. \\ \left. - \frac{\omega^2}{c^2}(\boldsymbol{\varepsilon}_j - \boldsymbol{\xi}_j\star\boldsymbol{\mu}_j^{-1}\star\boldsymbol{\zeta}_j)\star\boldsymbol{G}_j\right)\right] = \lim_{h\to 0}\int_{h'}\mathrm{d}h'\nabla\boldsymbol{\delta}. \tag{G.13}$$

The four boundary conditions can be thought of as equivalent to the conditions that are usually applied to the electromagnetic fields across a boundary. Multiplying Eq. (G.3) by $i\omega\mu_0\mathbf{j}_\mathrm{N}(\mathbf{r}',\omega)$ and integrating over $\mathbf{r}'$ shows that this condition is equivalent to

$$\hat{\mathbf{e}}_\parallel\cdot\Big[\mathbf{E}(\mathbf{r}_1) - \mathbf{E}(\mathbf{r}_2)\Big] = \hat{\mathbf{e}}_\perp\times\Big[\mathbf{E}(\mathbf{r}_1) - \mathbf{E}(\mathbf{r}_2)\Big] = 0. \tag{G.14}$$

Eq. (4.14c) in section 4.1 shows the relationship between the electric Green's function and the magnetic field. Therefore multiplying Eq. (G.7) by $\mathbf{j}_\mathrm{N}(\mathbf{r}',\omega)$ and integrating over $\mathbf{r}'$ leads to

$$\hat{\mathbf{e}}_\parallel\cdot\Big[\mathbf{H}(\mathbf{r}_1) - \mathbf{H}(\mathbf{r}_2)\Big] = \hat{\mathbf{e}}_\perp\times\Big[\mathbf{H}(\mathbf{r}_1) - \mathbf{H}(\mathbf{r}_2)\Big] = 0, \tag{G.15}$$

which is a familiar boundary condition. Eq. (G.9) can be transformed into the boundary condition

$$\hat{\mathbf{e}}_\perp\cdot\Big[\mathbf{B}(\mathbf{r}_1) - \mathbf{B}(\mathbf{r}_2)\Big] = 0, \tag{G.16}$$

by multiplying Eq. (4.14a) by $\mathbf{j}_\mathrm{N}(\mathbf{r}',\omega)$ and integrating over $\mathbf{r}'$. The final boundary condition can be shown to be

$$\hat{\mathbf{e}}_\perp\cdot\Big[\mathbf{D}(\mathbf{r}_1) - \mathbf{D}(\mathbf{r}_2)\Big] = 0, \tag{G.17}$$

when Eq. (4.14b) is multiplied by $\frac{i\omega}{c^2}\mathbf{j}_\mathrm{N}(\mathbf{r}',\omega)$ and integrated over $\mathbf{r}'$.