

Electromagnetic Fields and Forces in Nanostructures

by

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DECLARATION

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Στή γιαγιά τή Συμυρνιά, τή Μαλίλα μου·

πού ἂν κι ἐλαχίστων γραμμάτων ἤξερε πάντα νά διαβάζει τό ἀνείπωτο στά μάτια μου.

Στόν παππού τόν Μανώλη, τόν Μικρασιάτη·

γλεντζέ καί ταξιδευτή, ἕναν μικρό Διόνυσο μέ τό χαμόγελο πάντα στ' ἀχείλι.

Στή μητέρα μου Χρυσούλα,

τῆς Ἰωνίας γέννημα τῆς Κρήτης θρέμμα, τίς χάρες ἀμφοτέρων συμψηφίζοντας· Φίλη καί Μάνα
(κι ὥστόσο, ἀπό κάτω, πάντα ἕνα ἄθωο κοριτσάκι!).

Στόν πατέρα μου Γιάννη,

μ' ἔμαθε ν' ἀγαπῶ τή γῆ μας· τήν εὐθυκρισία τοῦ Παρθενώνα, πού στά χέρια του γινόταν κόσμημα,
ἤθελε νά τήν πῶ στόν πεντοζάλη.

Στό Καρινιό,

γιά ὅλα· γιά τήν ἀγάπη καί τήν ποίηση

γιά τόν τρόπο πού γεύεται τή θάλασσα

καί τή χαρά τοῦ παιδιοῦ στά καθαρά της μάτια.

Στόν ἀδερφό μου Νίκο, στήν Alison, στήν ἀνιψούλα μου *Εμιλυ-Χρυσή.

Στή Λιλή Ζωγράφου,

τήν ἄλλη μου Μάνα.

Abstract

We have developed a general methodology for computing electromagnetic (EM) fields and forces in matter, based on solving the macroscopic Maxwell's equations numerically in real space and adopting the time-averaged Maxwell Stress Tensor formalism. Our approach can be applied to both dielectric and metallic systems characterised by a local frequency-dependent dielectric function, and in principle to any size and geometry. In this study we are particularly interested in calculating forces on nanostructures, induced by a beam of monochromatic light (such as a laser) of frequency ω . These forces are the direct analogue of Van der Waals interactions at a single frequency: the presence of matter scatters the light and alters the EM field, resulting in an energy-change that manifests itself as a force. The motivation behind this particular direction is the facilitation of self-assembly in colloidal systems with the aim of aiding the fabrication of photonic crystals.

In order to understand the main features of light-induced EM forces, as well as to provide a testbed for our numerical methodology, we first solve (analytically and numerically) for two homogeneous systems: a half-space and a slab. We find that in passing from a low- ϵ to a high- ϵ medium, the light beam always attracts the interface (*i.e.* the *surface* force is negative). The implication is that light will generally induce an attraction between the surfaces of two liquids separated by a layer of lower ϵ . For attraction between solids there is a tougher requirement: the *total* force must also be negative. When the EM field is that of a travelling wave the total pressure is positive. In contrast, evanescent waves may cause the total pressure to become attractive (negative). Thus by shining evanescent light in the region between two solid bodies an attraction between them may be induced.

We then study numerically the influence of monochromatic light (a travelling wave) on a crystal of dielectric spheres of GaP, concentrating on total forces induced on each sphere and on the crystal as a whole. We identify three regimes in the response of the system to radiation:

- At large wavelengths the crystal may be approximated by a homogeneous slab with an effective permittivity ϵ_{eff} . The analytical results for reflectance and forces apply.

- At wavelengths comparable to the lattice constant, multiple scattering effects tune in: when ω is inside the photonic band gaps the reflectivity of a thick crystalline slab rises to unity, the beam bounces off the crystal and there is a maximum momentum exchange (and largest forces). Also, a multitude of force orientations results when the Bragg conditions for multiple outgoing waves are met.
- Much more interesting is the regime where the radiation couples to the EM eigenmodes supported by isolated spheres (Mie resonances). These modes are analogous to electronic orbitals and, like their electronic counterparts, can form bonding and anti-bonding interactions between neighbouring spheres. By irradiating the system with light at the bonding frequency an attractive interaction is induced between the spheres. The photo-induced attraction is strong: for a moderate $I_0 \sim 3 \times 10^8 \text{ W/m}^2$ it surpasses all other interactions present (gravitational, thermal and Van der Waals) by 1-2 orders of magnitude. These resonant forces are sensitive to absorption, but, for GaP spheres in water (a common liquid medium for colloids), their effect should still be clearly seen, even for a polydispersion of a few percent. Thus we suggest that by judicious selection of bonding states we can drive a system towards a desired structure, rather than rely on the structure dictated by gravitational and Van der Waals forces. Apart from possible applications in the fabrication of 3D photonic crystals, the resonant mechanism leading to the bonding/anti-bonding effect may contribute to our understanding of novel non-linear phenomena arising due to the application of laser light fields in nanostructures.

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“If we had never been molested by alarms at celestial and atmospheric phenomena, nor by the misgiving that death somehow affects us, nor by the neglect of the proper limits of pains and desires, we should have had no need to study natural sciences.”

Epicurus

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frequently. I am pleased that some of Theocharis's pioneering work, specifically his defence of the true scientific principles[1] has been recognised. I hope that more of his ideas in science, philosophy and politics will soon receive the credit they deserve.

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« Η μόνη δουλειά που δέχτηκα μέ τή θέλησή μου στή ζωή είναι ή έξυπνέτηση τής αλήθειας. " Αν δέν υπάρχει τίποτα γύρω μου νά στηριχτώ, μοῦ μένει ή αλήθεια καί ή πίστη μου σ' αὐτήν.»

Λ. ΖΩΓΡΑΦΟΥ

Παρέστω μόνον ἡ τετραφάρμακος·
ἄφοβον ὁ θεός
ἀναίσθητον ὁ θάνατος
καὶ τᾶγαθὸν μὲν εὖκτητον
τὸ δὲ δεινὸν εὐεκκαρτέρητον.

ΕΠΙΚΟΤΡΟΣ

“Mankind [...] is turning a deaf ear to all the fundamental political questions, and the case for techno-science is a chief political question, because it is the question [...] of the relation between the continuously increasing power of techno-science and the manifest powerlessness of contemporary societies.”

C Castoriades

Chapter 1

Introduction

1.1 Photonics in nanostructures

Over the recent years there has been great interest within the condensed matter physics and optics communities world-wide in the electromagnetic effects in *nanostructures* (materials where structure appears in the scale of a few up to a thousand nm).

The motivation for this lies both at the level of basic and applied research. It stems from the realisation that the electromagnetic wavefield in solids and their surfaces plays an equal and complementary role to the electron wavefield, a role emphasised by recent experimental developments. Inverse photoemission (IPE) observations from Scanning Tunnelling Microscope (STM) tips [2, 3, 4] show detailed structure in the visible region of the spectrum having its origin in electromagnetic resonances between the tip and the surface (whereas in more conventional IPE experiments structure is dominated by surface electronic band structure). The same electromagnetic fields are responsible for forces acting at large distances between an Atomic Force Microscope (AFM) tip and the surface: the electromagnetic field comes into its own in nanoscale structures.

In parallel with developments in surface science have been experiments on *photonic crystals*: bulk nanoscale structures possessing crystalline order at the scale of the wavelength of light which they therefore diffract strongly, just like x-rays are diffracted from atomic crystals. By adjusting the geometric and dielectric parameters of a photonic crystal, we can alter the *optical* properties at will [7]-[11], in the same way as a semiconductor crystal can be built to support

desirable *electronic* properties. For example, the dispersion relation (band structure) of light inside a photonic crystal is very different from the linear $\omega = kc$ behaviour we have in homogeneous media. Another startling effect is the possibility for no allowed optical modes to exist at certain frequency ranges (band gaps) which, coupled with the ability to inject optical modes at specific frequencies by introducing defects in the crystalline structure, has fuelled prospects for novel optical phenomena such as photon localisation, inhibition of spontaneous emission and enhanced nonlinear effects. Novel devices (single-mode LEDs, thresholdless lasers, etc.) are also expected.

Others [5, 6] have concentrated on the contribution of electromagnetic fields to the total energy of a structure and have shown that micron (and submicron) particles in colloidal suspensions can be controlled and ordered on being illuminated by a laser beam. The energetics of these structures is controlled by interplay of structural and electromagnetic properties of the material in the same way that energetics of atomic scale solid is controlled by interplay of electronic bands and atomic arrangements.

Where do we find photonic crystals ? Natural gemstone opals owe their beautiful iridescent colours to their structure which strongly diffracts visible light. However their relatively large amount of disorder as well as the poor refractive index contrast they display renders them impractical for the detailed tailoring of optical properties one needs to attain if the above mentioned novel effects are to be sought after. The first man-made photonic crystals were made ten years ago by Yablonovitch [12, 13] and they operated at the scale of microwaves. Intensified efforts for the past years have born fruit and we are now starting to have artificial opals at visible wavelengths, fabricated by a wealth of experimental techniques which bears witness to the ingenuity with which the problem has been tackled. Some methods aim at building crystals from colloidal suspensions utilising the influence of gravitational, electromagnetic or biochemical interactions, while others concentrate on selectively removing material from a matrix (etching) and thus forming a desired structure. ‘Mixed’ approaches are also used (removing material followed by ordering or stacking of crystal layers) [14]–[28].

The theoretical understanding of photonic structures has centred at solving Maxwell’s equations for the EM wavefield in the presence of matter. Thus the classical theory of vector fields is applied. This methodology, appropriate

for scales large enough compared to the underlying atomic structure, has been successfully applied to predict both the dispersion relation (band structure) as well as the scattering properties of such systems. The main approaches have involved plane-wave expansions [29]-[31], the transfer matrix formalism [32] and the order-N method [33].

1.2 Overview of photonic forces

Nature favours phenomena that are energetically profitable. For example, the Van der Waals force between two macroscopic objects can be found by calculating the change in the self-energy of the vacuum-fluctuation wavefields which multiply scatter off the objects. In electronic band structure, total energy calculations combined with variational arguments have been successfully used to predict the equilibrium structure of crystals and to realistically estimate a plethora of physical quantities [34]. But while the former example involves light waves at the continuum of frequencies and the latter employs electronic waves, there is an increasingly widening range of systems where photons at frequency intervals particular to specific applications apply; of those, photonic crystals form a subset whose potential for new physics and new applications is only beginning to be realised.

In such a crystal, the photonic band structure contains the energy levels accessible to photons. But unlike atomic crystals, where the sea of electrons occupies a range of modes, it is possible with an external source of monochromatic light to selectively populate a particular mode in a photonic crystal. The light source injects many photons but owing to the bosonic and non-interacting nature of light, each photon would remain in the same state. Thus photonic crystals are suitable for observing single-mode band-structure effects, a property implying rich force spectra.

To give an example, consider what happens in a photonic crystal made of dielectric spheres as we traverse with a laser of tunable frequency a resonant mode of the system (such as the modes discussed in chapter 5). At the resonance the fields inside the spheres are enhanced strongly and the corresponding energy density increases by a factor of 1000; away from the resonances the enhancement factor decreases to ~ 5 . This shifting of the wavefield from one region of the

crystal to another strongly affects the forces and may have important implications (and applications) on cohesion in colloidal assemblies.

Apart from performing total energy calculations, EM forces can be also computed in an alternative, more formal manner: namely within the Maxwell Stress Tensor (MST) formalism. In this methodology, the various components of the MST are constructed from the electric and magnetic fields in real space. Integration of the tensor around a closed surface surrounding the body of interest yields the total force acting on it. There is in principle no restriction as to the size and shape of the body, nor as to its dielectric properties. The generality of this method makes its applicability wide, ranging from purely electric to purely magnetic to fully electromagnetic effects; from virtual to real photons; and from travelling to evanescent wavefields.

Suppose we wish to calculate the dispersion forces [†] (these are a class of Van der Waals forces) acting across a plane. This may be the case if we calculate the pressure in a colloidal array, or the force between an AFM tip and a surface. The stress acting across a plane is given by the Maxwell stress tensor, which in turn can be written as an integral over all frequencies, of a function of the causal Green's function, $G^+(\omega)$, embodying the properties of the system. If we can calculate $G^+(\omega)$ on the plane in question then we can calculate the stress tensor. Within the context of our theory $G^+(\omega)$ has a useful on-shell interpretation: it represents the response of a system to electromagnetic waves of frequency ω injected into the system on the plane in question. In free space these waves escape to infinity but in the presence of a nanostructure (such as the cavity between tip and surface) the waves are multiply scattered between tip and surface modifying the net field on the plane in question. To calculate the force we need to know the reflection coefficient of the tip and of the surface to incident electromagnetic waves. That is exactly what our theory is set up to do.

Notice that all internal details of tip and surface are swept into the reflection coefficients: they are all we need to know. The theory also answers another problem that sometimes arises in calculation of dispersion forces: for real ϵ dispersion forces can be thought of as arising from zero point energy attributable to photonic bands. Each stationary state is assigned energy $(\hbar\omega/2)$ and the total

[†]A beautiful overview of the subject of intermolecular and surface forces, from both the experimental and the theoretical sides, is given by Israelachvili [35].

energy is a sum over all states. However this approach fails if ϵ is complex because there are no stationary states: all states decay with time. In contrast the reflection coefficient remains well defined and that is all we need to calculate forces by our method. In other words we have made an easy and elegant generalisation to complex ϵ , vitally important in practice because much of the force calculated with the Green's function approach comes from regions of ω where ϵ is complex, i.e. where $\hbar\omega$ is greater than the band gap of the material in question.

A simpler problem (involving one frequency at a time) is to calculate the forces induced by the application of an external electromagnetic field, as in the colloidal system investigated by Golovchenko [5, 6]. Such a calculation is motivated by the current state of affairs in self-assembly of colloidal spheres, the two main causes of which so far are the gravitational and Van der Waals forces. Of these two, gravity leads invariably to close-packed structures, whereas the Van der Waals force favours only one structure at a time. But for optimal optical properties in a photonic crystal, it is often necessary to obtain a particular structure that may not happen to be favoured by gravity or Van der Waals. Employing our theoretical tools we can go a step further than this, by studying how the crystal structure affects the EM modes (band structure) and then using knowledge of these modes to affect the crystal structure by inducing cohesion while also studying the stability of the final structure.

It would also be interesting to invert the IPE experiment of Gimzewski, sending a laser beam and measuring the response of forces on the tip as ω is varied.

1.2.1 The Atomic Force Microscope (AFM)

A rich variety of forces can be observed by atomic force microscopy [36]. In the non-contact mode (at distances greater than 1 nm between tip and surface, and therefore of special interest to this research) Van der Waals, electrostatic, magnetic or capillary forces produce images of topography. Because its operation does not require a current between the sample surface and the tip, the AFM can move into potential regions inaccessible to the STM or image fragile samples which would be damaged irreparably by the STM electronic current. Insulators, organic materials, biological macromolecules, polymers, ceramics and glasses are some of the many materials which can be imaged.

Since the recent invention of the AFM in 1986 [37], the basic principle of this microscope to measure forces or interactions between a sharp probing tip and the sample surface led to the creation of a variety of other scanning probe microscopes (SPM), such as the magnetic force microscope (MFM), the dipping force microscope (DFM), the friction force microscope (FFM), and the electrostatic force microscope (EFM). The current tendency to combine different methods such as STM/AFM, AFM/MFM, AFM/FFM [38], provides the unique opportunity to characterise a single nm-sized specimen by a combination of methods and therefore gain more information than by the separate application of a single method. The development of commercial microscopes has followed the pace of research. AFM and STM are the most successful instruments in the field of SPM. The possibility to apply AFM to conductive as well as to insulating materials has attracted not only surface physicists and chemists but also biologists, physicians, electrochemists, and mechanical engineers. Atomic force microscopy has thus become a new, rapidly expanding interdisciplinary field [39]. In cell biology, in particular, one of the greatest potential benefits of the AFM is that specimens can be observed in solution, provided they are securely attached to a surface. This capability, not only allows one to image samples at biologically relevant hydration states but also drastically reduces the adhesive interactions between the sample and the scanning tip in air due to meniscus forces caused by condensation on the tip and sample. An exciting prospect provided by the ability of the AFM to operate in solution is that it should be possible to observe dynamic events at high resolution in biological systems, including living cells. This has, in fact, been demonstrated to be true in several cases [40, 41].

“Few problems are so much ignored and at the same time so important, as the accelerated wiping out of the biological resources of the earth. By forcing the other species into extinction, mankind is zealously sawing off the branch on which it is sitting.”

C Castoriades

Chapter 2

Methodology

2.1 Theoretical framework

2.1.1 The need for a stress tensor

We are interested in calculating EM forces on macroscopic bodies in the nanometre scales. It is therefore natural to seek expressions for the forces in terms of macroscopic quantities (such as dielectric functions). Hence we make at the outset an implicit adoption of the continuum picture of matter. We follow the treatment of Landau and Lifshitz [42].

A force acting on a macroscopic system is a sum of the forces $\mathbf{f}_v(\mathbf{r})$ on each of the volume elements that constitute the system. Thus the total force $\mathbf{F}$ can be expressed in terms of the integral

$$\mathbf{F} = \int \mathbf{f}_v(\mathbf{r}) dv, \quad (2.1.1)$$

where $\mathbf{f}_v(\mathbf{r})$ is the force per unit volume on the volume element centred at $\mathbf{r}$. However, the interactions between neighbouring volume elements, being internal forces, must have a zero net effect on the total force. Thus only the forces on volume elements which are in contact with the space exterior to the body will contribute towards the total force. So it must be possible to transform the volume integral above into an integral over the closed surface surrounding the body. This is a consequence of the principle of conservation of momentum, namely that a force on a system must be attributed to a change in the net momentum entering the system (*momentum-balance picture*). From vector analysis, we know that we can transform the volume integral of the vector $\mathbf{f}_v$ into a surface integral as long

as $\mathbf{f}_v$ can be expressed as the divergence of a tensor of rank two:

$$f_{v,i} = \frac{\partial T_{ij}}{\partial x_j}, \quad i, j = \{x, y, z\}, \quad (2.1.2)$$

where a sum over repeated indices is implied from here onwards. The total force on the system then becomes

$$F_i = \int \frac{\partial T_{ij}}{\partial x_j} dv \quad (2.1.3)$$

$$= \oint T_{ij} dS_j, \quad (2.1.4)$$

where a sum over repeated indices is implied from here onwards. The *stress tensor* T_{ij} contains all the information we need. Note it has dimensions of energy density. This is not accidental, since forces can be also thought to arise due to spatial variations in the energy (energy-gradient picture). They act in order to lower the system's energy *i.e.* to push the system into regions where the energy is lowest. Therefore only the spatially varying part of T_{ij} contributes to forces, a fact which can be seen in Eq. (2.1.3). Eq. (2.1.4) is of great practical benefit in that it is often easier to calculate the stress tensor on the surface $\mathbf{S}$ enclosing a solid body rather than in its interior.

2.1.2 The stress tensor for electromagnetism

So far the discussion is general. For the case of electromagnetism, Eqs. (2.1.1)-(2.1.4) describe the force transmitted across the surface $\mathbf{S}$ and acting on the *combined* system of particles and fields inside the volume element [48]. From this we can obtain the force on matter alone by subtracting the rate of change of the radiation momentum internal to the volume; however for systems of interest to us this term is zero and therefore of no consequence, as we explain later. Now we need the stress tensor.

Many treatments exist in the literature (not all of them different): the subject of the volume force and the derivation of the stress tensor from it has been visited by Korteweg [43], Helmholtz [44], Kirchhoff [45], Hertz [46], Cohn [47], Abraham [48], Jeans [49], Gans [50], Sano [51], Pockels [52, 53], Larmor [54], Livens [55, 56], and more recently by Stratton [57], Landau & Lifshitz [58], Panofsky & Phillips [59], Jackson [60] and others. Indeed, to solve for the full force on a

volume element whose dielectric properties are a function of the temperature, density and field while incorporating inhomogeneity, anisotropy, nonlinearity in the ϵ, μ of both the matter inside the volume element and in the medium surrounding it, is not a trivial task. However we can make a few simplifying assumptions which reduce dramatically the difficulty of the problem while still allowing treatment of situations relevant to our purposes.

We consider an isotropic, non-pyroelectric and non-piezoelectric solid body of permittivity ϵ and permeability μ . We assume that ϵ, μ do not change with density changes or pure shears; in other words no electro-striction or magneto-striction effects are considered. Under these restrictions most authors ([43]-[53] and [57]-[60]) agree that the stress tensor that gives the volume force in Eq. (2.1.2) becomes

$$T_{ij}|_1 = \epsilon E_i E_j + \mu H_i H_j - \frac{1}{2} \delta_{ij} (\epsilon E^2 + \mu H^2) \quad (2.1.5)$$

and can be used to yield the stresses *inside* a solid dielectric. To determine the total force on the solid body immersed in a liquid, linear medium (of permittivity ϵ_m and permeability μ_m) which is in mechanical and thermal equilibrium, we can either (i) sum up the volume forces inside the body by integrating the divergence of $T_{ij}|_1$ over volume, in the manner of Eq. (2.1.3), or (ii) sum up the volume forces on a thin shell of liquid medium outside the body by integrating the tensor *in the medium* over a surface external to the body, in the manner of Eq. (2.1.4)[†]. For the latter purpose the above form of $T_{ij}|_1$ will do but with ϵ_m and μ_m in place of ϵ and μ and the external to the body fields (*i.e.* the fields in the liquid) in place of the internal fields.

The expression for $T_{ij}|_1$ is derived [‡] from the volume force which in turn is found via the ‘*energy-method*’: the EM energy of the system is calculated, then the body is allowed a virtual displacement and the force is found from the virtual work done. Under the restrictions imposed above the EM force acting on the matter enclosed in volume $\int d^3r$ is

$$\mathbf{F} = \int \left[\rho_{ext} \mathbf{E} - \frac{1}{2} E^2 \nabla \epsilon + \mathbf{J}_{ext} \times \mathbf{B} - \frac{1}{2} H^2 \nabla \mu + \frac{\tilde{\epsilon}\tilde{\mu} - 1}{c_0^2} \frac{\partial}{\partial t} (\mathbf{E} \times \mathbf{H}) \right] d^3r, \quad (2.1.6)$$

[†]For both (i) and (ii) we should generally subtract the rate of change of radiation momentum from the force yielded by the stress tensor integration, in order to obtain the forces on matter alone.

[‡]See Appendix B.

where ρ_{ext} and $\mathbf{J}_{ext}$ are the external sources of charges and currents. The term $(1/c_0^2) \partial(\mathbf{E} \times \mathbf{H})/\partial t$ is the rate of change of the radiation momentum inside the volume element. By subtracting this term from the force on the combined system of particles and fields, the volume force on particles alone is obtained. For the case of either static or time-averaged effects due to harmonic fields it is not necessary to explicitly carry out this subtraction since $\partial(\mathbf{E} \times \mathbf{H})/\partial t = 0$.

Larmor [54] and Livens [55, 56] have voiced objections against the form $T_{ij}|_1$, proposing instead

$$T_{ij}|_2 = E_i D_j + H_i B_j - \frac{1}{2} \delta_{ij} (\epsilon_0 E^2 + \mu_0 H^2). \quad (2.1.7)$$

Finally, one could be tempted to start by assuming an expression for the force of the type

$$\mathbf{F} = \int [\rho_{tot} \mathbf{E} + \mathbf{J}_{tot} \times \mathbf{B}] d^3 r, \quad (2.1.8)$$

where ρ_{tot} and $\mathbf{J}_{tot}$ are the *total* (external + induced) sources of charges and currents. From this force a third candidate for the stress tensor is derived readily,

$$T_{ij}|_3 = \epsilon_0 E_i E_j + \mu_0 H_i H_j - \frac{1}{2} \delta_{ij} (\epsilon_0 E^2 + \mu_0 H^2). \quad (2.1.9)$$

All three expressions for the stress tensor agree if we are to calculate total forces on solid bodies immersed in vacuum, as is the case with most of our results. For surface forces, however, or for forces on bodies immersed in dielectric liquids, the three forms $T_{ij}|_k$, $\{k = 1, 2, 3\}$ are different. We shall choose $T_{ij}|_1$ in these cases. Two are the reasons behind this choice:

- There appears to be a consensus in the literature to adopt $T_{ij}|_1$. From the early days of the discussion many notable names ([43]-[53], including Helmholtz, Kirchhoff, Hertz, Abraham, Minkowski and others) supported this form. In addition, more recent authors ([57]-[60]) who have either attempted their own derivations (Landau & Lifshitz) or written reviews on the issue (such as Stratton, Jackson and others), have adopted $T_{ij}|_1$; Stratton even claims to have found the fallacy in Livens's formula $T_{ij}|_2$. Certainly the weight of opinion leans to $T_{ij}|_1$.
- We have studied a few test-cases from electrostatics for which the force on a body is actually known from standard calculations. For these systems we have re-derived the force using the different forms for the stress tensor, showing that $T_{ij}|_1$ alone provides the correct answer. Three examples include:

(a) Two equal point charges q are immersed in an infinite, homogeneous, dielectric fluid of relative permittivity $\tilde{\epsilon}$. The charges are separated by a distance r along the $\hat{\mathbf{z}}$ direction. The force exerted on one charge by the other is well known to be

$$F = \frac{q^2}{4\pi\epsilon_0} \frac{1}{r^2} \frac{1}{\tilde{\epsilon}}. \quad (2.1.10)$$

Integration of $T_{ij}|_1$ on a surface enclosing one charge yields exactly this force. For comparison, integrating $T_{ij}|_2$ or $T_{ij}|_3$ on the surface yields

$$F_{2,3} = \frac{q^2}{4\pi\epsilon_0} \frac{1}{r^2} \frac{1}{\tilde{\epsilon}^2}, \quad (2.1.11)$$

which is clearly wrong.

(b) A conducting sphere of radius R and charge Q inside an isotropic, homogeneous dielectric medium of relative permittivity $\tilde{\epsilon}$ under the influence of an initially uniform electrostatic field $\hat{\mathbf{z}}E_0$. The force on the sphere can be derived by a method other than stress-tensor integration [61]. The known result is

$$\mathbf{F} = \hat{\mathbf{z}} Q E_0. \quad (2.1.12)$$

Only $T_{ij}|_1$ produces the correct result when integrated over a surface enclosing the sphere. For comparison, integrating $T_{ij}|_3$ yields

$$\mathbf{F}_3 = \hat{\mathbf{z}} \frac{Q E_0}{\tilde{\epsilon}}, \quad (2.1.13)$$

a wrong result. The same state of affairs occurs if the sphere is dielectric. The correct force acting on the sphere is still given Eq. (2.1.12), reproduced only by $T_{ij}|_1$.

(c) A point charge q at a distance d away from a plane boundary separating two different dielectric media of relative permittivities $\tilde{\epsilon}_1$ (in the half-space where the charge lies) and $\tilde{\epsilon}_2$. The problem has been solved by Landau & Lifshitz [62] employing the method of images. The force on the charge q is

$$F = \frac{q^2}{4\pi\epsilon_0} \frac{1}{(2d)^2} \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1(\tilde{\epsilon}_1 + \tilde{\epsilon}_2)}, \quad (2.1.14)$$

and is reproduced only by integrating $T_{ij}|_1$ over a surface enclosing the charge. Integration of $T_{ij}|_2$ yields

$$F_2 = \frac{q^2}{4\pi\epsilon_0} \frac{1}{(2d)^2} \frac{\tilde{\epsilon}_1^2 + \tilde{\epsilon}_2^2 - 2\tilde{\epsilon}_1\tilde{\epsilon}_2}{\tilde{\epsilon}_1^2(\tilde{\epsilon}_1 + \tilde{\epsilon}_2)^2}, \quad (2.1.15)$$

whereas $T_{ij}|_3$ gives

$$F_3 = \frac{q^2}{4\pi\epsilon_0} \frac{1}{(2d)^2} \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1^2(\tilde{\epsilon}_1 + \tilde{\epsilon}_2)}, \quad (2.1.16)$$

both of which are erroneous. For $\tilde{\epsilon}_2 \rightarrow -\infty$ Eq. (2.1.14) recovers the force on a charge near a conducting plane.

We shall refer to $T_{ij}|_1$ as ‘the stress tensor’ or ‘the Maxwell stress tensor (MST)’ from now on. We will omit the subscript for notational simplicity.

To conclude this section, by integrating $T_{ij}|_1$ of Eq. (2.1.5) over a closed surface $\mathbf{S}$ enclosing the body, the *full* EM force on the body is obtained; if the integration takes place over a region inside the body, the local stresses are obtained. Apart from the restrictions we imposed for the validity of Eq. (2.1.5), no further approximations or limiting assumptions need be made, as has often been done with other methodologies employed for EM effects (e.g. dipole approximation, vanishing particle size compared to the wavelength of light in scattering phenomena, etc.). Our methodology can be applied to dielectric or metallic objects of any shape and at any frequency range in principle. The only requirement for meaningful results is that the integration is carried out over scales that are large with respect to atomic sizes, so that the continuum picture of matter is justified. For nanostructure scales of interest to us (> 50 nm) this requirement is surely satisfied: the dynamics of the EM wavefield is more properly described by the equations of Maxwell rather than by the Schrödinger equation for all the electronic charges which make up the dielectric response of the system. This choice of macroscopic scales dictates a relevant timescale also for the type of fields we are interested in (no macroscopic object can respond synchronously to THz oscillations): we must time-average the forces (*i.e.* the stress tensor) over many cycles when dealing with harmonic fields at optical frequencies. A consequence of time-averaging is that the rate of change of the EM field momentum inside the volume of integration vanishes. Thus it is the material bodies that absorb all momentum transfer between our system and the outside world. The stress tensor integration yields exactly this force.

2.2 Numerical methodology

2.2.1 Description of approach

Fields

We discretise Maxwell's equations on a simple cubic (SC) mesh. For calculations of photonic band structure, periodic boundary conditions are used in all directions. For transmission results, we consider a crystal finite in the z direction, having a thickness of N unit cells (layers), N being allowed to vary. We shall discuss here the case where a monochromatic light beam of frequency ω , polarisation σ and wavevector $\mathbf{k}$ is incident on the crystal from the $-z$ side. Using the Transfer Matrix formalism [63] we calculate the complex reflection coefficients $R_{\mathbf{k}'\sigma'}$ for scattering into plane waves of wavevector $\mathbf{k}' = \mathbf{k}_{\parallel} + \mathbf{g}_{\parallel} + \hat{\mathbf{z}}k'_z$ and polarisation σ' , where $\mathbf{k}_{\parallel}$ is the component of $\mathbf{k}$ parallel to the xy plane and $\mathbf{g}_{\parallel}$ is a *two-dimensional* reciprocal lattice vector also lying on the xy plane. Our computer codes have been published and issues of stability and accuracy in the calculation of reflection coefficients and band structures have been discussed elsewhere [63]-[66].

Forces

The new ingredient is the calculation of forces. From the reflection coefficients we construct the total fields on the $-z$ side of the crystal,

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 \sum_{\mathbf{k}', \sigma'} (\delta_{\mathbf{k}, \mathbf{k}'} \delta_{\sigma, \sigma'} + R_{\mathbf{k}'\sigma'}) \hat{\mathbf{e}}_{\mathbf{k}'\sigma'} e^{i[\mathbf{k}' \cdot \mathbf{r} - \omega t]}, \quad (2.2.1)$$

$$\mathbf{H}(\mathbf{r}, t) = \mathbf{H}_0 \sum_{\mathbf{k}', \sigma'} (\delta_{\mathbf{k}, \mathbf{k}'} \delta_{\sigma, \sigma'} + R_{\mathbf{k}'\sigma'}) \hat{\mathbf{e}}_{\mathbf{k}'\sigma'} e^{i[\mathbf{k}' \cdot \mathbf{r} - \omega t]}. \quad (2.2.2)$$

Here $\mathbf{E}_0, \mathbf{H}_0$ are the amplitudes of the incident fields and $\hat{\mathbf{e}}_{\mathbf{k}'\sigma'}$ are the unit vectors for each $\mathbf{k}'$ and each polarisation σ' . The Kronecker delta functions serve as to add the contribution from the incident field, which is present on the $-z$ side, to the reflected field. For the total fields on the $+z$ side the only contribution comes from the transmitted fields.

Having constructed the total fields on either side of the crystal, we invoke once again the Transfer Matrix equations to find the total field at each mesh point inside the structure. In doing so we propagate the total fields on the $-z$ side forwards, and the total fields on the $+z$ side backwards.

When this is accomplished, the stress tensor components are calculated and time-averaged. The latter procedure can be made in a standard way since the fields are expressed as sums of plane waves at the same frequency ω . Thus, the time-averaged stress tensor is

$$\langle T_{ij} \rangle = \frac{1}{2} \Re \left\{ \epsilon^* E_i^* E_j + \mu^* H_i^* H_j - \frac{1}{2} \delta_{ij} (\epsilon^* \mathbf{E}^* \cdot \mathbf{E} + \mu^* \mathbf{H}^* \cdot \mathbf{H}) \right\}, \quad (2.2.3)$$

where the star denotes the complex conjugate. In a similar manner, all quantities involving products of fields are calculated:

$$\langle \rho \rangle = \frac{1}{2} \Re \left\{ \frac{1}{2} [\mathbf{E} \cdot \mathbf{D}^* + \mathbf{H} \cdot \mathbf{B}^*] \right\}, \quad (2.2.4)$$

for the EM energy density, and

$$\langle \mathbf{S}_P \rangle = \frac{1}{2} \Re \left\{ \mathbf{E} \times \mathbf{H}^* \right\}, \quad (2.2.5)$$

for the Poynting flux vector (which is proportional to the EM momentum density). $\mathbf{S}_P$ is calculated only for the travelling terms of the field expansions in Eqs. (2.2.1), (2.2.2), *i.e.* for the terms of real wavevectors $\mathbf{k}'$.

For notational simplicity we shall drop the brackets $\langle \rangle$ on $\mathbf{S}$, ρ and T_{ij} from now on, while still implying that time averaging has been carried out.

Finally, the forces are found by integrating the stress tensor over a surface enclosing the body of interest, which can in principle be of arbitrary shape and complexity. In this work we present force calculations on (i) isolated dielectric objects (artificial atoms), (ii) crystal layers or layers of homogeneous media and (iii) the entire crystal sample. For (i), the integration surface is confined in all directions as it covers the cubic unit cell, which never contains more than one artificial atom in our calculations. For (ii), the surface consists of two infinite sheets spreading out in the xy plane. In practice the integration needs not be carried out over more area than the xy faces of the unit cell due to the periodic boundary conditions. For (iii), two xy faces are used, one on the $-z$ side, the other on the $+z$ side of the crystal such that the whole crystal is enclosed. When the integration has taken place yielding the force, the pressure may be derived by dividing by the surface area of integration.

2.2.2 Computational machinery & technicalities

Familiarity with the OPAL codes is assumed in this section. The numerical details of these codes are described in detail in Refs. [64], [65].

Normalisation and units

Whatever the system of units being used, it is important to relate calculated quantities like pressure, energy, and force to experimental variables such as the intensity I_0 of the incident beam of light. Careful normalisation of the fields is therefore required, and a system of units easily compatible with experiment is desirable. The OPAL suite of codes operates in atomic units (a.u.); the permittivity, permeability, frequency, wavevectors and fields are all expressed in these units. However we shall express the final outcome of force, pressure and energy calculations in SI units.

Units in diagrams

In all diagrams the force, pressure, stress tensor and the energy density will be in SI units (N, N/m², N/m² and J/m³ respectively). The frequency will always be in eV. For the band structure plots the wavevectors k_z will be in multiples of π/l_0 , where l_0 is the unit-cell length which is always be $l_0 = 900$ nm in our calculations. The incident beam intensity will be $I_0 = 3 \times 10^8$ W/m², unless otherwise noted.

Fields

After the reflection coefficients are computed, we renormalise the polarisation vectors $\hat{\mathbf{e}}_{\mathbf{k}'\sigma'}$ to unit magnitude each (taking care to retain all 6 components of the eigenvectors RVEC, since we shall need all 3+3 components of the $\mathbf{E}, \mathbf{H}$ fields for the force calculation). This is accomplished by multiplying each current-carrying polarisation vector with its current, reversing the procedure carried out in subroutine PLANEW. Had we not done this the total fields (and hence the forces) would have a normalisation that varied with frequency ω , since the currents are frequency-dependent. We also take $|E_0| = \sqrt{\frac{2I_0}{\epsilon_0 c}}$ (in a.u.). With these two measures the incident $\mathbf{E}$ field has the correct magnitude in a.u., corresponding to an

incident beam with intensity I_0 . The incident $\mathbf{H}$ field has magnitude $|\frac{k}{\omega\mu_0} \cdot E_0|^\dagger$. Then the total $\mathbf{E}$ and $\mathbf{H}$ fields in a.u. are obtained from Eqs. (2.2.1), (2.2.2).

The conversion of the fields into SI units gives:

$$[E]_{SI} = \frac{\text{Hartree}}{e a_B} [E]_{au} = (5.14222 \times 10^{11}) [E]_{au} \quad (2.2.6)$$

$$[H]_{SI} = \frac{e \text{ Hartree}}{\hbar a_B} [H]_{au} = (1.25168 \times 10^8) [H]_{au}. \quad (2.2.7)$$

Maxwell Stress Tensor

Numerically, a ‘local’ expression for the stress tensor is applied:

$$T_{ij}(\mathbf{r}) = \epsilon E_i(\mathbf{r}) E_j(\mathbf{r}) + \mu H_i(\mathbf{r}) H_j(\mathbf{r}) - \frac{1}{2} \delta_{ij} [\epsilon E^2(\mathbf{r}) + \mu H^2(\mathbf{r})]. \quad (2.2.8)$$

T_{ij} has the units of the product ϵE^2 . Inserting the factors ϵ and μ in a.u. and converting into SI units yields

$$[T_{ij}]_{SI} = \frac{\text{Hartree}}{a_B^3} [T_{ij}]_{au} = (2.94209 \times 10^{13}) [T_{ij}]_{au}. \quad (2.2.9)$$

Force and pressure

Pressure has the same units as T_{ij} . For the force we have

$$[F]_{SI} = \frac{\text{Hartree}}{a_B} [F]_{au} = (8.23870 \times 10^{-8}) [F]_{au}. \quad (2.2.10)$$

Energy

The energy density ρ has the same units as T_{ij} . As for the energy, U , its units are

$$[U]_{SI} = \text{Hartree} [U]_{au} = (4.35973 \times 10^{-18}) [U]_{au}. \quad (2.2.11)$$

Both ρ and U are defined using a ‘local’ expression.

[†] $\mathbf{H}$ is obtained from $\mathbf{H}'$ by a straightforward multiplication with the factor $\frac{c\omega\epsilon_0}{i}$.

Momentum density

For the Poynting vector $\mathbf{S}_P$ a ‘non-local’ definition has been adopted [63] such that the conservation of energy (Poynting theorem) is obeyed exactly. For the units of $\mathbf{S}_P$ we have

$$[S_P]_{SI} = \frac{\text{Hartree}^2}{\hbar a_B^2} [S_P]_{au} = (6.43639 \times 10^{19}) [S]_{au}. \quad (2.2.12)$$

Numerical tests

Several numerical tests were carried out:

- The transmitted field, propagated backwards until outside the system, should equal the sum of the incident + reflected field on the $-z$ side of the system. We checked this and verified the equality for all field components. Typically the agreement was up to at least 6 decimal digits for the systems studied and the energy range examined.
- At each mesh point we checked that the divergence equations $\nabla \cdot \mathbf{D} = 0$ and $\nabla \cdot \mathbf{H} = 0$ were satisfied, typically up to 12 decimal digits or more.
- Where possible, checks of the numerically computed reflectance, transmittance, fields, force, pressure and energy density were made against analytical calculations. For homogeneous half-spaces and homogeneous layers the scattering problem was solved analytically and such tests were made. Accuracy to at least 6 decimal digits was obtained.

“Forgive my facile symbolism, but our age is exactly commensurate with this truth: a stunning technical progress strives to heal mankind of ills caused by a stunning technical progress. How long since a man dreamt of a bird!”

Odysseus Elytis

Chapter 3

Homogeneous system

3.1 Homogeneous systems studied: a half-space and a layer

Probably the simplest system for studying macroscopic radiation forces on materials is a homogeneous half-space (dielectric or metallic) upon which a single beam of light is incident (Fig. 3.1). The simplicity of this model system will allow us to put forward in a transparent manner some important features of our methodology. Furthermore, as this problem is analytically solvable it provides a nice test-ground for our numerical results.

We model the semi-infinite medium by considering a layer of material of sufficient thickness in the z direction so that all light entering the layer is absorbed before reaching the far end. For a metal this would only take a thickness equal to the skin depth (a few nm typically) when ω lies below the plasma frequency ω_p ; for a real dielectric, the tiny absorption present makes it necessary to consider much thicker samples, perhaps as much as several cm. In either case, only the incident and specularly reflected wavefields are present outside the system. The cross section of the light beam is regarded as infinite so that edge effects are removed. The total force on the half-space is found by integrating the stress tensor for the total field (incident plus reflected) on a surface S_z which lies on the $-z$ side immediately above the medium and extends to infinity in the xy plane (the contribution from the integration on the far end is zero, since the fields vanish there). A more appropriate quantity for this system is the EM pressure $P_i = (1/S_z) \oint T_{iz} dS_z$ on the layer. The incident beam has intensity I_0 and travels

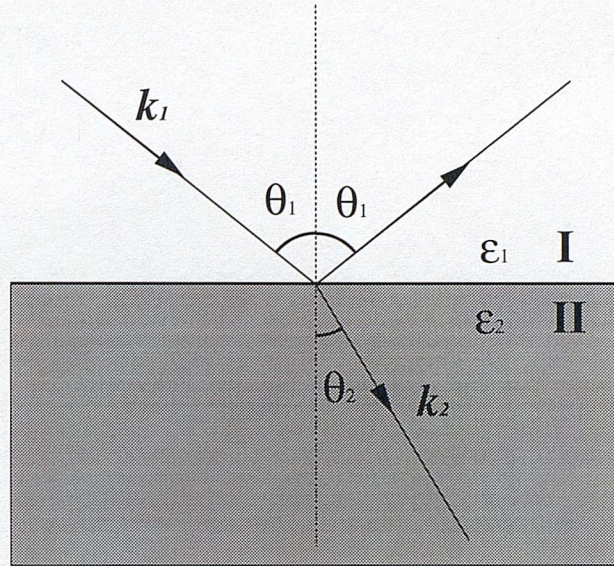


Figure 3.1: Reflection and transmission of a beam of monochromatic light upon incidence on the interface between two different semi-infinite dielectric media (half-spaces).

at the speed c_0 , which implies an incident radiation pressure

$$P_{inc} = \frac{I_0}{c_0}. \quad (3.1.1)$$

Another system which is analytically tractable is a homogeneous layer of finite width d such that some light is transmitted through the far end (see Fig. 3.2). The layer extends to infinity in the x and y directions. The integration of the stress tensor takes place on the $z = 0^-$ and $z = d^+$ surfaces. Like before, the light beam is regarded as infinitely wide. Given the cross section of modern lasers, this is realistic approximation.

We will be studying the forces on both systems with an aim to understand how they arise physically, what their limiting values are and whether they can become attractive. We shall be studying the influence of both travelling and evanescent wavefields.

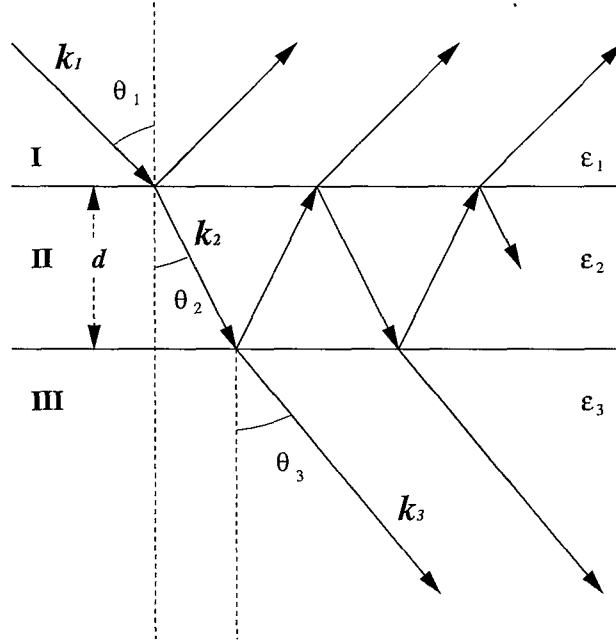


Figure 3.2: A wave incident at angle θ_1 from medium I is scattered by the interfaces with media II and III. When $\epsilon_2 < \epsilon_1$ the fields in II become evanescent for large enough θ_1 . For visual clarity all beams are pictured as rays with zero cross section; in our calculations we regard each beam as having infinite cross section.

3.2 Media studied: GaP and Al

As a dielectric system we chose GaP which is transparent (i.e. has low absorption and the permittivity ϵ is mostly real and positive) over near infrared and visible frequencies. In the ultraviolet spectrum it possesses a plasma frequency at ~ 13.5 eV and the absorptive part of ϵ becomes significant with respect to the real part. GaP's dielectric properties in the infrared will be discussed in detail in the following chapter.

As a metallic system we use Al, for which the dielectric function can be modelled as

$$\epsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma_D)}, \quad (3.2.1)$$

where $\omega_p = 15$ eV and the damping coefficient for the intraband transitions is $\gamma_D = 0.1$ eV.

Our data for the dielectric properties of GaP and Al materials come from [67].

3.3 Analytical formulae: half-space

We will now calculate the reflection and transmission coefficients for a light beam of wavelength λ incident obliquely on a material of permittivity $\epsilon_2 = \epsilon'_2 + i\epsilon''_2$. No free charges or electrical currents are present on the surface or in the bulk of the dielectric. The medium from which light is incident has a real positive permittivity $\epsilon_1 = \epsilon'_1$. For all plots $\epsilon'_1 = 1$, however the analytical formulae we derive cater for the general case $\epsilon'_1 \neq 1$.

3.3.1 Units and notation

In order to avoid confusion we must define our units and notation carefully. The equations of Maxwell for a transverse wavefield that varies as $\sim e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$ give us

$$\nabla \times (\nabla \times \mathbf{E}) = -\mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2} \quad (3.3.1)$$

$$\Rightarrow \mathbf{k} \times (\mathbf{k} \times \mathbf{E}) = -\mu_0 \epsilon_0 \tilde{\epsilon} \omega^2 \mathbf{E}, \quad (3.3.2)$$

where from now on all symbols with a tilde are dimensionless, *i.e.* they are pure numbers. We expand the left-hand side,

$$\mathbf{k} (\mathbf{k} \cdot \mathbf{E}) - (\mathbf{k} \cdot \mathbf{k}) \mathbf{E} = -\mu_0 \epsilon_0 \tilde{\epsilon} \omega^2 \mathbf{E} \quad (3.3.3)$$

$$\Rightarrow \mathbf{k} \cdot \mathbf{k} = \mu_0 \epsilon_0 \tilde{\epsilon} \omega^2, \quad (3.3.4)$$

which is the dispersion relation in a non-magnetic medium with permittivity $\epsilon_0 \tilde{\epsilon}$. Separating the units from the wavevectors we can re-express the left-hand side term as

$$\mathbf{k} \cdot \mathbf{k} = \mu_0 \epsilon_0 \omega^2 (\tilde{\mathbf{k}} \cdot \tilde{\mathbf{k}}), \quad (3.3.5)$$

which leads us to the *dimensionless* dispersion relation,

$$\tilde{\mathbf{k}} \cdot \tilde{\mathbf{k}} = \tilde{\epsilon}. \quad (3.3.6)$$

The advantage of Eq. (3.3.6) is that we can derive the wavevectors from the dielectric function without worrying about units. We are interested in cases where the wavevectors can have an imaginary component along the z -direction, *i.e.*

$$\mathbf{k} = \hat{\mathbf{x}} \beta + \hat{\mathbf{z}} (\gamma' + i\gamma''), \quad (3.3.7)$$

for which Eq. (3.3.6) gives us

$$\tilde{\beta}^2 + \tilde{\gamma}'^2 - \tilde{\gamma}''^2 = \tilde{\epsilon}' \quad (3.3.8)$$

$$2 \tilde{\gamma}' \tilde{\gamma}'' = \tilde{\epsilon}'' . \quad (3.3.9)$$

3.3.2 Boundary conditions (BCs)

The standard boundary conditions apply at the interface, namely the continuity of the following field components:

$$\mathbf{B}_\perp, \mathbf{D}_\perp, \mathbf{E}_\parallel, \mathbf{H}_\parallel . \quad (3.3.10)$$

In addition, due to the homogeneity of both materials we expect that the boundary conditions should not change with time or location on the interface, which implies that the harmonic terms are such that

$$\mathbf{k}_{inc} \cdot \mathbf{r}_{z=0} = \mathbf{k}_{ref} \cdot \mathbf{r}_{z=0} = \mathbf{k}_{tr} \cdot \mathbf{r}_{z=0} , \quad (3.3.11)$$

which is Snell's law.

Given these conditions and starting with an incident wave of amplitude $\mathbf{E}_0$ and wavevector $\mathbf{k}_0$ we can solve for the reflected and transmitted waves with straightforward algebra.

In what follows we have assumed an incident wavevector

$$\mathbf{k}_{inc} = \mathbf{k}_1 = (\beta, 0, \gamma_1) = (k_1 \sin \theta_1, 0, k_1 \cos \theta_1) \quad (3.3.12)$$

without any loss of generality. Snell's law then gives us the reflected wavevector

$$\mathbf{k}_{ref} = (\beta, 0, -\gamma_1) = (k_1 \sin \theta_1, 0, -k_1 \cos \theta_1) \quad (3.3.13)$$

and the transmitted wavevector

$$\mathbf{k}_{tr} = \mathbf{k}_2 = (\beta, 0, \gamma_2' + i\gamma_2'') . \quad (3.3.14)$$

3.3.3 Reflection coefficients

Polarisation is conserved for scattering from a dielectric half-space.

S polarisation ($\mathbf{E} \perp$ plane of incidence)

For the electric fields we have

$$\mathbf{E}_{inc} = \hat{\mathbf{y}} E_0 e^{i(\beta x + \gamma_1 z - \omega t)} \quad (3.3.15)$$

$$\mathbf{E}_{ref} = \hat{\mathbf{y}} |R_{ss}| E_0 e^{i(\beta x - \gamma_1 z - \omega t + \phi_1)} \quad (3.3.16)$$

$$\mathbf{E}_{tr} = \hat{\mathbf{y}} |T_{ss}| E_0 e^{i(\beta x + \gamma'_2 z - \omega t + \phi_2) - \gamma''_2 z} . \quad (3.3.17)$$

The magnetic fields can be found readily from the electric fields, since

$$\mathbf{H} = \frac{1}{\omega \mu_0} \mathbf{k} \times \mathbf{E} . \quad (3.3.18)$$

Thus we obtain

$$\mathbf{H}_{inc} = \frac{1}{\omega \mu_0} (-\hat{\mathbf{x}} \gamma_1 + \hat{\mathbf{z}} \beta) E_0 e^{i(\beta x + \gamma_1 z - \omega t)} \quad (3.3.19)$$

$$\mathbf{H}_{ref} = \frac{1}{\omega \mu_0} (\hat{\mathbf{x}} \gamma_1 + \hat{\mathbf{z}} \beta) |R_{ss}| E_0 e^{i(\beta x - \gamma_1 z - \omega t + \phi_1)} \quad (3.3.20)$$

$$\mathbf{H}_{tr} = \frac{1}{\omega \mu_0} [-\hat{\mathbf{x}}(\gamma'_2 + i\gamma''_2) + \hat{\mathbf{z}} \beta] |T_{ss}| E_0 e^{i(\beta x + \gamma'_2 z - \omega t + \phi_2) - \gamma''_2 z} . \quad (3.3.21)$$

Thus in our notation the amplitudes $|R_{ss}|, |T_{ss}|$ are real positive (*i.e.* they are the *Fresnel* coefficients), and all phase dependence is taken care of by the exponential factors. The boundary conditions at the interface (Eq. (3.3.10)) provide the equations from which the reflections coefficients are determined:

$$1 + |R_{ss}| e^{i\phi_1} = |T_{ss}| e^{i\phi_2} \quad (3.3.22)$$

$$\gamma_1 (-1 + |R_{ss}| e^{i\phi_1}) = -(\gamma'_2 + i\gamma''_2) |T_{ss}| e^{i\phi_2} . \quad (3.3.23)$$

Thus we have four equations (real and imaginary parts of the above two) for an equal number of unknowns. From these we find

$$|R_{ss}| = \sqrt{\frac{(\gamma_1 - \gamma'_2)^2 + \gamma''_2{}^2}{(\gamma_1 + \gamma'_2)^2 + \gamma''_2{}^2}} \quad (3.3.24)$$

for reflection and

$$|T_{ss}| = \frac{2\gamma_1}{\sqrt{(\gamma_1 + \gamma'_2)^2 + \gamma''_2{}^2}} \quad (3.3.25)$$

for transmission. The phases are given by

$$\tan \phi_1 = \frac{2\gamma_1\gamma_2''}{(\gamma_2'^2 + \gamma_2''^2) - \gamma_1^2} \quad (3.3.26)$$

$$\tan \phi_2 = -\frac{\gamma_2''}{\gamma_1 + \gamma_2'} , \quad (3.3.27)$$

and are *uniquely* determined from the relations

$$\cos \phi_2 = \frac{+1}{\sqrt{1 + \tan^2 \phi_2}} \quad (3.3.28)$$

$$\sin \phi_2 = \tan \phi_2 \cos \phi_2 \quad (3.3.29)$$

$$\sin \phi_1 = \frac{|T_{ss}|}{|R_{ss}|} \sin \phi_2 \quad (3.3.30)$$

$$\cos \phi_1 = \frac{|T_{ss}| \cos \phi_2 - 1}{|R_{ss}|} . \quad (3.3.31)$$

P polarisation (E || plane of incidence)

For the electric fields we have

$$\mathbf{E}_{inc} = (-\hat{\mathbf{x}} \cos \theta_1 + \hat{\mathbf{z}} \sin \theta_1) E_0 e^{i(\beta x + \gamma_1 z - \omega t)} \quad (3.3.32)$$

$$\mathbf{E}_{ref} = (+\hat{\mathbf{x}} \cos \theta_1 + \hat{\mathbf{z}} \sin \theta_1) |R_{pp}| E_0 e^{i(\beta x - \gamma_1 z - \omega t + \phi_1)} \quad (3.3.33)$$

$$\mathbf{E}_{tr} = (-\hat{\mathbf{x}} \cos \theta_2 + \hat{\mathbf{z}} \sin \theta_2 e^{i\alpha}) |T_{pp}| E_0 e^{i(\beta x + \gamma_2' z - \omega t + \phi_2) - \gamma_2'' z} , \quad (3.3.34)$$

where the angle of the transmitted beam with respect to the surface normal is given by

$$\sin \theta_2 = \frac{\beta}{k_2} = \frac{\beta}{\sqrt{\beta^2 + \gamma_2'^2 + \gamma_2''^2}} \quad (3.3.35)$$

$$\cos \theta_2 = \frac{|\gamma_2' + i \gamma_2''|}{k_2} = \sqrt{\frac{\gamma_2'^2 + \gamma_2''^2}{\beta^2 + \gamma_2'^2 + \gamma_2''^2}} . \quad (3.3.36)$$

For the magnetic fields,

$$\mathbf{H}_{inc} = -\hat{\mathbf{y}} \frac{k_1}{\omega \mu_0} E_0 e^{i(\beta x + \gamma_1 z - \omega t)} \quad (3.3.37)$$

$$\mathbf{H}_{ref} = -\hat{\mathbf{y}} \frac{k_1}{\omega \mu_0} |R_{pp}| E_0 e^{i(\beta x - \gamma_1 z - \omega t + \phi_1)} \quad (3.3.38)$$

$$\begin{aligned} \mathbf{H}_{tr} = & -\hat{\mathbf{y}} \frac{1}{\omega \mu_0} \left[(\gamma'_2 + i\gamma''_2) \cos \theta_2 + \beta \sin \theta_2 e^{i\alpha} \right] \times \\ & \times |T_{pp}| E_0 e^{i(\beta x + \gamma'_2 z - \omega t + \phi_2) - \gamma''_2 z}. \end{aligned} \quad (3.3.39)$$

The boundary conditions become

$$\cos \theta_1 (1 - |R_{pp}| e^{i\phi_1}) = \cos \theta_2 |T_{pp}| e^{i\phi_2} \quad (3.3.40)$$

$$k_1 (1 + |R_{pp}| e^{i\phi_1}) = \left[(\gamma'_2 + i\gamma''_2) \cos \theta_2 + \beta \sin \theta_2 e^{i\alpha} \right] |T_{pp}| e^{i\phi_2} \quad (3.3.41)$$

$$k_1^2 \sin \theta_1 (1 + |R_{pp}| e^{i\phi_1}) = (\beta^2 + \gamma_2'^2 - \gamma_2''^2 + i \cdot 2\gamma_2' \gamma_2'') \sin \theta_2 e^{i\alpha} |T_{pp}| e^{i\phi_2}. \quad (3.3.42)$$

For the reflection coefficients we find (after long algebra and many coffees through the darkest nights !),

$$|R_{pp}| = \sqrt{1 - \frac{4\gamma_1 \gamma_2' k_2^2}{k_1^2 (\gamma_2'^2 + \gamma_2''^2) + k_2^4 \cos^2 \theta_1 + 2\gamma_1 \gamma_2' k_2^2 - 4\beta^2 \gamma_2''^2 \cos^2 \theta_1}}, \quad (3.3.43)$$

and for transmission,

$$|T_{pp}| = \frac{2\gamma_1 k_2}{\sqrt{k_1^2 (\gamma_2'^2 + \gamma_2''^2) + k_2^4 \cos^2 \theta_1 + 2\gamma_1 \gamma_2' k_2^2 - 4\beta^2 \gamma_2''^2 \cos^2 \theta_1}}. \quad (3.3.44)$$

For the phases we have,

$$\tan \alpha = -\frac{\gamma_2''}{\gamma_2'} \quad , \quad \alpha \in \left[-\frac{\pi}{2}, 0\right] \quad (3.3.45)$$

$$\tan \phi_1 = \frac{2\gamma_1 \gamma_2'' (\beta^2 - \gamma_2'^2 - \gamma_2''^2)}{k_1^2 (\gamma_2'^2 + \gamma_2''^2) - k_2^4 \cos^2 \theta_1 + 4\beta^2 \gamma_2''^2 \cos^2 \theta_1} \quad (3.3.46)$$

$$\tan \phi_2 = \frac{\gamma_1 \gamma_2'' (\beta^2 - \gamma_2'^2 - \gamma_2''^2)}{k_1^2 (\gamma_2'^2 + \gamma_2''^2) + \gamma_1 \gamma_2' k_2^2}, \quad (3.3.47)$$

and in order to determine the phase factors uniquely,

$$\cos \phi_2 = \frac{+1}{\sqrt{1 + \tan^2 \phi_2}} \quad (3.3.48)$$

$$\sin \phi_2 = \tan \phi_2 \cos \phi_2 \quad (3.3.49)$$

$$\cos \phi_1 = \frac{1}{|R_{pp}|} - \frac{\cos \theta_2}{\cos \theta_1} \frac{|T_{pp}|}{|R_{pp}|} \cos \phi_2 \quad (3.3.50)$$

$$\sin \phi_1 = -\frac{\cos \theta_2}{\cos \theta_1} \frac{|T_{pp}|}{|R_{pp}|} \sin \phi_2. \quad (3.3.51)$$

These solutions for both the $\mathbf{E}$ and $\mathbf{H}$ fields and for both polarisations have been tested and found to satisfy Maxwell's equations in both media. In addition they fulfil the boundary conditions for all field components. All special cases ($\gamma_2'' = 0, \theta_1 = 0$, total internal reflection, total polarisation at the Brewster angle for p light) have been checked.

3.3.4 Maxwell Stress Tensor

Since both half-spaces and their interface do not vary in any way in the parallel directions (*i.e.* $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$) and since the parallel wavevector β is real, it follows that the stress tensor T_{ij} is invariant along the parallel directions also. Therefore the only non-zero contributions to the force arise from the integration along the $\hat{\mathbf{z}}$ direction. Hence the only components of T_{ij} that are needed are T_{xz}, T_{yz} and T_{zz} . Of those T_{yz} is zero since for s polarisation $E_z = H_y = 0$, whereas for p polarisation $E_y = H_z = 0$. This reduces the number of necessary components to two.

S polarisation ($\mathbf{E} \perp$ plane of incidence)

Having computed the EM fields we can obtain the stress tensor components. For medium 1, we need the resultant field:

$$\mathbf{E}^{(1)} = \mathbf{E}_{inc} + \mathbf{E}_{ref} \quad (3.3.52)$$

$$= \hat{\mathbf{y}} E_0 e^{i(\beta x + \gamma_1 z - \omega t)} \left[1 + |R_{ss}| e^{i(-2\gamma_1 z + \phi_1)} \right], \quad (3.3.53)$$

and

$$\mathbf{H}^{(1)} = \mathbf{H}_{inc} + \mathbf{H}_{ref} \quad (3.3.54)$$

$$= \frac{E_0}{\omega \mu_0} e^{i(\beta x + \gamma_1 z - \omega t)} \times \\ \times \left[\hat{\mathbf{x}} \gamma_1 (-1 + |R_{ss}| e^{i(-2\gamma_1 z + \phi_1)}) + \hat{\mathbf{z}} \beta (1 + |R_{ss}| e^{i(-2\gamma_1 z + \phi_1)}) \right]. \quad (3.3.55)$$

Thus the time-averaged stress tensor components are

$$\begin{aligned} T_{xz}^{(1)} &= T_{zx}^{(1)} \\ &= \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left(\frac{\beta \gamma_1}{k_1^2} \right) (|R_{ss}|^2 - 1) \end{aligned} \quad (3.3.56)$$

$$T_{zz}^{(1)} = -\frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left(\frac{\gamma_1^2}{k_1^2} \right) (1 + |R_{ss}|^2). \quad (3.3.57)$$

For medium 2,

$$\begin{aligned} T_{xz}^{(2)} &= T_{zx}^{(2)} \\ &= -\frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 |T_{ss}|^2 \left(\frac{\beta \gamma_2'}{k_1^2} \right) e^{-2\gamma_2'' z} \end{aligned} \quad (3.3.58)$$

$$T_{zz}^{(2)} = -\frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 |T_{ss}|^2 \left(\frac{\gamma_2'^2}{k_1^2} \right) e^{-2\gamma_2'' z}. \quad (3.3.59)$$

P polarisation ($\mathbf{E} \parallel$ plane of incidence)

From the total fields we derive the stress tensor components. It turns out that exactly the same expressions for T_{xx} and T_{zz} are found, but with the substitutions $|R_{pp}|$ for $|R_{ss}|$ and $|T_{pp}|$ for $|T_{ss}|$, and of course with the phase angles for p light as given by Eqs. (3.3.46)-(3.3.51).

3.3.5 Energy density

The time-averaged EM energy density is

$$\rho(\mathbf{r}) = \frac{1}{4} \Re \left\{ \left(\mathbf{E}(\mathbf{r}) \cdot \mathbf{D}^*(\mathbf{r}) + \mathbf{H}(\mathbf{r}) \cdot \mathbf{B}^*(\mathbf{r}) \right) \right\}, \quad (3.3.60)$$

from which the EM energy is found:

$$U = \int \rho(\mathbf{r}) d^3 r. \quad (3.3.61)$$

Thus we obtain for s polarisation,

$$\rho^{(1)} = \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left[1 + |R_{ss}|^2 + 2|R_{ss}| \left(\frac{\beta^2}{k_1^2} \right) \cos(2\gamma_1 z - \phi_1) \right] \quad (3.3.62)$$

$$\rho^{(2)} = \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 |T_{ss}|^2 E_0^2 \left(\frac{\beta^2 + \gamma_2'^2}{k_1^2} \right) e^{-2\gamma_2'' z}. \quad (3.3.63)$$

For p polarisation,

$$\rho^{(1)} = \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left[1 + |R_{pp}|^2 + 2|R_{pp}| \left(\frac{\beta^2}{k_1^2} \right) \cos(2\gamma_1 z - \phi_1) \right] \quad (3.3.64)$$

$$\rho^{(2)} = \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 |T_{pp}|^2 E_0^2 \left(\frac{\beta^4 + \gamma_2'^4 + 2\beta^2 \gamma_2'^2 + \gamma_2'^2 \gamma_2''^2 - \beta^2 \gamma_2''^2}{k_1^2 k_2^2} \right) e^{-2\gamma_2'' z}. \quad (3.3.65)$$

3.3.6 Forces

Volume forces inside medium 1

They are zero since neither $T_{xz}^{(1)}$ nor $T_{zz}^{(1)}$ varies with z .

Volume forces inside medium 2

They are nonzero only if there is absorption *i.e.* if $\gamma_2'' \neq 0$.

$$P_x = \left(T_{xz}^{(2)}(z_f) - T_{xz}^{(2)}(z_i) \right) \quad (3.3.66)$$

$$= -\frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 |T_{\sigma\sigma}|^2 \left(\frac{\beta \gamma_2'}{k_1^2} \right) (e^{-2\gamma_2'' z_f} - e^{-2\gamma_2'' z_i}), \quad (3.3.67)$$

where $\sigma = s$ or p , and

$$P_z = \left(T_{zz}^{(2)}(z_f) - T_{zz}^{(2)}(z_i) \right) \quad (3.3.68)$$

$$= -\frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 |T_{\sigma\sigma}|^2 \left(\frac{\gamma_2'^2}{k_1^2} \right) (e^{-2\gamma_2'' z_f} - e^{-2\gamma_2'' z_i}). \quad (3.3.69)$$

Interface forces

The normal pressure on the interface is

$$P_{z,int} = \left(T_{zz}^{(2)}(0^+) - T_{zz}^{(1)}(0^-) \right) \quad (3.3.70)$$

$$= -\frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left[|T_{\sigma\sigma}|^2 \left(\frac{\gamma_2'^2}{k_1^2} \right) - (1 + |R_{\sigma\sigma}|^2) \left(\frac{\gamma_1'^2}{k_1^2} \right) \right], \quad (3.3.71)$$

which for s -polarisation becomes

$$P_{z,int} = \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left[\frac{\gamma_1'^2 - \gamma_2'^2 + \gamma_2''^2}{(\gamma_1' + \gamma_2')^2 + \gamma_2''^2} \right] \left(\frac{\gamma_1'^2}{k_1^2} \right) \quad (3.3.72)$$

and for p -polarisation,

$$P_{z,int} = \epsilon_0 \tilde{\epsilon}_1 E_0^2 \cos \theta_1 \times \\ \times \frac{k_1^2(\gamma_2'^2 + \gamma_2''^2) + k_2^4 \cos^2 \theta_1 - 2\gamma_2'^2 k_2^2 - 4\beta^2 \gamma_2''^2 \cos^2 \theta_1}{k_1^2(\gamma_2'^2 + \gamma_2''^2) + k_2^4 \cos^2 \theta_1 + 2\gamma_1 \gamma_2' k_2^2 - 4\beta^2 \gamma_2''^2 \cos^2 \theta_1}. \quad (3.3.73)$$

Total forces on medium 2

The total force on medium 2 is the sum of the interface force and all the volume forces that act throughout its entirety. Since for a half-space the contribution from the far end is zero, we have

$$P_{x,tot} = -T_{xz}^{(1)}(0) \quad (3.3.74)$$

$$= \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left(\frac{\beta \gamma_1}{k_1^2} \right) (1 - |R_{\sigma\sigma}|^2), \quad (3.3.75)$$

and

$$P_{z,tot} = -T_{zz}^{(1)}(0) \quad (3.3.76)$$

$$= \frac{1}{2} \epsilon_0 \tilde{\epsilon}_1 E_0^2 \left(\frac{\gamma_1^2}{k_1^2} \right) (1 + |R_{\sigma\sigma}|^2). \quad (3.3.77)$$

3.3.7 Discussion and physical understanding of results

(a) *Scattering directions.* A homogeneous system can only scatter light into directions belonging to the scattering plane (the plane defined by $\mathbf{k}$ and $\hat{\mathbf{z}}$) so EM forces must lie on this plane. Also the polarisation of light is conserved upon scattering.

(b) *Natural limits.* The response to light incident on a homogeneous half-space ranges from full reflection (e.g. for a perfect metal at frequencies below ω_p) to full absorption (for a dielectric at the Brewster angle and p -polarised light). For normal incidence, the pressure on the half-space will range respectively from $2P_{inc}$ (full reflection) to P_{inc} (full absorption). These effects are demonstrated for GaP and Al samples in Figs. 3.3, 3.4 (solid black lines).

It is clear from these plots that the force on the system equals minus the net momentum exchange between the light and the system, just as one would expect from the action-reaction principle. Thus the explanation of forces in terms of the momentum-balance picture is valid. The values P_{inc} and $2P_{inc}$ constitute therefore the *natural limits* for *total* (i.e. external) EM forces on a general dielectric half-space due to incidence of a single beam of light.

(c) *Volume forces.* Inside the semi-infinite dielectric there will be no volume force in any direction if there are no losses, since the transmitted light will propagate unimpeded as in free space (although with a different wave velocity). Therefore

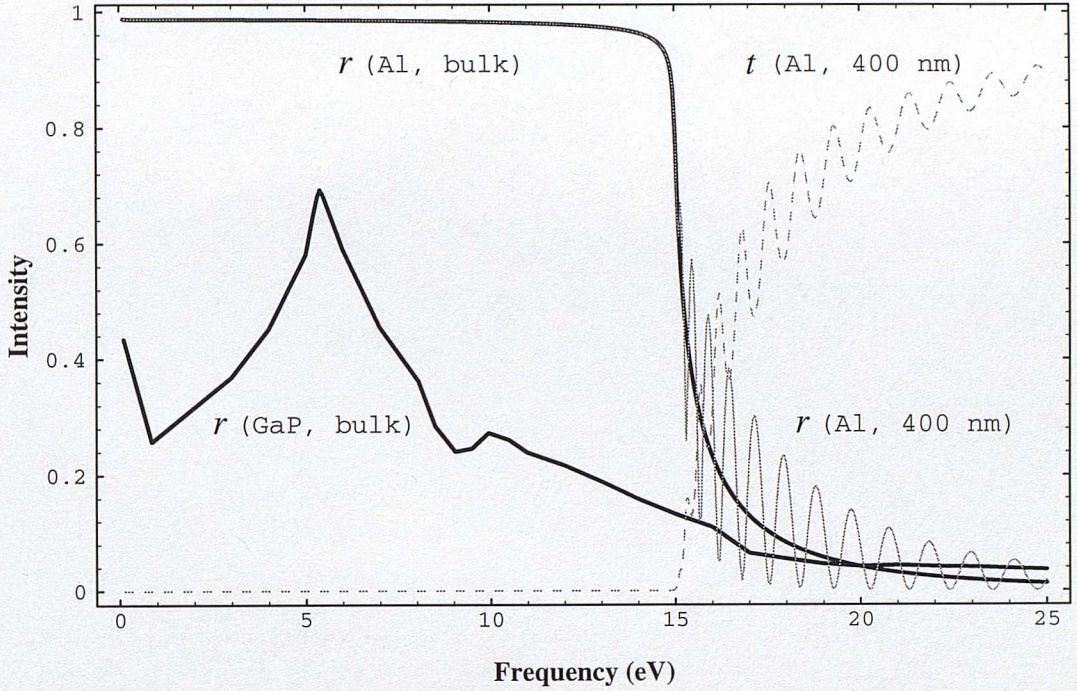


Figure 3.3: Intensity of reflected light from two homogeneous half-spaces (GaP and Al; black lines) and a homogeneous slab (400 nm of Al; grey lines) for normal incidence from air. For the slab the transmittance appears in broken grey lines. The ripple structure is due to multiple reflections from the sides of the slab (Fabry-Perot oscillations).

the stress tensor will be constant throughout the medium. However when absorption is present, volume forces occur and are accompanied by a continuous decay of the T_{ij} components with z . For a homogeneous system with very low absorption the volume force will be small compared to the interface force.

(d) *Origin of surface forces.* Momentum transfer between the incident light and matter can occur due to a scattering or absorption process. Therefore a surface force must generally exist on the boundary between dielectrics, since the reflection/transmission process causes the momentum of the incident light to change. This surface force will be accompanied by a discontinuity in one or more stress tensor components across the interface (T_{zz} in our case).

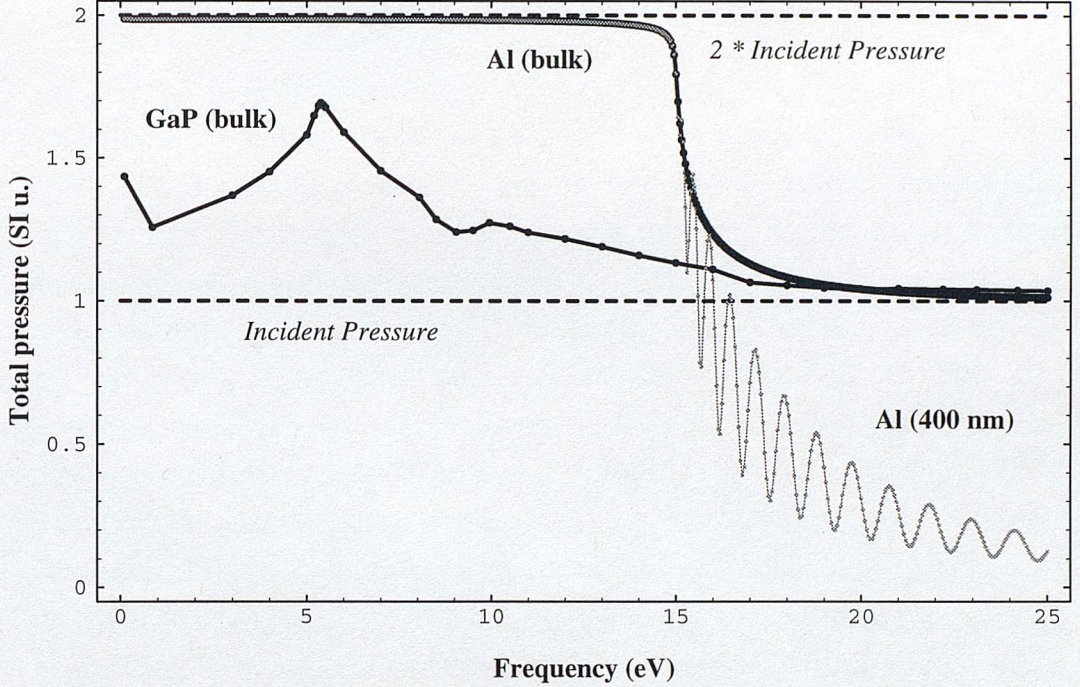


Figure 3.4: Light-induced pressure on a homogeneous half-space (GaP and Al; black curves) and a homogeneous slab (400 nm of Al; grey curves). Normal incidence from air along $\hat{z}$. The incident radiation pressure is $P_{inc} = I_0/c_0 = 1$. Positive values for the pressure imply that the light is pushing the system. (i) For the half-space, the pressure is calculated with the stress-tensor method analytically (solid black lines) and numerically (dotted black lines) in perfect agreement. It ranges from P_{inc} (full absorption) to $2P_{inc}$ (full reflection). (ii) In contrast, a slab of finite thickness may under certain conditions allow the light to pass through with transmittance ~ 1 , in which case it experiences a pressure close to zero. For the slab the pressure is calculated numerically with the stress-tensor method (solid grey line) and with the energy-gradient method (dotted grey line).

(e) *Sign of surface forces.* For $\epsilon_1 < \epsilon'_2$ and s polarisation the interface force is always negative (*i.e.* the interface is attracted by the beam), since

$$P_{z,int} \propto (\gamma_1^2 - \gamma_2'^2 + \gamma_2''^2) = (\epsilon_1 - \epsilon'_2). \quad (3.3.78)$$

For p -polarisation the algebra is complicated. To simplify matters we assume that there is no absorption in medium 2 ($\epsilon_2'' = 0$) and that total internal reflection is

not happening ($\gamma_2'' = 0$). Then for the interface force we have

$$P_{z,int} \propto (\epsilon_1 - \epsilon_2) [(\epsilon_2 - \epsilon_1) \sin^2 \theta_1 + \epsilon_2], \quad (3.3.79)$$

i.e. again for $\epsilon_1 < \epsilon_2'$ the interface force is always negative. (However it may also be negative for some range of values of $\epsilon_1, \epsilon_2, \theta_1$ even when $\epsilon_1 > \epsilon_2'$).

We can physically understand this in terms of the spatial distribution of the EM energy. For *s*-light $D_\perp = 0$ and therefore *all* components of $\mathbf{E}, \mathbf{B}$ and $\mathbf{H}$ are continuous across the interface (see Eq. (3.3.10)). The only discontinuity is in $D_\parallel$ which is higher in medium 2. Hence the energy density in medium 2 is always higher than in medium 1 at $z = 0$. Put in another way, the total energy is conserved, but the lower wave velocity in medium 2 means that more energy is ‘squeezed’ near the interface on the 0^+ side than on the 0^- side. Since the interface volume element is infinitely thin (and thus contains equal volumes of either medium) this imbalance in energy density shows up as a negative force. For *p*-light there is a nonzero $D_\perp$ which complicates matters so that the balance of the energy density is not straightforward to predict without doing the calculation; but otherwise the same arguments apply. This energy-gradient argument explains qualitatively the force, but for normal incidence it is quantitatively correct also. Thus the normal component of the interface pressure can be found from

$$P_{z,int} = -[\rho(0^+) - \rho(0^-)] \quad (3.3.80)$$

$$= -\frac{1}{4} \Re \left\{ (\mathbf{E} \cdot \mathbf{D}^* + \mathbf{H} \cdot \mathbf{B}^*) \Big|_{0^+} - (\mathbf{E} \cdot \mathbf{D}^* + \mathbf{H} \cdot \mathbf{B}^*) \Big|_{0^-} \right\} \quad (3.3.81)$$

$$= -\frac{1}{4} \Re \left\{ (\mathbf{E} \cdot \mathbf{E}^*) \Big|_{0^\pm} (\epsilon_2^* - \epsilon_1) \right\}, \quad (3.3.82)$$

where $0^\pm$ signifies that it does not matter if we evaluate $\mathbf{E} \cdot \mathbf{E}^*$ in either medium since it is continuous in the interface. We show in Fig. 3.5 the MST and the energy-gradient calculations for both Al and GaP. The agreement is excellent. The spatial variation of ρ across the interface is shown in Figs. 3.6, 3.7.

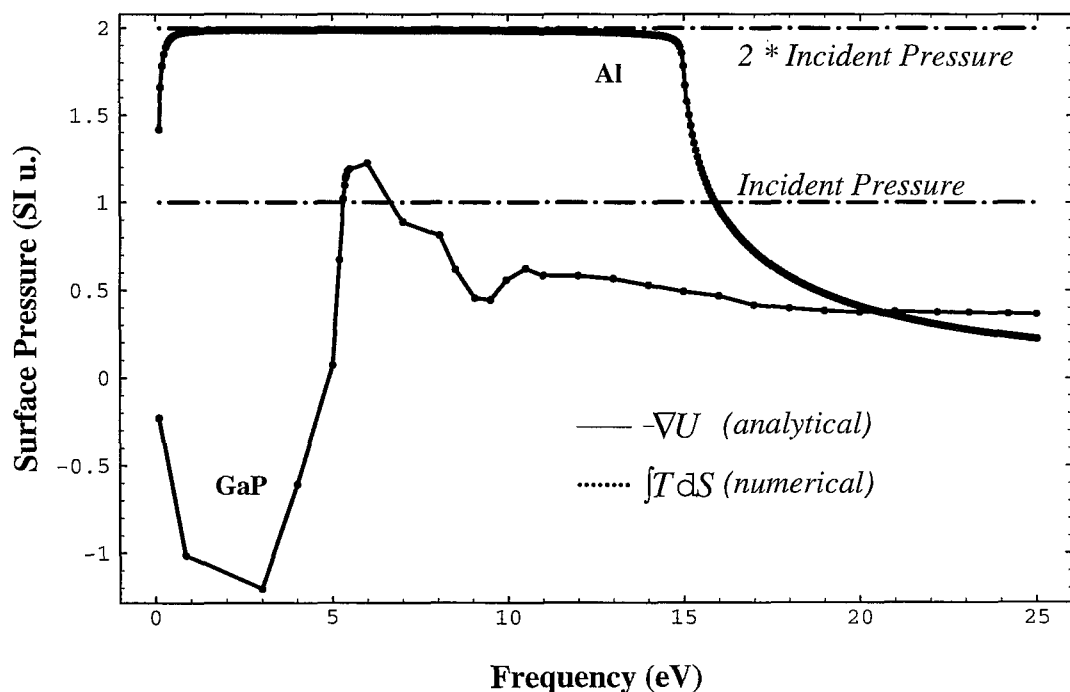


Figure 3.5: Light-induced interface pressure on a homogeneous half-space (GaP and Al). Normal incidence from air along $\hat{z}$. The pressure is calculated analytically with the energy-gradient method of Eq. (3.3.82) (solid black lines) and numerically with the stress-tensor method of Eq. (3.3.72) (dotted black lines) in perfect agreement.

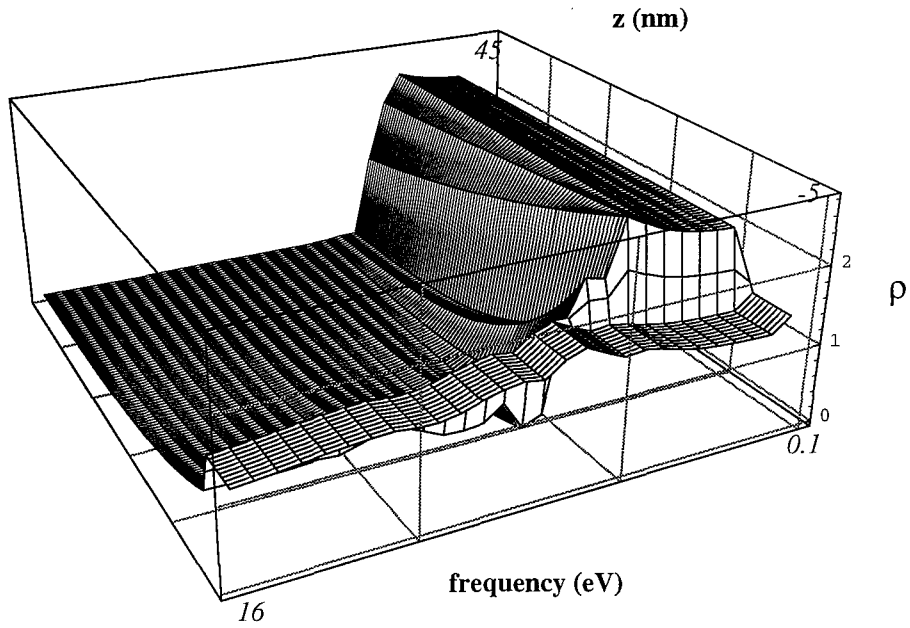


Figure 3.6: Spatial and frequency distribution of the energy density ρ for a half-space of GaP at normal incidence from air. In air ($z < 0$) ρ is spatially constant and varies only with ω due to the frequency-dependent ϵ_2 that affects the scattering. Inside GaP ($z > 0$) ρ varies as $e^{-2\gamma_2''z}$ as seen from Eq. (3.3.63). At low frequencies ρ rises across the interface, hence the force is negative.

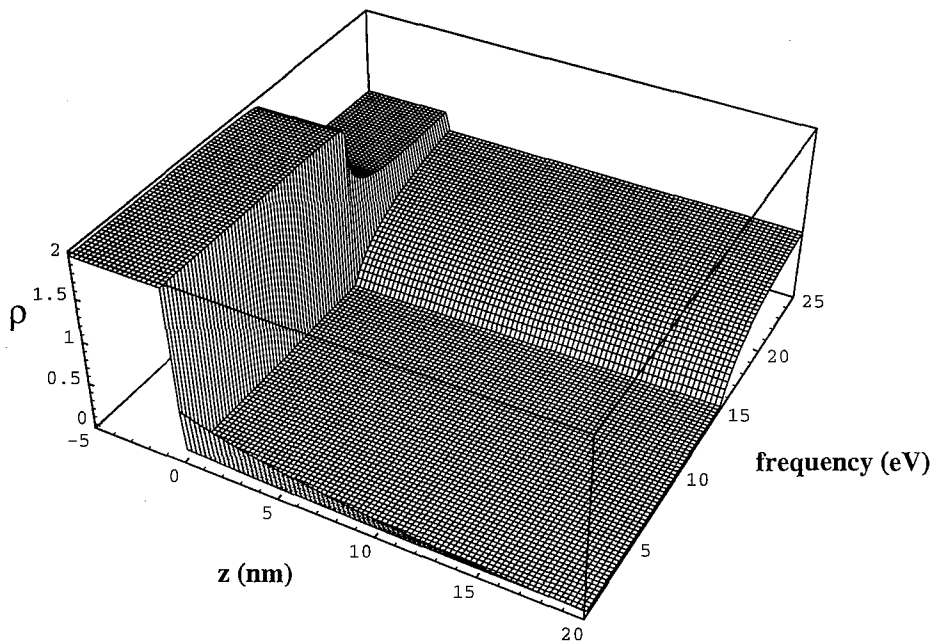


Figure 3.7: Spatial and frequency distribution of ρ for a half-space of Al at normal incidence from air. As with GaP, ρ is spatially constant in air ($z < 0$). Inside Al ($z > 0$) there are three regimes: (i) the high-absorption limit at $\omega < 0.2$ eV, where some light enters Al and ρ is strongly attenuated; (ii) the high-reflectivity limit at $0.2 \text{ eV} < \omega < \omega_p = 15 \text{ eV}$, where virtually no light enters the half-space, and (iii) the ‘transparency’ limit at $\omega > \omega_p$, where most light penetrates the interface and the absorption is low, so that the exponential decay of ρ is not noticeable over the distance shown here (20 nm). The interface force is always positive.

3.4 Analytical formulae: layer

Now we concentrate on the scattering properties of a non-magnetic, homogeneous dielectric layer of thickness d (Fig. 3.2). We analytically solve for s -polarised light.

3.4.1 Boundary conditions

At the two interfaces ($z = 0, z = d$), the boundary conditions are the same as those for the half-space.

3.4.2 Reflection coefficients

Light is incident from medium I, where it propagates with no absorption. However in media II, III absorption is possible, and we deal with the general case. The incident beam will multiply scatter off both interfaces as shown in Fig. 3.2. The total fields that result from this scattering mechanism are shown in Fig. 3.8: in region I there is the incident field travelling downwards with amplitude $\hat{A}$, and the superposition of all reflected beams travelling upwards with a complex amplitude $\hat{B}$. All the phase factors resulting from multiple scattering are intrinsic in $\hat{B}$. Similarly in II the total (steady-state) field can be split up in two counter-propagating fields with amplitudes $\hat{C}, \hat{D}$. Finally in III there is only a downwards travelling beam with amplitude $\hat{E}$, again incorporating all multiple reflections of light before it eventually enters medium III. This way of defining the problem simplifies the algebra significantly. First we checked that the above proposed fields satisfy all the equations of Maxwell in regions I, II, III. Then we used the BCs to find the amplitudes.

The resultant electric fields in region I, II, III respectively are

$$\mathbf{E}_1 = \hat{\mathbf{y}} e^{i(\beta x - \omega t)} \left[\hat{A} e^{i\gamma'_1 z} + \hat{B} e^{-i\gamma'_1 z} \right] \quad (3.4.1)$$

$$\mathbf{E}_2 = \hat{\mathbf{y}} e^{i(\beta x - \omega t)} \left[\hat{C} e^{i\gamma_2 z} + \hat{D} e^{-i\gamma_2 z} \right] \quad (3.4.2)$$

$$\mathbf{E}_3 = \hat{\mathbf{y}} e^{i(\beta x - \omega t)} \hat{E} e^{i\gamma_3 z}, \quad (3.4.3)$$

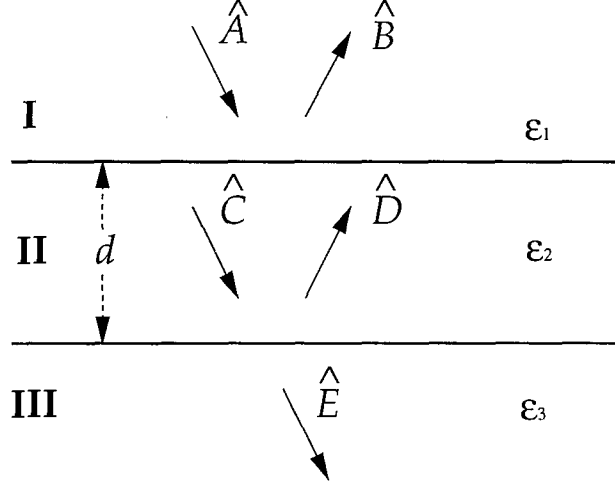


Figure 3.8: Method of calculating the fields for the slab system of Fig. 3.2. The incident and all the multiply reflected beams in region I are superposed, resulting in two waves (that propagate into and out of the slab) with complex amplitudes $\hat{A}, \hat{B}$. Similarly for region II there are two resultant waves with complex amplitudes $\hat{C}, \hat{D}$. In region III the superposition of all multiply transmitted beams leads to an outwards propagating wave with amplitude $\hat{E}$.

where γ_2, γ_3 are complex in general but γ'_1 is real. The magnetic fields are

$$\mathbf{H}_1 = \frac{e^{i(\beta x - \omega t)}}{\omega \mu_0} \left[-\hat{\mathbf{x}} \gamma'_1 (\hat{A} e^{i\gamma'_1 z} - \hat{B} e^{-i\gamma'_1 z}) + \hat{\mathbf{z}} \beta (\hat{A} e^{i\gamma'_1 z} + \hat{B} e^{-i\gamma'_1 z}) \right] \quad (3.4.4)$$

$$\mathbf{H}_2 = \frac{e^{i(\beta x - \omega t)}}{\omega \mu_0} \left[-\hat{\mathbf{x}} \gamma_2 (\hat{C} e^{i\gamma_2 z} - \hat{D} e^{-i\gamma_2 z}) + \hat{\mathbf{z}} \beta (\hat{C} e^{i\gamma_2 z} + \hat{D} e^{-i\gamma_2 z}) \right] \quad (3.4.5)$$

$$\mathbf{H}_3 = \frac{e^{i(\beta x - \omega t)}}{\omega \mu_0} \left[-\hat{\mathbf{x}} \gamma_3 + \hat{\mathbf{z}} \beta \right] \hat{E} e^{i\gamma_3 z}. \quad (3.4.6)$$

The boundary conditions become

$$\hat{A} + \hat{B} = \hat{C} + \hat{D} \quad (3.4.7)$$

$$\gamma'_1(\hat{A} - \hat{B}) = \gamma_2(\hat{C} - \hat{D}) \quad (3.4.8)$$

$$\hat{C} e^{i\gamma_2 d} + \hat{D} e^{-i\gamma_2 d} = \hat{E} e^{i\gamma_3 d} \quad (3.4.9)$$

$$\gamma_2(\hat{C} e^{i\gamma_2 d} - \hat{D} e^{-i\gamma_2 d}) = \gamma_3 \hat{E} e^{i\gamma_3 d}. \quad (3.4.10)$$

From these we can finally derive the amplitudes:

$$\hat{B} = \frac{(\gamma_2 - \gamma_3)(\gamma'_1 + \gamma_2) e^{2i\gamma_2 d} + (\gamma_2 + \gamma_3)(\gamma'_1 - \gamma_2)}{(\gamma_2 - \gamma_3)(\gamma'_1 - \gamma_2) e^{2i\gamma_2 d} + (\gamma_2 + \gamma_3)(\gamma'_1 + \gamma_2)} \hat{A} \quad (3.4.11)$$

$$\hat{C} = \frac{2\gamma'_1(\gamma_2 + \gamma_3)}{(\gamma_2 - \gamma_3)(\gamma'_1 - \gamma_2) e^{2i\gamma_2 d} + (\gamma_2 + \gamma_3)(\gamma'_1 + \gamma_2)} \hat{A} \quad (3.4.12)$$

$$\hat{D} = \frac{2\gamma'_1(\gamma_2 - \gamma_3) e^{2i\gamma_2 d}}{(\gamma_2 - \gamma_3)(\gamma'_1 - \gamma_2) e^{2i\gamma_2 d} + (\gamma_2 + \gamma_3)(\gamma'_1 + \gamma_2)} \hat{A} \quad (3.4.13)$$

$$\hat{E} = \frac{4\gamma'_1\gamma_2 e^{i(\gamma_2 - \gamma_3)d}}{(\gamma_2 - \gamma_3)(\gamma'_1 - \gamma_2) e^{2i\gamma_2 d} + (\gamma_2 + \gamma_3)(\gamma'_1 + \gamma_2)} \hat{A}. \quad (3.4.14)$$

The above expressions for the fields satisfy Maxwell's equations in all regions I, II and III. Furthermore the complex amplitudes $\hat{B}, \hat{C}, \hat{D}, \hat{E}$ are such that the BCs at each interface are also satisfied. Therefore Eqs. (3.4.1-3.4.6) together with Eqs. (3.4.11-3.4.15) provide the *unique* solution to our problem.

3.4.3 Maxwell Stress Tensor

Having solved for the fields in all regions we can now derive the Maxwell Stress Tensor. We obtain

$$T_{zz}^I = -\frac{1}{2}\epsilon_0\tilde{\gamma}_1'^2(|\hat{A}|^2 + |\hat{B}|^2) \quad (3.4.15)$$

$$T_{zz}^{II} = \frac{1}{2}\epsilon_0 \left[-\tilde{\gamma}_2'^2(|\hat{C}|^2 e^{-2\gamma_2''z} + |\hat{D}|^2 e^{+2\gamma_2''z}) + \right. \\ \left. + \tilde{\gamma}_2''^2(\hat{C}\hat{D}^* e^{2i\gamma_2'z} + \hat{C}^*\hat{D} e^{-2i\gamma_2'z}) \right] \quad (3.4.16)$$

$$T_{zz}^{III} = -\frac{1}{2}\epsilon_0\tilde{\gamma}_3'^2|\hat{E}|^2 e^{-2\gamma_3''z}. \quad (3.4.17)$$

In the particular case where medium II has no absorption ($\epsilon_2'' = 0$), the z dependence in T_{zz}^{II} disappears:

$$T_{zz}^{II} = \begin{cases} -\frac{1}{2}\epsilon_0\tilde{\gamma}_2'^2(|\hat{C}|^2 + |\hat{D}|^2), \gamma_2'' = 0 \\ +\frac{1}{2}\epsilon_0\tilde{\gamma}_2''^2(\hat{C}\hat{D}^* + \hat{C}^*\hat{D}), \gamma_2' = 0 \end{cases}. \quad (3.4.18)$$

3.4.4 Energy density

For the energy density ρ we find

$$\begin{aligned} \rho^I = \frac{1}{2}\epsilon_0 & \left[(\tilde{\gamma}_1'^2 + \tilde{\beta}^2)(|\hat{A}|^2 + |\hat{B}|^2) + \right. \\ & \left. + \tilde{\beta}^2(\hat{A}\hat{B}^* e^{2i\gamma_1'z} + \hat{A}^*\hat{B} e^{-2i\gamma_1'z}) \right] \end{aligned} \quad (3.4.19)$$

$$\begin{aligned} \rho^{II} = \frac{1}{2}\epsilon_0 & \left[(\tilde{\gamma}_2'^2 + \tilde{\beta}^2)(|\hat{C}|^2 e^{-2\gamma_2''z} + |\hat{D}|^2 e^{+2\gamma_2''z}) + \right. \\ & \left. + (-\tilde{\gamma}_2''^2 + \tilde{\beta}^2)(\hat{C}\hat{D}^* e^{2i\gamma_2'z} + \hat{C}^*\hat{D} e^{-2i\gamma_2'z}) \right] \end{aligned} \quad (3.4.20)$$

$$\rho^{III} = \frac{1}{2}\epsilon_0(\tilde{\gamma}_3'^2 + \tilde{\beta}^2)|\hat{E}|^2 e^{-2\gamma_3''z}. \quad (3.4.21)$$

3.4.5 Forces

We discuss only normal components of the forces (along $\hat{z}$).

Volume force in medium I

It is zero since T_{zz}^I is independent of position.

Volume force in medium II

It is nonzero only if there is absorption ($\epsilon_2'' \neq 0$):

$$P_{z,vol} = T_{zz}^{II}(z_f) - T_{zz}^{II}(z_i) \quad (3.4.22)$$

$$\begin{aligned} = \frac{1}{2}\epsilon_0 & \left\{ -\tilde{\gamma}_2'^2[|\hat{C}|^2 (e^{-2\gamma_2''z_f} - e^{-2\gamma_2''z_i}) + |\hat{D}|^2 (e^{+2\gamma_2''z_f} - e^{+2\gamma_2''z_i})] + \right. \\ & \left. + \tilde{\gamma}_2''^2[\hat{C}\hat{D}^* (e^{2i\gamma_2'z_f} - e^{2i\gamma_2'z_i}) + \hat{C}^*\hat{D} (e^{-2i\gamma_2'z_f} - e^{-2i\gamma_2'z_i})] \right\}. \end{aligned} \quad (3.4.23)$$

Volume force in medium III

It is nonzero only if there is absorption ($\epsilon_3'' \neq 0$):

$$P_{z,vol} = T_{zz}^{III}(z_f) - T_{zz}^{III}(z_i) \quad (3.4.24)$$

$$= \frac{1}{2} \epsilon_0 \tilde{\gamma}_3'^2 |\hat{E}|^2 (e^{-2\gamma_3'' z_i} - e^{-2\gamma_3'' z_f}). \quad (3.4.25)$$

Interface forces

At the upper interface ($z = 0$):

$$P_{z,int}|_{z=0} = T_{zz}^{II}(0) - T_{zz}^I(0), \quad (3.4.26)$$

and at the lower interface ($z = d$):

$$P_{z,int}|_{z=d} = T_{zz}^{III}(d) - T_{zz}^{II}(d). \quad (3.4.27)$$

Total force on medium I

The total pressure on medium I is

$$P_{z,tot} = T_{zz}^{II}(0) \quad (3.4.28)$$

$$= \frac{1}{2} \epsilon_0 \left[-\tilde{\gamma}_2'^2 (|\hat{C}|^2 + |\hat{D}|^2) + \tilde{\gamma}_2''^2 (\hat{C} \hat{D}^* + \hat{C}^* \hat{D}) \right]. \quad (3.4.29)$$

Total force on layer

The total pressure on the layer is

$$P_{z,tot} = T_{zz}^{III}(d) - T_{zz}^I(0) \quad (3.4.30)$$

$$= \frac{1}{2} \epsilon_0 \left[\tilde{\gamma}_1'^2 (|\hat{A}|^2 + |\hat{B}|^2) - \tilde{\gamma}_3'^2 |\hat{E}|^2 e^{-2\gamma_3'' d} \right]. \quad (3.4.31)$$

Total force on medium III

The total pressure on medium III is

$$P_{z,tot} = -T_{zz}^{II}(d) \quad (3.4.32)$$

$$= -\frac{1}{2} \epsilon_0 \left[-\tilde{\gamma}_2'^2 (|\hat{C}|^2 e^{-2\gamma_2'' d} + |\hat{D}|^2 e^{+2\gamma_2'' d}) + \right. \\ \left. + \tilde{\gamma}_2''^2 (\hat{C} \hat{D}^* e^{2i\gamma_2' d} + \hat{C}^* \hat{D} e^{-2i\gamma_2' d}) \right]. \quad (3.4.33)$$

Special case: medium II is absorption-free

We are interested primarily in forces that act on medium III across a dielectric layer of practically negligible absorption (say a vacuum layer). In this case $\epsilon_2'' = 2\gamma_2'\gamma_2'' = 0$ and the waves in medium II are either *travelling* (in which case $\gamma_2'' = 0$) or *evanescent* ($\gamma_2' = 0$). The crucial quantity that determines which of these two types of waves exists is the parallel momentum β . For large enough β the wavevector picks up an imaginary component along $\hat{\mathbf{z}}$:

$$\epsilon_2' = \beta^2 + \gamma_2'^2 ; \quad \mathbf{k}_2 = (\beta, 0, \gamma_2') , \quad \text{when } \beta^2 < \epsilon_2' \quad (3.4.34)$$

$$\epsilon_2' = \beta^2 - \gamma_2''^2 ; \quad \mathbf{k}_2 = (\beta, 0, i\gamma_2'') , \quad \text{when } \beta^2 > \epsilon_2' . \quad (3.4.35)$$

By choosing $\epsilon_2' < \epsilon_1'$ and adjusting θ_1 (since $\beta = k_1 \sin \theta_1$) appropriately, we can create an evanescent wave in region II with the desirable imaginary momentum γ_2'' . Now we are ready to obtain the forces.

Total force on III

$$P_{z,tot} = -T_{zz}^{II}(d) \quad (3.4.36)$$

$$= \begin{cases} +\frac{1}{2}\epsilon_0\tilde{\gamma}_2'^2(|\hat{C}|^2 + |\hat{D}|^2), \gamma_2'' = 0 \\ -\frac{1}{2}\epsilon_0\tilde{\gamma}_2''^2(\hat{C}\hat{D}^* + \hat{C}^*\hat{D}), \gamma_2' = 0 \end{cases} \quad (3.4.37)$$

$$= \begin{cases} +4\epsilon_0\tilde{\gamma}_2'^2[\gamma_1'^2(\gamma_2'^2 + |\gamma_3|^2)]|\hat{A}|^2/|\text{den}(\hat{B})|^2, \gamma_2'' = 0 \\ -4\epsilon_0\tilde{\gamma}_2''^2[\gamma_1'^2 e^{-2\gamma_2''d} \cdot (\gamma_2''^2 - |\gamma_3|^2)]|\hat{A}|^2/|\text{den}(\hat{B})|^2, \gamma_2' = 0, \end{cases} \quad (3.4.38)$$

where $\text{den}(\hat{B})$ is the denominator of the coefficient $\hat{B}$ (all $\hat{B}, \hat{C}, \hat{D}, \hat{E}$ have the same denominator).

Total force on I

$$P_{z,tot} = T_{zz}^{II}(0) \quad (3.4.39)$$

$$= \begin{cases} -\frac{1}{2}\epsilon_0\tilde{\gamma}_2'^2(|\hat{C}|^2 + |\hat{D}|^2), \gamma_2'' = 0 \\ +\frac{1}{2}\epsilon_0\tilde{\gamma}_2''^2(\hat{C}\hat{D}^* + \hat{C}^*\hat{D}), \gamma_2' = 0 \end{cases} , \quad (3.4.40)$$

which is equal in magnitude and opposite in direction to the total pressure on medium III.

Interface forces

At the upper interface ($z = 0$),

$$P_{z,int}|_{z=0} = T_{zz}^{II}(0) - T_{zz}^I(0) \quad (3.4.41)$$

$$= \begin{cases} \frac{1}{2}\epsilon_0 \left[-\tilde{\gamma}_2'^2 (|\hat{C}|^2 + |\hat{D}|^2) + \tilde{\gamma}_1'^2 (|\hat{A}|^2 + |\hat{B}|^2) \right], \gamma_2'' = 0 \\ \frac{1}{2}\epsilon_0 \left[\tilde{\gamma}_2''^2 (\hat{C}\hat{D}^* + \hat{C}^*\hat{D}) + \tilde{\gamma}_1'^2 (|\hat{A}|^2 + |\hat{B}|^2) \right], \gamma_2' = 0 \end{cases} \quad (3.4.42)$$

At the lower interface ($z = d$)

$$P_{z,int}|_{z=d} = T_{zz}^{III}(d) - T_{zz}^{II}(d) \quad (3.4.43)$$

$$= \begin{cases} -\frac{1}{2}\epsilon_0 \left[\tilde{\gamma}_3'^2 |\hat{E}|^2 e^{-2\gamma_3''d} - \tilde{\gamma}_2'^2 (|\hat{C}|^2 + |\hat{D}|^2) \right], \gamma_2'' = 0 \\ -\frac{1}{2}\epsilon_0 \left[\tilde{\gamma}_3'^2 |\hat{E}|^2 e^{-2\gamma_3''d} + \tilde{\gamma}_2''^2 (\hat{C}\hat{D}^* + \hat{C}^*\hat{D}) \right], \gamma_2' = 0 \end{cases} \quad (3.4.44)$$

$$= \begin{cases} -4\epsilon_0 [\tilde{\gamma}_1'^2 \gamma_2'^2 (\gamma_3'^2 - \gamma_3''^2 - \gamma_2'^2)] |\hat{A}|^2 / |\text{den}(\hat{B})|^2, \gamma_2'' = 0 \\ -4\epsilon_0 [\tilde{\gamma}_1'^2 \gamma_2''^2 (\gamma_3'^2 - \gamma_3''^2 + \gamma_2''^2) e^{-2\gamma_2''d}] |\hat{A}|^2 / |\text{den}(\hat{B})|^2, \gamma_2' = 0. \end{cases} \quad (3.4.45)$$

3.4.6 Discussion and physical understanding of results

We summarise our main findings for the forces on this system:

(a) *Natural limits revisited.* It is recalled that in the case of a half-space the two limits for reflectance are 1 and 0, leading to the natural limits for the pressure of $2P_{inc}$ and P_{inc} respectively. If we replace the half-space with a layer of finite thickness such that some light is transmitted through the far end, then the lower bound for the force becomes zero, corresponding to full transmission (i.e. zero reflection and zero absorption). These effects are demonstrated for normal incidence on an Al sample of thickness $d = 400$ nm in Figs. 3.3, 3.4 (grey lines). Therefore the three values ($0, P_{inc}$ and $2P_{inc}$) constitute the natural limits for *total*

(i.e. external) EM forces on a macroscopic body (homogeneous or not, finite or not) of arbitrary size and linear properties, upon the incidence of a single beam of light. The only way for these limits to be exceeded is when a resonance condition is fulfilled. We shall come to this interesting case when dealing with forces on dielectric crystals.

(b) *Energy-gradient for total forces.* The force calculation for normal incidence on the Al layer of Fig. 3.4 is done both with the stress-tensor method and also with the energy-gradient method yielding perfect agreement again, for total forces this time (compare with Eq. (3.3.80)):

$$P_{z,tot} = - \left[\rho(d^+) - \rho(0^-) \right]. \quad (3.4.46)$$

(c) *Interface forces at $z = d$.* With motivation for self-assembly, we are interested in producing attractive forces between media I and III. For the case of II being an absorption-free medium the waves in II may be either travelling ($\gamma_2'' = 0$) or evanescent ($\gamma_2' = 0$). We find that irrespective of whether the waves are travelling or evanescent, the interface force at $z = d$ is attractive (negative) when $(\epsilon_3' - \epsilon_2') > 0$. This is satisfied for at least some range of frequencies when II is vacuum and III is a semiconductor/insulator - but not if III is a metal. The same holds true for the interface force at $z = 0$ (having assumed $\epsilon_3'' = 0$ for algebraic simplicity): the sign of the force on the interface is given by $(\epsilon_2 - \epsilon_1)$. Consequently, any light beam passing through two liquid media separated by a layer of lower ϵ will induce an attraction between the liquid surfaces. If the liquids are incompressible their surfaces will move towards each other. Fig. 3.9 illustrates this point. The inverse situation occurs for light falling from air (medium I) onto a liquid layer (medium II), as shown in Fig. 3.10: the liquid expands in both interfaces. Such behaviour has been described by Kats and Kontorovich [68] in connection with the self-focusing of a laser beam in a liquid medium.

(d) *Total forces.* When only travelling waves exist in all three media, the two half-spaces repel each other (see upper part of Eq. (3.4.38)). In this case, the photons can be thought of as tennis balls: as they scatter off the walls of I and II, they exert a positive pressure on each wall. Hence two solid media I and III can never be attracted in this way (for hard solids it is the *total* forces that govern any overall attraction).

However, when $\mathbf{k}_2$ has a sufficiently large imaginary component along $\hat{\mathbf{z}}$,

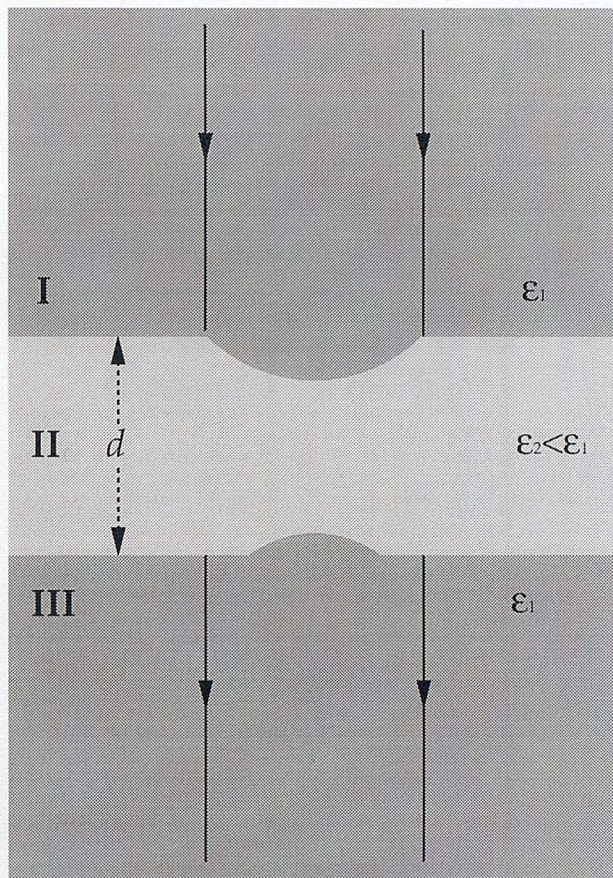


Figure 3.9: A light beam falling on a layer II separating two liquid media of higher ϵ . The light repels (attracts) the upper (lower) interface. The total force on the layer is positive, hence the higher degree of bending at the upper interface.

the force changes sign and becomes attractive (lower part of Eq. (3.4.38)). In this case the tennis-ball analogy breaks down. Our results indicate that at the electrostatic limit $\lambda \gg d$, the total force on III per unit area varies as

$$P_{z,tot}|_{\lambda \gg d} = \epsilon_0 \frac{\tilde{\gamma}_1^2 (|\gamma_3|^2 - \gamma_2''^2)}{|\gamma_1' + \gamma_3|^2} |\hat{A}|^2. \quad (3.4.47)$$

Thus the force is attractive for large angles θ_1 , repulsive for small angles. This is seen more clearly for the case of III being free of absorption, in which case the formulae simplify to give

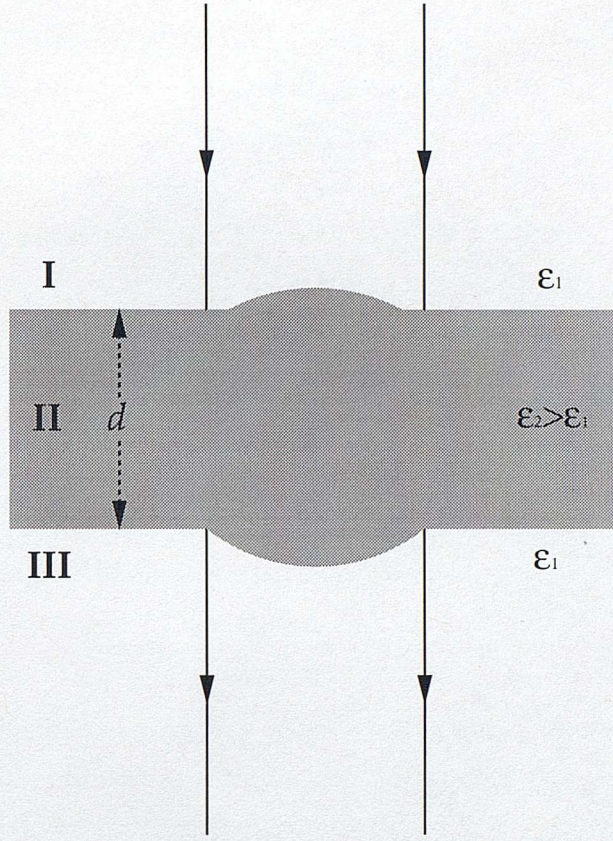


Figure 3.10: A light beam falling on liquid layer II from a medium of lower ϵ attracts (repels) the upper (lower) interface. The total force on the layer is positive, hence the higher degree of bending at the lower interface.

$$P_{z,tot} \sim (|\gamma_3|^2 - \gamma_2'^2) \quad (3.4.48)$$

$$= \begin{cases} (\epsilon_2' - \epsilon_3') & , \gamma_3' = 0 \\ \epsilon_3' + \epsilon_2' - 2\beta^2 & , \gamma_3'' = 0 \end{cases} \quad (3.4.49)$$

$$= \begin{cases} (\epsilon_2' - \epsilon_3') & , \gamma_3' = 0 \\ \epsilon_3' + \epsilon_2' - 2\epsilon_1' \sin^2 \theta_1 & , \gamma_3'' = 0. \end{cases} \quad (3.4.50)$$

When $\epsilon_1 > \epsilon'_3 > \epsilon'_2$ and θ_1 increases from 0 the wavefield in region II becomes evanescent first; then, at a larger value of θ_1 the wavefield in region III becomes evanescent also ('double evanescence'). At this point the force between media I and III becomes attractive and it remains so for all angles greater.

On the other hand, for a travelling wave in medium III the force also becomes attractive beyond a threshold for θ_1 . In Fig. 3.11 we plot the sign of T_{zz}^{II} versus ϵ'_1 and γ'_1 (recall that $\gamma'_1 = \sqrt{\epsilon'_1} \cos \theta_1$) for the system with II being vacuum, III being absorption-free and $\epsilon'_3 = \epsilon'_1$ for simplicity. The force is attractive for positive T_{zz}^{II} as seen from Eq. (3.4.32). It is clear that for $\epsilon'_1 < \epsilon'_2$ the force is repulsive (travelling waves in II), whereas for $\epsilon'_1 > \epsilon'_2$ the force becomes attractive for small enough γ'_1 (*i.e.* for large angles θ_1). Values of γ'_1 greater than $\sqrt{\epsilon'_1}$ are forbidden and are excluded from the plot (white area).

The force is zero when $|\gamma_3| = \gamma''_2$ (see Figs. 3.12, 3.13). Curiously, this is the same condition for the angles of refraction θ_2, θ_3 to be equal, since

$$|\tan \theta_i| = \frac{\beta}{|\gamma_i|}, \quad i = \{1, 2, 3\}. \quad (3.4.51)$$

When $\gamma''_2 < |\gamma_3|$ the force is repelling, and $\theta_2 > \theta_3$, just as in the case of a travelling wave scattering off a denser medium. On the other hand, when $\gamma''_2 > |\gamma_3|$, we have $\theta_2 < \theta_3$, which is reminiscent of a travelling wave scattering off a dense/rare interface; but at the same time the total force is attractive, a fact for which there is no analogue in waves of real $\mathbf{k}$.

The magnitude of the attractive force is sensitive to the ratio of the wavelength λ to the layer thickness d over which the evanescent fields decay before entering medium III. We find that for $\lambda/d \sim 5$ the force has already dropped by more than three orders of magnitude compared to its value at the electrostatic limit $\lambda \gg d$. In Fig. 3.14 we plot the normal pressure P_z versus $\cos \theta_1$ for three cases, (i) $\epsilon'_1 < \epsilon'_3$, (ii) $\epsilon'_1 = \epsilon'_3$ and (iii) $\epsilon'_1 > \epsilon'_3$. For (i) and (ii) the attractive part of P_z displays a smooth maximum, whereas for (iii) it peaks sharply. In all cases the highest value of P_z at the electrostatic limit is of the same order of magnitude as the incident radiation pressure. For (i) and (ii) the field in region III is that of a travelling wave for all angles and therefore $|\gamma_3| > 0$. However for (iii) the field in III is evanescent when $\theta_1 > \theta_0$, θ_0 being the angle for total internal reflection at an interface between media I and III[†]. At θ_0 we have $|\gamma_3| = 0$. From

[†]It is interesting to note that a sharp feature in P_z is present always at θ_0 , provided the wave

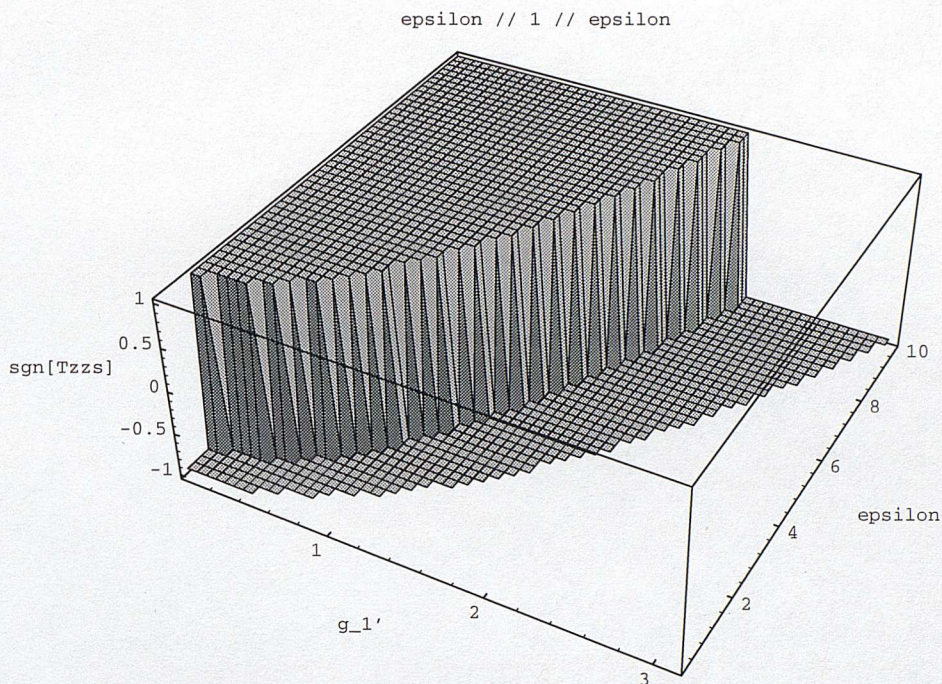


Figure 3.11: For the slab system of $\tilde{\epsilon}_1 = \tilde{\epsilon}$, $\tilde{\epsilon}_2 = 1$, $\tilde{\epsilon}_3 = \tilde{\epsilon}$ (no absorption in any medium) we plot the sign of T_{zz}^{II} vs. $\tilde{\epsilon}$ and $\tilde{\gamma}'_1 = (\tilde{\epsilon})^{1/2} \cos \theta_1$. There exists a region of the parameter space where $T_{zz}^{II} > 0$, causing the *total* light-induced force on medium III to be negative, and the total force on medium I to be positive. Hence the two half-spaces attract each other under the influence of light. Compare this situation with that of the *surface* forces (at $z = 0$ and $z = d$) which lead to attraction between the two interfaces as long as $\tilde{\epsilon} > 1$ (*i.e.* for *all* values of $\tilde{\gamma}'_1$), as explained in the text.

Eq.(3.4.47) it becomes clear why a vanishing γ_3 results in a sharp peak in P_z . For moderate absorption in III, $|\gamma_3|$ does not quite reach zero at θ_0 and the strength of the attraction is reduced although the peak is still clearly seen: for example, the maximum drops by 40 % when $\tilde{\epsilon}_3'' \sim 0.1$. Since the EM force is proportional to the light intensity I_0 , we can adjust the latter so that the force induced by the evanescent (or the travelling) wavefield supersedes the dispersion forces between the half-spaces. We have indicated the strength of the Van der Waals attraction

is evanescent in the layer. Its position on the θ_1 axis is independent of the dielectric properties of medium II, since the peak occurs when $|\gamma_3| = 0$ *i.e.* when $\epsilon'_3 = \beta^2 = \epsilon_1 - \gamma_1'^2$.

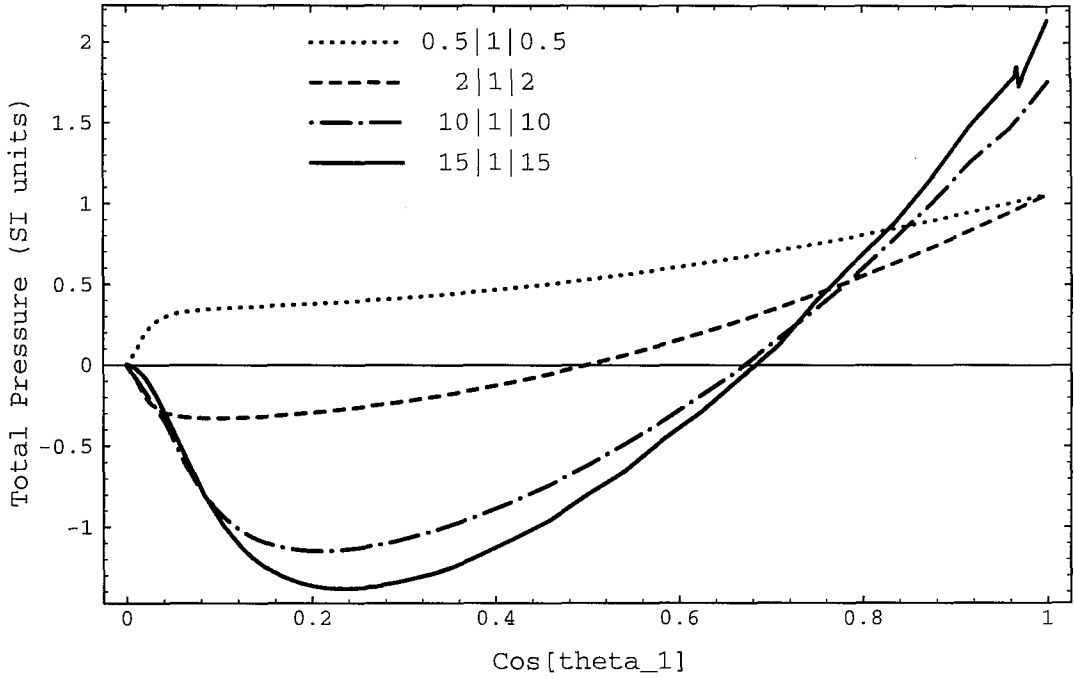


Figure 3.12: Total pressure on medium III vs. $\cos \theta_1$, for four systems of $\tilde{\epsilon}_1 = \tilde{\epsilon}$, $\tilde{\epsilon}_2 = 1$, $\tilde{\epsilon}_3 = \tilde{\epsilon}$ (no absorption): (i) $\tilde{\epsilon} = 0.5$, (ii) $\tilde{\epsilon} = 2$, (iii) $\tilde{\epsilon} = 10$, (iv) $\tilde{\epsilon} = 15$. For $\tilde{\epsilon} < 1$ the pressure is always repulsive (as professed by the previous graph). However for $\tilde{\epsilon} > 1$ there is attraction at large enough angles, the threshold-angle being a function of $\tilde{\epsilon}$.

in the figure, calculated in the macroscopic Lifshitz theory with retardation and non-additivity effects considered [69]. Owing to the d^{-4} behaviour of the Van der Waals force, only large enough separations will allow the photo-induced force to dominate.

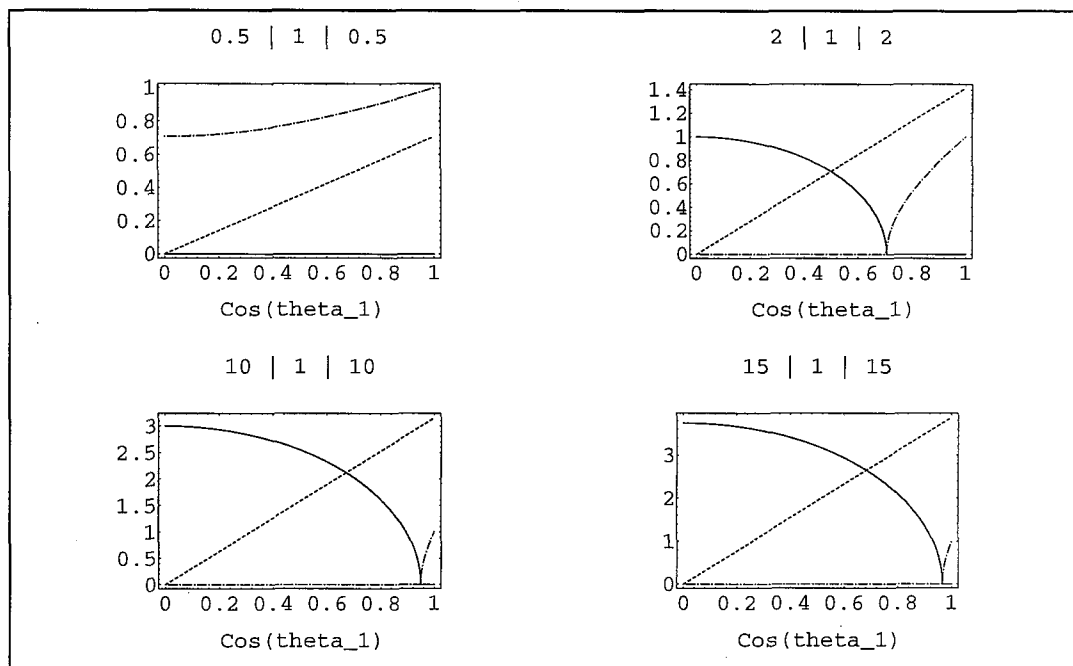


Figure 3.13: The wavevector arrangements for each of the four systems of the previous figure. The solid line denotes $\tilde{\gamma}_2''$, the dashed line is $\tilde{\gamma}_3'$ and the dash-dotted line is $\tilde{\gamma}_2'$. In all cases the angles for which $\tilde{\gamma}_2'' > \tilde{\gamma}_3'$ lead to attractive forces (see previous figure).

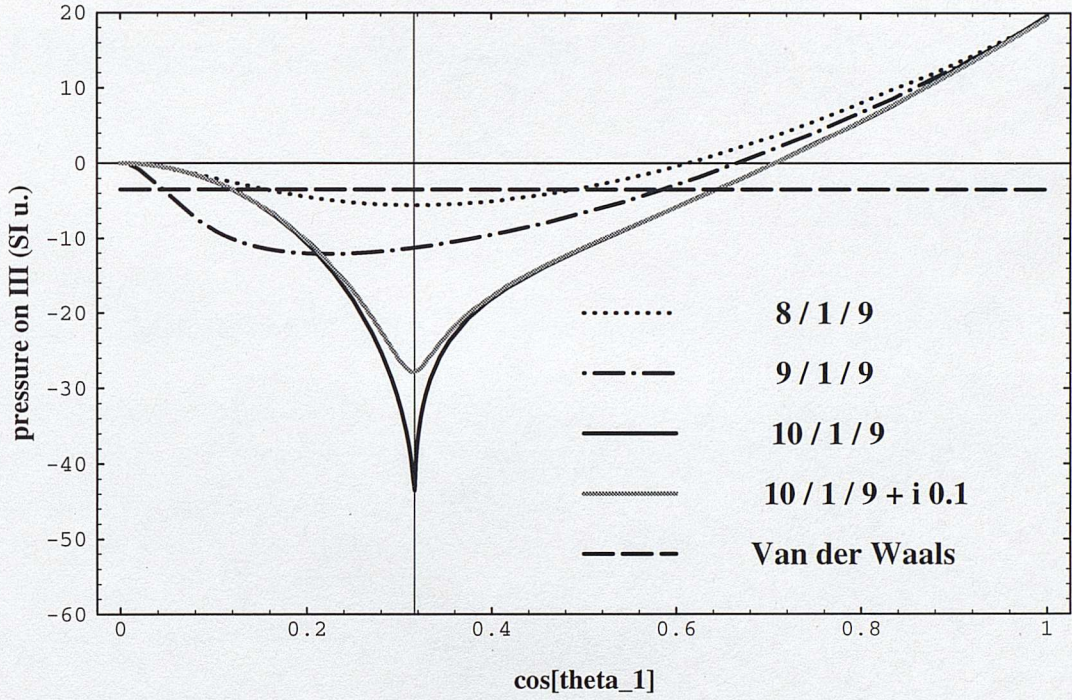


Figure 3.14: Total pressure on medium III for (i) $\tilde{\epsilon}_1 = 8, \tilde{\epsilon}_2 = 1, \tilde{\epsilon}_3 = 9$, (ii) $\tilde{\epsilon}_1 = 9, \tilde{\epsilon}_2 = 1, \tilde{\epsilon}_3 = 9$ and (iii) $\tilde{\epsilon}_1 = 10, \tilde{\epsilon}_2 = 1, \tilde{\epsilon}_3 = 9$. For system (iii) a nonzero absorption ($\tilde{\epsilon}_3 = 9 + i 0.1$) is considered also (grey line). The sharp attraction peak for (iii) occurs when $|\gamma_3| = 0$. Zero pressure corresponds to $\gamma_2'' = |\gamma_3|$. All curves are for $d = 100$ nm, $\lambda/d = 100$ and $I_0 = 3.5 \times 10^9$ W/m². The Van der Waals attraction between I and III is smaller than the photo-induced force for most angles; however for weaker light intensities or for separations smaller than 50 nm it dominates all photo-induced effects over all angles.

“I have not the slightest doubt that the main danger today is not from the rational act of responsible statesmen, but is due to essentially irrational acts of irresponsible, frightened, humiliated, revengeful or just mad people - or perhaps, more likely still, from the confused actions of well-meaning people overwhelmed by complex circumstances beyond their mental or moral ceiling.”

P M Blackett

Chapter 4

Dielectric crystals - non resonant effects

4.1 Dielectric and other properties of GaP

We are now ready to turn our attention to a crystal of dielectric (GaP) spheres arranged in a SC lattice. Since we will work extensively with GaP we give some of its main properties in Table 4.1.

Structure	zinc blende
Molecular mass	100.69
Average atomic mass	50.35
Density ρ	4.13 g/cm ³
Melting point	1750 K
Specific heat (typical value)	$\in [100,800]$ J/Kg·K
Microhardness	9450 N/mm ²
Debye temperature	446 K
Coef. linear thermal expansion	5.3×10^{-6} K ⁻¹
Thermal conductivity	752 mW/cm·K

Table 4.1: Main mechanical, thermal and molecular properties of GaP.

The relative permittivity of GaP has been modelled analytically by Barker [70]:

$$\tilde{\epsilon}(\nu) = 1 + \sum_{i=1}^3 \frac{S_{ei}\nu_{ei}^2}{\nu_{ei}^2 - \nu^2} +$$

$$+ \frac{S_0 \left(1 - \sum_{j=1}^2 S_j \right) \nu_0^2}{\nu_0^2 - \nu^2 + i\nu\gamma_0 - \nu_0^2 \sum_{j=1}^2 \frac{S_j \nu_j^2}{\nu_j^2 - \nu^2 + i\nu\gamma_j}} . \quad (4.1.1)$$

Eq. (4.1.1) has the form of a classical-oscillator equation, where S_i , ν_i^2 and γ_i^2 are the oscillator strength, restoring force and linewidth respectively, and the various parameters are shown in Table 4.2 (data from Parsons and Coleman [71]).

Electronic		Ionic	
$\tilde{\epsilon}(0)$	11.147	$\tilde{\epsilon}(\infty)$	9.091
S_{e1}	2.570	S_0	2.056
ν_{e1}	29,000 cm ⁻¹	ν_0	363.4 cm ⁻¹
S_{e2}	4.131	γ_0	1.1 cm ⁻¹
ν_{e2}	42,700 cm ⁻¹	S_1	7.0 × 10 ⁻⁴
S_{e3}	1.390	ν_1	349.4 cm ⁻¹
ν_{e3}	58,000 cm ⁻¹	γ_1	21 cm ⁻¹
		S_2	3.5 × 10 ⁻⁴
		ν_2	358.4 cm ⁻¹
		γ_2	12.6 cm ⁻¹

Table 4.2: Dielectric parameters for GaP at 300 K. $\tilde{\epsilon}(0)$ and $\tilde{\epsilon}(\infty)$ are the relative permittivities at zero and infinite frequency respectively.

The second term in the equation for $\tilde{\epsilon}$ is the electronic contribution and the third term the lattice contribution with frequency-dependent damping. The parameters have been fitted [71] so as to allow an accuracy for the real part of the refractive index n better than four figures from microwave to visible frequencies. In particular at infrared frequencies near the transverse optic mode (~ 0.045 eV) the absorptive part of $\tilde{\epsilon}$ becomes very large and the real part varies abruptly. We find that in the frequency range [0.1, 2] eV, $\tilde{\epsilon}'$ is reproduced to high accuracy whereas $\tilde{\epsilon}''$ is underestimated by less than 2 orders of magnitude (Fig. 4.1). Still, $\tilde{\epsilon}''$ is too small to have any sizeable effect on our force calculations, as we later show. Because $\tilde{\epsilon}'$ does not vary appreciatively at frequencies between 0.1 eV and 1.5 eV which are of interest to us, we shall regard it as constant from now on and model it as $\tilde{\epsilon}' = 8.9$ (marked by the horizontal line in the upper part of Fig. 4.1).

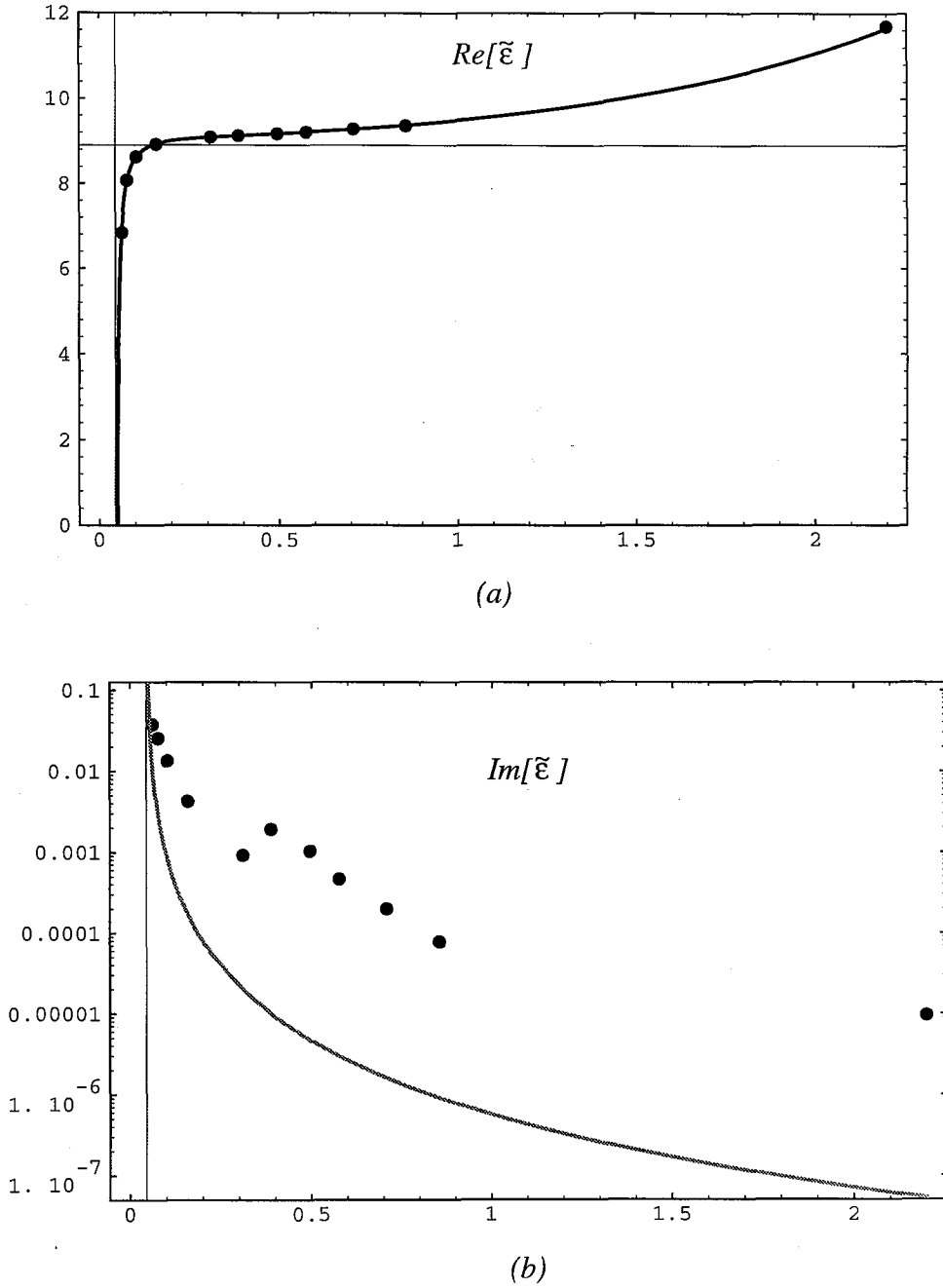


Figure 4.1: The relative permittivity of GaP, $\tilde{\epsilon} = \tilde{\epsilon}' + \tilde{\epsilon}''$ versus frequency. $\tilde{\epsilon}'$ is in the upper plot, $\tilde{\epsilon}''$ in the lower plot. Eq. 4.1.1 is plotted in solid line, dots are experimental results. Convergence is excellent for $\tilde{\epsilon}'$. For $\tilde{\epsilon}''$ the theoretical curve is wrong by less than 2 orders of magnitude for the frequencies shown.

4.2 System of Study

Our system is a SC crystal of spheres with a radius $r_s = 200 - 400$ nm with a lattice spacing $l_0 = 900$ nm, so the spherical surfaces are separated by $D_s = 100 - 500$ nm (Fig. 4.2). We often consider the reference medium in which the spheres are embedded to be air ($\tilde{\epsilon}_m = 1$), although, for reasons of experimental feasibility, it is really a liquid medium we have in mind. Such a medium does not alter qualitatively the effects we describe provided its absorption is low enough. For example, we show that water is suitable. Most of our results are for a discretisation mesh $10 \times 10 \times 10$. The crystal is infinite in the x and y directions, and has thickness N unit cells (layers) in the z direction.

For the EM properties of this crystal we expect the following behaviour:

- (a) Propagation of EM waves in an “effective medium” at large wavelengths $\lambda \gg l_0$ where light senses the crystal as an homogeneous dielectric medium.
- (b) Multiple scattering effects: *i*) appearance of forbidden ranges of frequencies as $\lambda \sim l$ where the multiply scattered waves interfere destructively and annihilate each other inside the crystal; *ii*) scattering by specific crystal planes into non-specular directions as $\lambda^{-1} \sim |\mathbf{k} + \mathbf{G}|/(2\pi)$, $\mathbf{G}$ being a reciprocal lattice vector.
- (c) Resonant features at frequencies corresponding to the EM eigenmodes of each sphere.

Each of the above types of behaviour is portrayed in the photonic band structure of Fig. 4.3. We shall begin by discussing the non-resonant effects (a) and (b) above; the very rich subject of resonances will be dealt with extensively in a chapter of its own.

4.3 Effective medium

We observe this behaviour in the band structure (dispersion relation) of Fig. 4.3.a and in Fig. 4.4, where the transmittance is shown alongside the pressure for a crystal monolayer, at normal incidence. At low energies ω is proportional to the wavevector k_z , just as in free-wave propagation, the constant of proportionality being the wave velocity of a homogeneous effective medium replacing the crystal. For a system of unconnected spheres like ours, the permittivity ϵ_{eff} of the effective

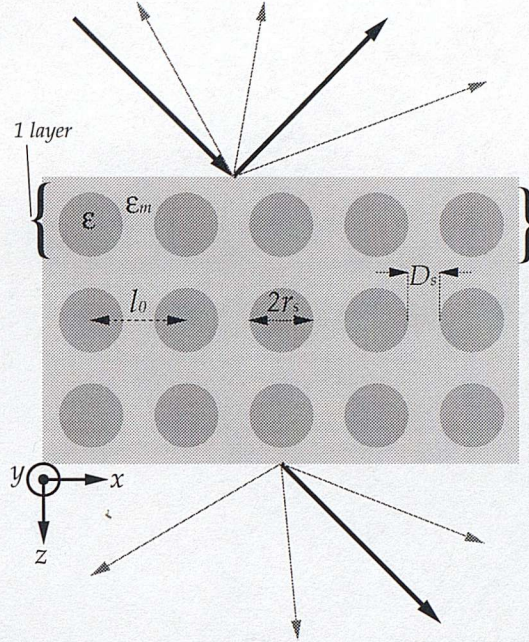


Figure 4.2: The dielectric crystal studied in chapters 4 and 5: A 3D crystal (of lattice spacing l_0) of GaP spheres (r_s, ϵ) embedded in a medium ϵ_m (usually $\tilde{\epsilon}_m=1$ here). The forces we study are induced by the incidence of one laser beam on the top surface. The crystal responds by reflecting light into the specular beam (black ray) and under special conditions into the Bragg-reflected beams (grey). Equivalently there is the directly-transmitted beam and possible Bragg-transmitted beams. Beams that result from Bragg scattering are also termed ‘off-diagonal’. The crystal is infinite in the xy plane and has thickness N layers (unit cells) in the z -direction.

medium is well approximated within the Garnett theory[72, 73],

$$\epsilon_{eff} = \frac{\epsilon_m(1 + 2\xi)}{1 - \xi}, \quad \xi = f \frac{\epsilon - \epsilon_m}{\epsilon + 2\epsilon_m}, \quad (4.3.1)$$

f being the volume fraction of the spheres in the crystal. For our system Eq. (4.3.1) yields $\epsilon_{eff} = 1.647$. We find that at energies up to 0.1 eV (or wavelengths $\lambda > 10 \times l_0$) the pressure on the crystal is approximately equal to the analytical expression for the force on a homogeneous film of the same thickness and

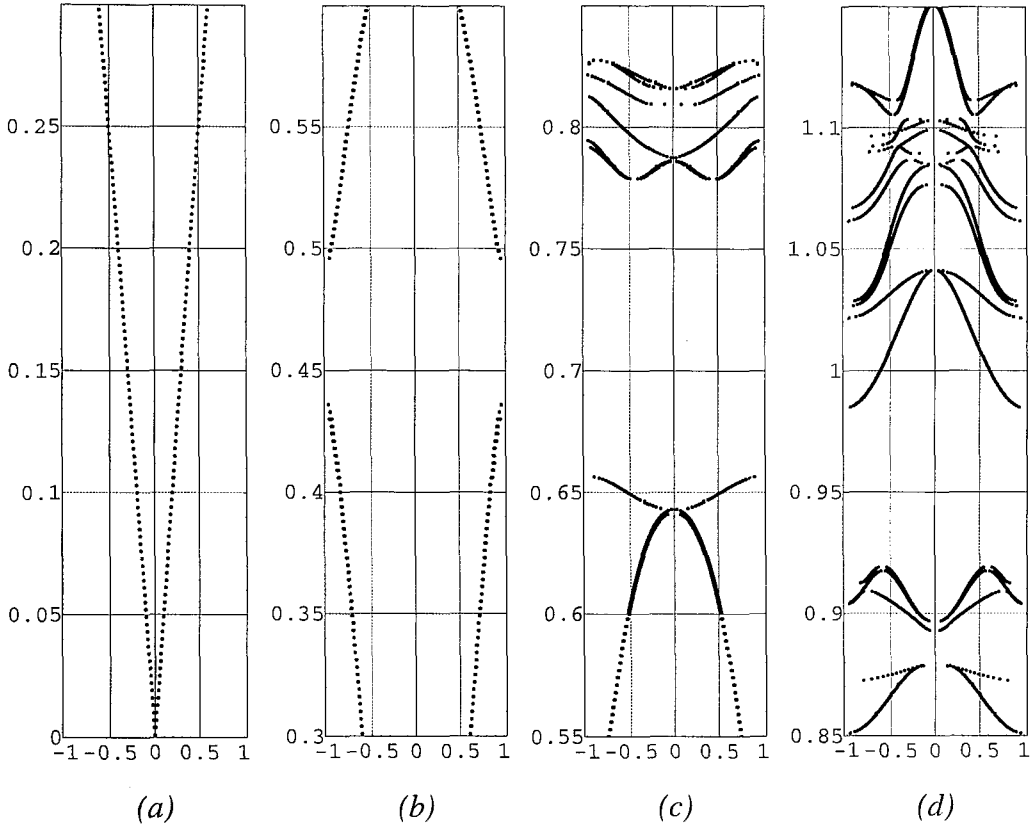


Figure 4.3: Photonic band structure along the ΓX direction for a simple cubic lattice of GaP spheres ($r_s = 350$ nm, $\tilde{\epsilon} = 8.9$) in air. ω is plotted vertically (in eV units), k_z horizontally (in multiples of π/l_0). The frequency range from 0 to 1.15 eV is scanned and shown in four parts. (a) 0-0.3 eV: light propagates as if in an homogeneous medium; (b) 0.3-0.6 eV: appearance of photonic band gaps; (c) 0.55-0.85 eV: appearance of resonant modes; (d) 0.85-1.15 eV: more resonant modes amid other bands and band gaps.

permittivity ϵ_{eff} :

$$P_{eff,z} = \frac{1}{2} \epsilon_0 E_0^2 \left[\tilde{\gamma}_1'^2 (1 + |R|^2) - \tilde{\gamma}_3'^2 |T|^2 e^{-2\gamma_3'' d} \right] \quad (4.3.2)$$

$$= \frac{I_0}{c_0} \frac{\gamma_1'^2}{\omega^2 \epsilon_0 \mu_0} \left[1 + |R|^2 - |T|^2 \right] \quad (4.3.3)$$

$$= \frac{I_0}{c_0} \frac{\gamma_1'^2}{\omega^2 \epsilon_0 \mu_0} 2|R|^2, \quad (4.3.4)$$

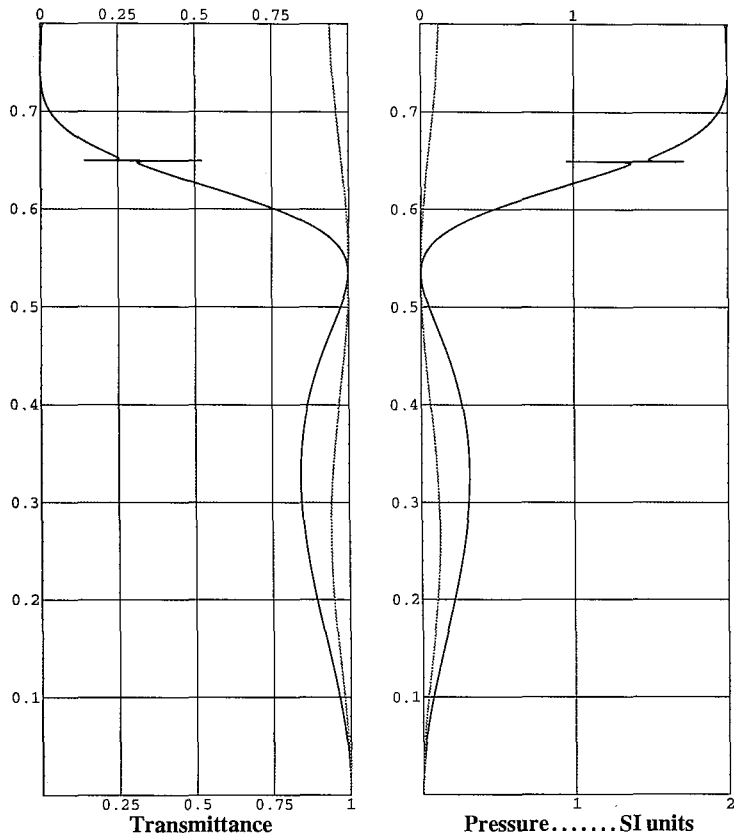


Figure 4.4: Frequency spectra of (a) transmittance and (b) normal pressure, for a monolayer of a dielectric crystal and normal incidence along $\hat{z}$ (solid black curves). For low ω the crystal can be replaced by an homogeneous slab of the same thickness and ϵ_{eff} which is given by Eq. 4.3.1; thus both spectra are reproduced reasonably well up to 0.1 eV (grey curves). At higher ω the effective medium approximation ceases to hold. This is especially true for the band gap frequencies.

where the symbols Eq. 4.3.2 are the same as in section 3.4.5. At energies higher than 0.1 eV the Garnett-type of effective medium theory loses out in accuracy, apart from capturing the unity-transmittance feature at ~ 0.54 eV which results from destructive interference between the singly and multiply reflected beams. The limitations of this effective-medium approach are especially evident in its failure to reproduce the force and transmission spectra for frequencies inside the second band gap *i.e.* at $\omega > 0.65$ eV (Fig. 4.4). To extend the effective-medium

concept to these frequencies we would need to generalise the Garnett method to finite wavelength as has been shown by Barrera and Fuchs [74]. Whatever the value of ϵ_{eff} , it will affect only the coefficient of the $|R|^2$ term in Eq. 4.3.4 and the total pressure will still be proportional to reflectance, even though the proportionality coefficient may be a function of frequency.

4.4 Multiple scattering

4.4.1 Band gaps

At higher energies band gaps open up and the propagation of all EM modes is forbidden inside an infinite crystal: all incident light will be reflected back when the frequency lies within a band gap. It is impressive that even for one layer of thickness the second band gap, centred at around 0.7 eV, causes a low transmission (see Fig. 4.4). Consequently the pressure on the layer approaches the upper natural limit $2P_{inc}$ (*i.e.* $\sim 2 \text{ N/m}^2$ for $I_0 \sim 3 \times 10^8 \text{ W/m}^2$) for frequencies inside this gap. The first band gap requires a much thicker sample (of 16 layers) before it displays maximum reflectance and force, but otherwise it affects the system's behaviour in the same way (see Fig. 4.7).

This ‘asymmetry’ in the readiness with which the first and second band gaps make their presence felt prompts us to wonder whether the two have different origins. The first gap is clearly a lattice effect. It forms at energies near the first crossing of 4 bands at the edge of the first Brillouin zone: out of these four, two bands (one for each polarisation) are associated with the reciprocal-lattice vector $\mathbf{G}_0 = 0$, and another two are associated with the $\mathbf{G}_1 = \frac{2\pi}{l_0} \hat{\mathbf{z}}$ vector. The degeneracy of the 4 bands leads to ‘interaction’, *i.e.* hybridisation and subsequent repulsion, and the gap opens up. Being a lattice effect, it is not surprising that it requires the stacking of a significant number of crystal layers in order to show up in the spectra.

The fact that the second gap causes almost zero transmittance even for one layer of thickness suggests that its origins are not to be found in the organisation of spheres in a lattice. It would be more plausible if this gap were associated with the onset of the resonance at 0.65 eV, which is a *single-sphere* effect and is present for any thickness. This point will be appreciated more when we thoroughly discuss Mie resonances in the following chapter.

4.4.2 Off-diagonal scattering

Another consequence of multiple scattering is that under certain conditions, the scattered fields can interfere constructively and produce outgoing waves in additional directions to those of the specularly reflected and directly transmitted beams. We call this scattering *off-diagonal*, since when it occurs the wavevector of the scattered field picks up a 2D reciprocal lattice vector component,

$$\mathbf{k}' = \mathbf{k}_{\parallel} + \mathbf{g}_{\parallel} + \hat{\mathbf{z}} k'_z. \quad (4.4.1)$$

Most often the energetics of the problem is such that k'_z is imaginary and all the off-diagonal fields are evanescent. However at certain ranges of frequencies a real k'_z is allowed and the off-diagonally scattered beams are travelling waves. For a SC lattice the minimum energy for this to occur is when $\mathbf{k}_{\parallel}$ lies at the edge of the first Brillouin zone along the ΓX direction: $k_{\parallel} = \pi/l_0$. At this frequency the empty-space dispersion relation allows a second beam with planar wavevector $-\mathbf{k}_{\parallel}$. Considering an incident beam with $\mathbf{k} = (k_x, 0, k_z)$, the condition becomes

$$k \sin \theta = \frac{\pi}{l_0} \Rightarrow \omega = k c_0 = \frac{\pi}{l_0} \frac{c_0}{\sin \theta}. \quad (4.4.2)$$

Eq. (4.4.2) gives the lowest frequency for off-diagonal scattering for a crystal of lattice constant l_0 and a wave incident at angle θ .

As one imagines, the effects on forces are rich as the possible combinations of momentum-carrying outgoing waves are endless. To give one example, consider the incidence at $\theta = 70^\circ$ of a laser beam on our crystal of one layer thickness. For this system we plot the intensity of reflection r and transmission t of each outgoing wave in Fig. 4.5. Off-diagonal scattering sets in for $\mathbf{g}_{\parallel} = (-2\pi/l_0, 0, 0)$ at frequency 0.71 eV in good agreement with Eq. (4.4.2). At slightly higher frequencies the extra energy is given to k'_z while k'_x is fixed to the value π/l_0 , provided the angle of incidence is not changed. Thus the outgoing waves include two reflected and two transmitted beams, all forming an angle θ with the surface normal (Fig. 4.6). In addition, notice that at 0.76 eV the intensities of the leftwards and the rightwards travelling waves are equal: $r_{\rightarrow} + t_{\rightarrow} = r_{\leftarrow} + t_{\leftarrow}$ (the incident beam is assumed rightwards travelling). This means that the total momentum of the scattered EM fields along $\hat{\mathbf{x}}$ is zero. In other words, the crystal absorbs all incident momentum along $\hat{\mathbf{x}}$, and the x -force on a surface S equals

$$F_x = (P_{inc} S) \sin \theta. \quad (4.4.3)$$

Our force calculations confirm that F_x approaches this value at $\omega \sim 0.76$ eV.

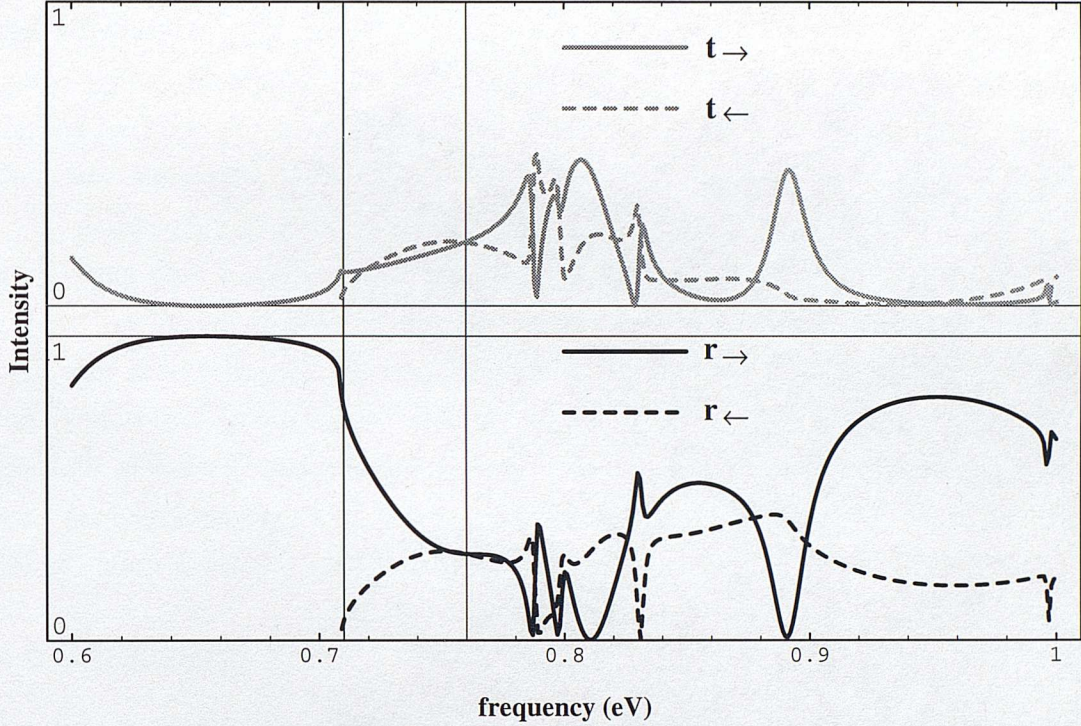


Figure 4.5: Onset of off-diagonal scattering from a crystal monolayer (GaP spheres of $r_s = 365$ nm, suspended in air). The incident beam is assumed travelling rightwards, as shown in the following figure, at an angle $\theta = 70$ deg to the surface normal. Shown are the intensity spectra of: the specularly reflected (solid black line), directly transmitted (solid grey line), off-diagonally reflected (dashed black line) and off-diagonally transmitted (dashed grey line) beams. For low ω , corresponding to wavevectors away from the edge of Brillouin zone, only the diagonal beams exist. However at the condition of Eq. (4.4.2) off-diagonal scattering starts to occur (at 0.71 eV). For the frequency range shown here only two off-diagonal beams exist.

Off-diagonal scattering survives a moderate amount of absorption in the crystal. We find that for $\tilde{\epsilon}'' = 0.1$ the reflectivities of the off-diagonal beams decrease by less than 40% (depending on the frequency), which is a little larger than the decrease the diagonal beams suffer ($< 20\%$).

4.5 Rationalisation of forces

4.5.1 Reflectance

The effective-medium theory for one crystal layer showed the force to be proportional to the reflectance $r(=|R|^2$ for a single reflected beam of coefficient R). If the media I and III are identical and absorption-free (not necessarily vacuum), then this result must always hold true, as Eqs. (4.3.2), (4.3.3), (4.3.4) profess.

A photonic crystal of spheres, arranged into many crystal layers is of course more than just homogeneous medium. It reflects light into several beams in general, each one with its own reflection coefficient. But whenever there is only one reflected and one transmitted beam, however complicated the reflection/transmission coefficients may be, we can regard the crystal as a homogeneous medium of some (equally complicated) effective ϵ , and expect the total force on it to be proportional to r , possibly with a frequency-dependent proportionality factor. Such is indeed the state of affairs in Fig. 4.7.

4.5.2 Energy

Even more telling than the reflectance is the energy. It was shown in the previous chapter that for normal incidence on a homogeneous layer, the force is found by simply taking the gradient of the EM energy, U , along the direction of propagation of the beam of light. The same holds true for dielectric crystals. For light incident along $\hat{z}$ the local force (normal component) is

$$F_z|_{loc} = -\frac{dU}{dz} . \quad (4.5.1)$$

The total (*i.e.* average) force on crystal of thickness d is

$$F_z|_{tot} = -\frac{U(d) - U(0)}{d} , \quad (4.5.2)$$

where $U(\zeta)$ is the average EM energy at depth ζ (*i.e.* averaged over all x, y points of the $z = \zeta$ surface of a unit cell). The total (*i.e.* average) pressure on the crystal is

$$P_{z,tot} = -\left[\rho(d) - \rho(0)\right] \quad (4.5.3)$$

where $\rho(\zeta)$ is the average EM energy density at depth ζ . In Fig. 4.8.a we plot P_z in SI units. It shares all the features of the force.

4.5.3 Momentum

It was pointed out earlier that another way of viewing forces is via the momentum-balance picture. In this picture the net force on a system can be attributed to the change of the net momentum entering the system. In our case we have an incident light beam bringing momentum $\mathbf{p}_{i;k}$ into the dielectric crystal, and the outgoing (reflected and transmitted) beams taking momentum out ($\mathbf{p}_{r;k'}$ and $\mathbf{p}_{t;k'}$ respectively). The net momentum given to the crystal is minus the momentum loss suffered by the light in a time-interval Δt ,

$$\mathbf{F}|_{tot} = -\frac{(\mathbf{p}_{r;k'} + \mathbf{p}_{t;k'}) - \mathbf{p}_{i;k}}{\Delta t}. \quad (4.5.4)$$

Formally the momentum enclosed in a volume element $\int d^3r$ may be found from the momentum density

$$\mathbf{p} = \int \mathbf{g} d^3r \quad (4.5.5)$$

$$= \int \frac{\mathbf{S}_P}{c^2} d^3r, \quad (4.5.6)$$

where $\mathbf{S}_P = \mathbf{E} \times \mathbf{H}$ is the Poynting vector. The subject of the EM momentum as such has attracted heated debate over the years, and is not generally regarded as settled, except when light travels in vacuum. Therefore we avoid performing the integral of Eq. (4.5.5); instead we confine ourselves to calculating $\mathbf{g}$ in vacuum. The total pressure then becomes

$$\mathbf{P}|_{tot} = -c_0 [(\mathbf{g}_{r;k'} + \mathbf{g}_{t;k'}) - \mathbf{g}_{i;k}] \quad (4.5.7)$$

$$= -\frac{1}{c_0} [(\mathbf{S}_{r;k'} + \mathbf{S}_{t;k'}) - \mathbf{S}_{i;k}]. \quad (4.5.8)$$

In our computer code we have calculated the right-hand side of Eq. (4.5.8), taking care to include only waves with real wavevectors $\mathbf{k}'$. The result appears in Fig. 4.8.b.

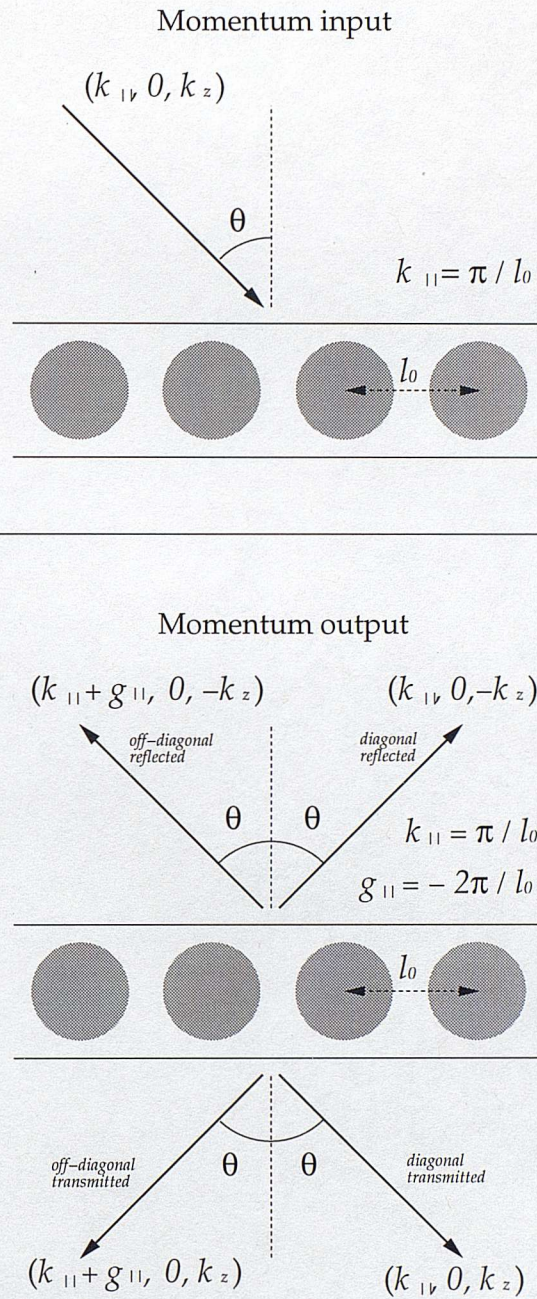


Figure 4.6: Off-diagonal scattering from a crystal monolayer at 70 deg incidence (s polarisation). The incident beam (upper panel) imports momentum into the crystal, the scattered waves (lower panel) export momentum away from it. The rightwards travelling scattered waves are diagonal, the leftwards travelling off-diagonal. At 0.76 eV, when the intensities of the waves travelling to the right and to the left balance each other (see Fig. 4.5), the total scattered x -momentum is zero; the crystal absorbs all incident momentum along $\hat{x}$.

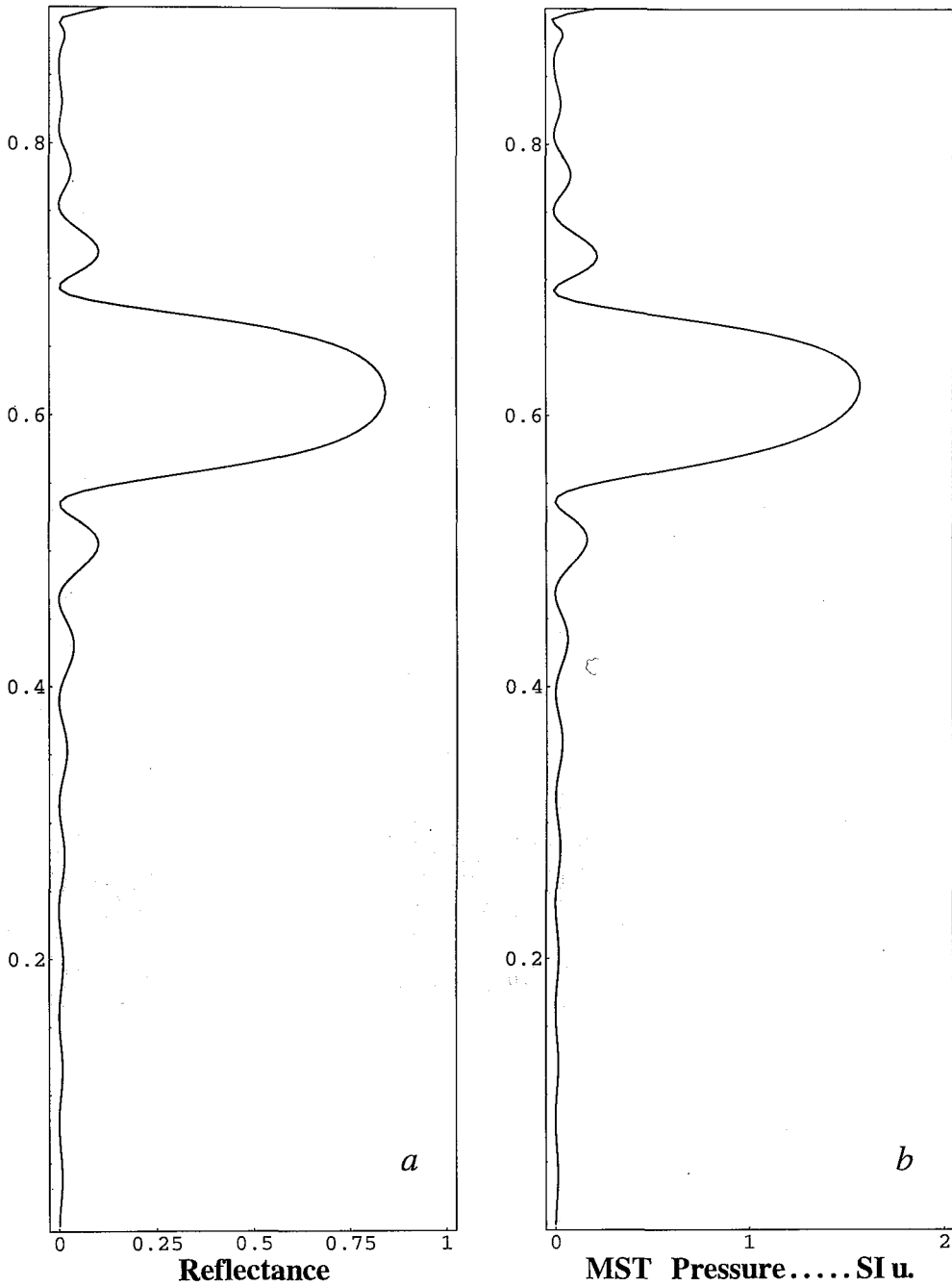


Figure 4.7: Reflectance (a) and normal force (b) for the system of Fig. 4.2 but with thickness of 8 layers. Normal incidence along $\hat{z}$. As the sample becomes thicker the reflectance slowly rises to unity inside the band gap.

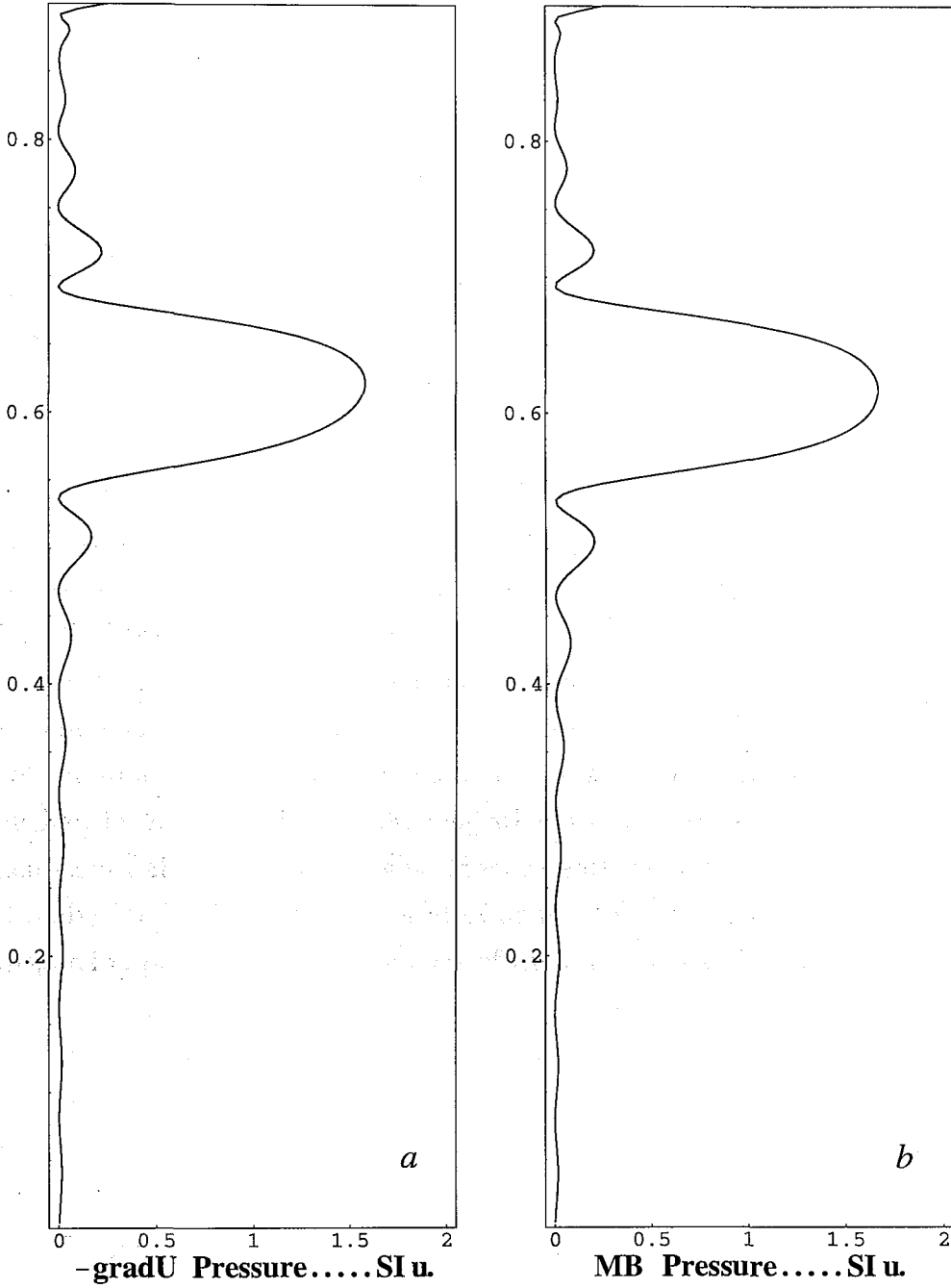


Figure 4.8: Normal pressure on the crystal of the previous figure, calculated via (a) the energy-gradient method, (b) the momentum-balance method.

“Philosophy requires a pure, clear, youthful mind and a zealous heart, elevated and noble sense and ardour. Every teacher who started from similar motives knows that this is enough for the young person to grasp the meaning of life.”

Ch. Theodorides

Chapter 5

Dielectric crystals - resonant effects

In this section we deal with the resonance effects identified as rather flat modes in the band structure (see parts (c),(d) in Fig. 4.3), combined with sharp features in the transmittance and force spectra (Fig. 5.5). As has been argued elsewhere [75, 76, 77, 83], these resonance peaks relate to the EM eigenmodes of isolated spheres (*Mie resonances*). Modes of similar nature (but corresponding to elastic waves in periodic composite materials) were identified by Kafesaki *et al* [78]. In the case of photonic crystals such resonant modes were first demonstrated (to our knowledge) by Zhang and Satpathy [30], whereas Stefanou *et al* [79] commented on their excitability by incident light. More recently they were studied by Ohtaka and Tanabe [75, 76, 77] who appropriately named them *heavy photons* due to their low dispersion behaviour. The presence of these resonances is known to strongly affect the position of the band gaps on the frequency axis [80, 81, 82]. Here we shall refer to the resonances that occur in the photonic crystal as ‘heavy-photon modes’, reserving the term ‘Mie modes’ for the resonant behaviour of *isolated* objects.

5.1 Mie scattering of an isolated sphere

In order to understand the heavy-photon modes and resonances in dielectric crystals, we describe how the Mie resonances [84] arise from the natural modes of the sphere. This discussion is based on the treatments of Stratton [85] and Bohren & Huffman [86].

5.1.1 Natural modes

Here we study the natural modes of an isolated sphere embedded in an infinite, homogeneous medium. These modes are solutions of Maxwell's equations incorporating spherical geometry and satisfying the appropriate boundary and limiting conditions.

A closed domain of a linear, homogeneous, isotropic material with no sources and dielectric properties ϵ, μ, σ_c can only support EM fields that satisfy the vector-wave equation which derives from the full Maxwell's equations [85]:

$$\nabla^2 \mathbf{C} - \mu_0 \mu \epsilon_0 \epsilon \frac{\partial^2 \mathbf{C}}{\partial t^2} - \mu_0 \mu \sigma_c \frac{\partial \mathbf{C}}{\partial t} = 0, \text{ where } \mathbf{C} = \{\mathbf{E}, \mathbf{B}, \mathbf{D}, \mathbf{H}\}, \quad (5.1.1)$$

and the operator ∇^2 implies

$$\nabla^2 \mathbf{C} = \nabla(\nabla \cdot \mathbf{C}) - \nabla \times (\nabla \times \mathbf{C}), \quad (5.1.2)$$

the rest of the symbols having their usual meanings. Due to the linearity of Eq. (5.1.1), a full solution for $\mathbf{C}$ can be constructed as the sum of many particular solutions, a fact that allows the arbitrary time evolution of $\mathbf{C}$ to be described in terms of harmonic solutions. Taking $e^{-i\omega t}$ as the time dependence of any such solutions, the wave equation becomes

$$\nabla^2 \mathbf{C} + k^2 \mathbf{C} = 0, \quad (5.1.3)$$

where $k^2 = \mu_0 \mu \epsilon_0 \epsilon \omega^2 + i \sigma_c \mu_0 \mu \omega$. Eq. (5.1.3) has three solutions,

$$\mathbf{L} = \nabla \psi, \quad \mathbf{M} = \nabla \times (\mathbf{a} \psi), \quad \mathbf{N} = \frac{1}{k} \nabla \times \mathbf{M}, \quad (5.1.4)$$

where ψ a solution of the *scalar* wave equation

$$\nabla^2 \psi + k^2 \psi = 0 \quad (5.1.5)$$

and $\mathbf{a}$ any constant vector of unit length. The function $\mathbf{L}$ is associated with a non-zero free-charge density. This can easily be seen when we decompose into Cartesian coordinates, whereby $\mathbf{L}$ provides the longitudinal-wave solution. For cases of interest to us there are no sources present and therefore the EM fields

are made up solely from the $\mathbf{M}, \mathbf{N}$ functions:

$$\mathbf{E} = \sum_n (a_n \mathbf{N}_n + b_n \mathbf{M}_n) \quad (5.1.6)$$

$$\mathbf{H} = -\frac{i k}{\omega \mu_0 \mu} \sum_n (a_n \mathbf{M}_n + b_n \mathbf{N}_n), \quad (5.1.7)$$

where n is a *general index* denoting particular solutions of the wave equation and the coefficients a_n, b_n remain to be determined. The particular solutions can be written down once the geometry suitable to the problem is chosen. In our case we choose spherical coordinates with origin at the centre of the dielectric sphere. The scalar-wave particular solutions of Eq. (5.1.5) are then

$$\psi_{\{e,o\}l,m_l} = z_l(kr) P_l^{m_l}(\cos \theta) e^{-i\omega t} \begin{cases} \cos(m_l \phi) \\ \sin(m_l \phi) \end{cases}. \quad (5.1.8)$$

In Eq. (5.1.8), $z_l(kr)$ are spherical Bessel's functions of the first kind ($j_l(kr)$) when the domain includes the origin (*i.e.* inside the sphere), of the third kind ($h_l^{(1)}(kr)$) for the space external to the sphere and extending to infinity. $P_l^{m_l}(\cos \theta)$ are the associated Legendre functions. The indices e and o of ψ denote even and odd dependence on ϕ respectively. Finally the principal (l) and azimuthal (m_l) quantum numbers provide information about the nodes with respect to the angles θ, ϕ respectively and take the values

$$\{l = 0, 1, 2, \dots, \infty\}, \quad \{m_l = 0, \pm 1, \pm 2, \dots, \pm l\}. \quad (5.1.9)$$

Since ψ are the generating functions for the vector solutions, the same indices apply to the latter, which therefore become the *vector spherical harmonic functions* $\mathbf{M}_{\{e,o\}l,m_l}, \mathbf{N}_{\{e,o\}l,m_l}$. We shall use the superscripts (1) and (3) with these solutions to denote the appropriate Bessel function.

How do the natural modes arise in an isolated sphere ? Out of the continuum of frequencies appearing in Eq. (5.1.8), only a discrete set survive when we require that the appropriate boundary conditions are met at the sphere surface, while demanding that the fields be finite at the origin and regular at infinity. These requirements also determine the mode coefficients a_n, b_n . In fact for each natural frequency the denominator of an expansion coefficient a_n or b_n becomes zero, and the corresponding vector spherical harmonic dominates. There are infinitely many frequencies at which a particular coefficient diverges, and they are

complex in general. For the a_n modes the frequencies are the solutions of the transcendental equation

$$\frac{[x h_n^{(1)}(x)]'}{h_n^{(1)}(x)} = \frac{\mu_s [m x j_n(mx)]'}{\mu_m m^2 j_n(mx)}, \quad (5.1.10)$$

where the derivatives are with respect to the variable x , and

$$x \equiv k_m r_s = \frac{2 \pi N_m r_s}{\lambda} \quad (5.1.11)$$

$$m \equiv \frac{k_s}{k_m} = \frac{N_s}{N_m}. \quad (5.1.12)$$

Here N is the refractive index and the subscripts 's' and 'm' refer to the sphere and the surrounding medium respectively. The quantity x is called the *order parameter*, and m is the *refractive-index contrast (ratio)*. The natural frequencies for the b_n modes satisfy

$$\frac{[x h_n^{(1)}(x)]'}{h_n^{(1)}(x)} = \frac{\mu_m [m x j_n(mx)]'}{\mu_s j_n(mx)}. \quad (5.1.13)$$

Due to the spherical symmetry the natural modes possess a $(2l + 1)$ degeneracy. The functions $\mathbf{M}_{\{e,o\}l,m_l}$ have the property that their radial component is zero. Hence, for the a_n modes, the $\mathbf{E}$ field has a radial component whereas the $\mathbf{H}$ field has not and is always perpendicular to the radius vector. There is therefore a distribution of oscillating electric charge on the surface of the sphere. For this reason these modes are called *transverse magnetic*, or of *electric type*, or *electric multipoles*. On the other hand, when only the b_n modes are excited, the fields are such as if they were produced by oscillating magnetic charges on the surface of the sphere. The natural modes are then called *transverse electric*, or of *magnetic type*, or *magnetic multipoles*. The electric modes have even dependence on ϕ , the magnetic modes odd. Thus the natural modes are completely identified by the following quantum numbers:

$$\left\{ \begin{array}{ll} a \text{ or } b & \text{(electric or magnetic multipoles)} \\ l & \text{(principal quantum number)} \\ m_l & \text{(azimuthal quantum number)} \end{array} \right. . \quad (5.1.14)$$

5.1.2 Mie modes

Here we study how an isolated sphere, embedded in an infinite, homogeneous medium responds to plane-wave radiation of frequency ω and field amplitude E_0 ,

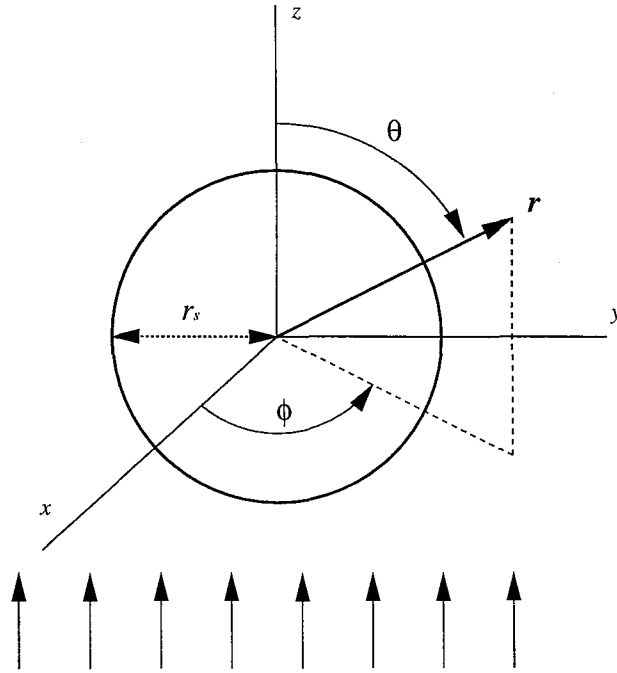


Figure 5.1: Geometry of the Mie scattering problem. A plane wave scatters off a sphere of radius r_s . Without loss of generality we consider the incidence to be along $\hat{z}$, and the incident (primary) field polarised along $\hat{x}$. Due to the spherical geometry the Maxwell's equations for the scattered (secondary) wavefields are solved in spherical polar coordinates r, θ, ϕ .

incident along $\hat{z}$ and polarised along $\hat{x}$ (Fig. 5.1). Analysed in spherical waves, the incident plane wave is rewritten as

$$\mathbf{E}_{inc} = \sum_{l=1}^{\infty} E_l (\mathbf{M}_{ol1}^{(1)} - i \mathbf{N}_{el1}^{(1)}) \quad (5.1.15)$$

$$\mathbf{H}_{inc} = -\frac{k_m}{\omega \mu_0 \mu_m} \sum_{l=1}^{\infty} E_l (\mathbf{M}_{el1}^{(1)} + i \mathbf{N}_{ol1}^{(1)}), \quad (5.1.16)$$

where $E_l = \frac{i^l E_0 (2l+1)}{l(l+1)}$. As the wave scatters off the sphere, the resultant field is the vector sum of the primary (incident) and secondary (scattered) fields. The solution can be expressed in terms of the vector spherical harmonics (since these form a complete and orthonormal set) and constructed in two parts, one for the interior and another for the exterior of the sphere. The solutions are then

matched at the boundary of the sphere. The presence of the plane wave incident along $\hat{\mathbf{z}}$ breaks the spherical symmetry and modifies the boundary conditions so that *only one* eigenmode for each l can be excited, the $m_l = 1$ mode[†]. The oscillations excited by the primary wave are not natural modes, since they are synchronous to the incident field. However they are of electric or magnetic type for the same reason as the natural modes. The coefficients a_n are still the amplitudes of oscillations of the electric type, while the b_n are the amplitudes of the magnetic type oscillations. At this stage we can identify the general index n of the a_n, b_n coefficients with the whole set of allowed indices:

$$a_n \equiv a_{\{l, m_l=1, e\}} \equiv a_l \quad (5.1.17)$$

$$b_n \equiv b_{\{l, m_l=1, o\}} \equiv b_l, \quad (5.1.18)$$

where the right-hand notations are used for simplicity from now on. The fields scattered by the sphere are

$$\mathbf{E}_{sca} = \sum_{l=1}^{\infty} E_l (i a_l \mathbf{N}_{el1}^{(3)} - b_l \mathbf{M}_{ol1}^{(3)}) \quad (5.1.19)$$

$$\mathbf{H}_{sca} = \frac{k_m}{\omega \mu_0 \mu_m} \sum_{l=1}^{\infty} E_l (i b_l \mathbf{N}_{ol1}^{(3)} + a_l \mathbf{M}_{el1}^{(3)}). \quad (5.1.20)$$

The coefficients a_l, b_l depend only on x, m and are given in Appendix C. Similarly, for the internal fields,

$$\mathbf{E}_{int} = \sum_{l=1}^{\infty} E_l (c_l \mathbf{M}_{ol1}^{(1)} - i d_l \mathbf{N}_{el1}^{(1)}) \quad (5.1.21)$$

$$\mathbf{H}_{int} = -\frac{k_s}{\omega \mu_0 \mu_s} \sum_{l=1}^{\infty} E_l (d_l \mathbf{M}_{el1}^{(1)} + i c_l \mathbf{N}_{ol1}^{(1)}), \quad (5.1.22)$$

where the coefficient c_l (d_l) has the *same denominator* as b_l (a_l), the roots of which are the natural frequencies for magnetic (electric) multipoles, as given by Eq. (5.1.13) (Eq. (5.1.10)) above. The natural frequencies are complex, however if their imaginary part is small and the frequency of the incident plane-wave approaches the real part of the natural frequency, a resonance phenomenon will occur. This is a Mie resonance. As ω of the incident field spans all values, the denominator of each coefficient a_l, b_l will become zero many times. In order

[†]Only one mode per polarisation is allowed; but since there are two independent polarisations of the incident field, it follows that a total of two modes are excitable by plane-polarised light.

to discriminate between such like-natured resonances, we introduce a further ‘quantum number’, the sequential order of each resonance $i = \{1, 2, \dots\}$. Hence for each Mie resonance of electric type we use the notation a_l^i , whereas for magnetic type we use b_l^i .

We show in Table 5.1 the natural frequencies for a sphere of $r_s = 365$ nm and $\tilde{\epsilon}_s = 8.9$ in air ($\tilde{\epsilon}_m = 1$). Since the Eqs. (5.1.10), (5.1.13), (5.1.12) for the natural frequencies ω_n depend only on m and on $x \propto \Re\{\omega_n\} \cdot r_s$, it follows that the frequencies of any-sized sphere of $\tilde{\epsilon}_s = 8.9$ can be deduced from this table.

As can be seen from the form of the vector spherical harmonic functions (see Appendix C), the scattered wavefield consists of spherical waves with *elliptical polarisation* in general. Therefore when the scattered fields are decomposed into plane waves (as is done in the OPAL codes) the polarisation of the primary radiation will not be conserved. We shall confirm this to be the case when we study Mie resonances in dielectric crystals (Figs. 5.6, 5.7).

A very useful quantity in understanding the properties of the different Mie resonances is the *scattering cross section*, defined as the ratio of the total scattered energy per unit time to the intensity of the incident radiation (incident energy per unit time per unit surface)

$$C_{sca} = \frac{W_{sca}}{I_0} \quad (5.1.23)$$

$$= \int \mathbf{S}_{sca} \cdot d\hat{\mathbf{e}}_\theta \times d\hat{\mathbf{e}}_\phi \quad (5.1.24)$$

$$= \int S_{sca,r} r^2 \sin \theta d\theta d\phi, \quad (5.1.25)$$

where the surface integration is over a closed spherical shell of radius $r > r_s$ concentric to the sphere, and the scattered energy flow per unit time is given by the Poynting flux vector $\mathbf{S}_{sca}$ (radial part $S_{sca,r}$):

$$S_{sca,r} = \frac{1}{2} \Re\{(\mathbf{E}_{sca} \times \mathbf{H}_{sca}^*) \cdot \frac{\hat{\mathbf{e}}_r}{|\hat{\mathbf{e}}_r|}\} \quad (5.1.26)$$

$$= \frac{1}{2} \Re\{E_{sca,\theta} H_{sca,\phi}^* - E_{sca,\phi} H_{sca,\theta}^*\}. \quad (5.1.27)$$

In the case of absorption present in the sphere ($\epsilon_s'' \neq 0$), the combined effects of scattering and absorption lead to *extinction*, and the relevant cross section is

$$C_{ext} = \frac{W_{ext}}{I_0} \quad (5.1.28)$$

$$= \int S_{ext,r} r^2 \sin \theta d\theta d\phi, \quad (5.1.29)$$

Type of mode n	$\Re\{\omega_n\}$ (eV)	$\Im\{\omega_n\}$ (eV)	$\Re\{\omega_n\} \cdot r_s$ (eV $\times$ nm)	$\lambda_{n,m}$ (nm)	$\lambda_{n,s}$ (nm)
b_1^1	0.512244	0.012107	187	2414	809
a_1^1	0.588025	0.085604	214	2103	705
b_2^1	0.733065	0.005524	267	1687	565
a_2^1	0.859172	0.033103	313	1439	482
b_3^1	0.948930	0.002098	346	1303	436
a_3^1	1.109970	0.011996	405.1	1114.4	373.5
b_1^2	1.110400	0.017353	405.3	1114	373.4
a_1^2	1.124760	0.205135	410	1099	368
b_4^1	1.160300	0.000697	423	1066	357
a_4^1	1.346380	0.003945	491	918	307
b_2^2	1.353710	0.013234	493	913	306
b_5^1	1.368030	0.000211	499	904	303
a_2^2	1.391720	0.123184	507	888	297
a_5^1	1.572220	0.001196	573	786.8	263.7
b_6^1	1.573000	0.000060	574	786.4	263.6
b_3^2	1.589580	0.008955	579	778	260

Table 5.1: Mie modes of an isolated GaP sphere ($\tilde{\epsilon}_s = 8.9, r_s = 365$ nm) in air. b_l^i (a_l^i) modes represent magnetic (electric) multipoles oscillating at the light frequency. The complex eigenfrequencies ($\Re\{\omega_n\}$, $\Im\{\omega_n\}$) are shown in eV. $\Im\{\omega_n\}$ is a measure of how strongly damped each oscillation is. The product ($\Re\{\omega_n\} \cdot r_s$) is proportional to the order parameter x and is independent of the radius r_s for each mode n , allowing the eigenfrequencies for any-sized GaP sphere to be calculated. The resonant wavelengths in the reference medium ($\lambda_{n,m}$) and inside the sphere ($\lambda_{n,s} = \lambda_{n,m}/\tilde{\epsilon}_s^{1/2}$) are shown in nm.

where $S_{ext,r}$ involves the products of the scattered and incident fields:

$$S_{ext,r} = \frac{1}{2} \Re\{E_{inc,\phi} H_{sca,\theta}^* - E_{inc,\theta} H_{sca,\phi}^* + E_{sca,\phi} H_{inc,\theta}^* - E_{sca,\theta} H_{inc,\phi}^*\}. \quad (5.1.30)$$

It can be shown that

$$C_{sca} = \frac{2\pi}{k_m^2} \sum_{l=1}^{\infty} (2l+1) (|a_l|^2 + |b_l|^2) \quad (5.1.31)$$

and

$$C_{ext} = \frac{2\pi}{k_m^2} \sum_{l=1}^{\infty} (2l+1) \Re\{(a_l + b_l)\} \quad (5.1.32)$$

$$= C_{sca}, \text{ for } \epsilon_s'' = \epsilon_m'' = 0. \quad (5.1.33)$$

The usefulness of the cross sections C_{sca}, C_{ext} lies in that it allows visualisation of the following properties of the Mie resonances: location (in the frequency axis), strength, resolution. Since the resonances affect strongly the EM forces on the sphere (and on photonic crystals of spheres) the cross sections are of high relevance to our work.

Below (Fig. 5.2) we see the scattering cross section C_{sca} for the sphere of Table 5.1 ($\tilde{\epsilon}_s = 8.9, r_s = 365$ nm in air). All peaks are associated with the resonances of the table. The sharpest peaks are the ones with smallest $\Im\{\omega_n\}$ (*e.g.* b_6^1, b_5^1, b_4^1). However not all resonances are visible, either due to damping *i.e.* large $\Im\{\omega_n\}$ (as in a_1^2, a_2^2 , which have the largest $\Im\{\omega_n\}$), or due to close proximity with other resonances in the frequency axis ($\{a_3^1, b_1^2\}$ and $\{a_5^1, b_6^1\}$ appear as a single peak; b_2^2 is masked by the nearby sharper peaks of a_4^1 and b_5^1). The peak height is indicative of the resonance phenomenon taking place: C_{sca} is up to 10 times the physical cross section $\pi r_s^2 \sim 0.4 \mu\text{m}^2$.

Compare Fig. 5.2 with the plot of the *radiation pressure* on a GaP sphere, Fig. 5.3. The time-averaged radiation pressure on the sphere has been found to be [87]

$$P = \frac{F}{\pi r_s^2} \quad (5.1.34)$$

$$= \frac{1}{c_m} \frac{1}{\pi r_s^2} (C_{ext} - \overline{\cos \theta} C_{sca}). \quad (5.1.35)$$

The quantity $\overline{\cos \theta}$ is called the *asymmetry factor* and measures the mean deviation of the scattered radiation from the direction of propagation of the incident wave. Such deviations imply changes in the EM momentum and therefore carry information about the force on the sphere by the light. Debye [88] has found a convenient expression for the asymmetry factor, yielding

$$P = \frac{1}{c_m} \frac{2}{x^2} \sum_{l=1}^{\infty} (2l+1) \Re\{a_l + b_l\}$$

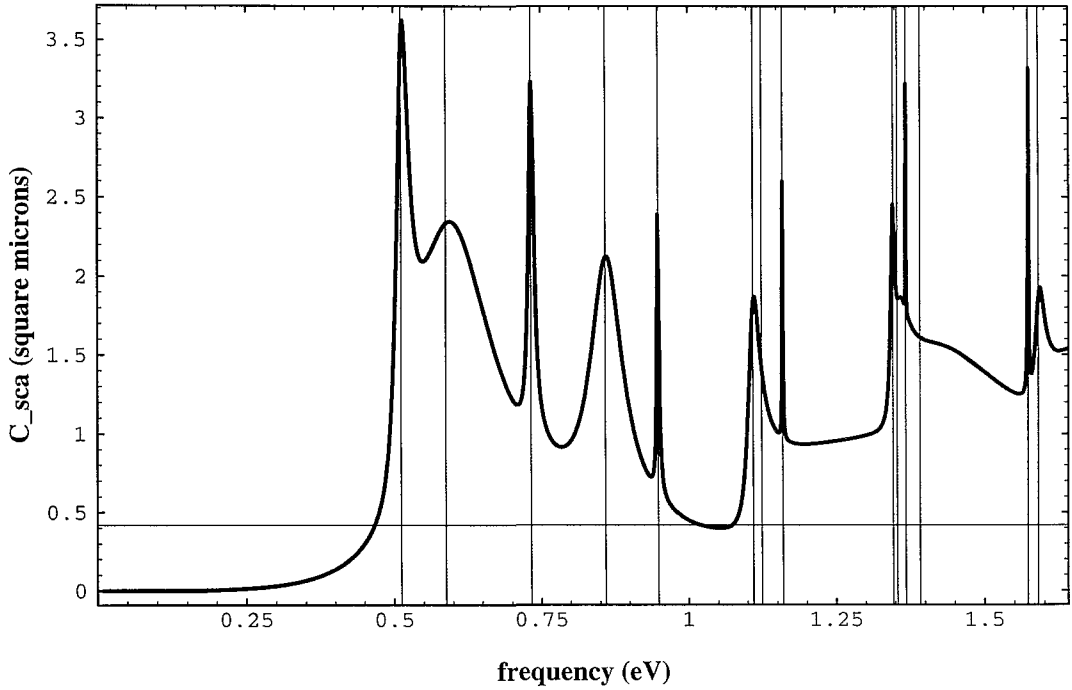


Figure 5.2: The scattering cross section C_{sca} for a GaP sphere of Table 5.1. The vertical lines show the position of the Mie resonances. The horizontal line shows the physical cross section $\pi r_s^2 = 0.418 \mu\text{m}^2$. For simplicity, no absorption or frequency-dependence in the dielectric properties of the sphere has been considered.

$$\begin{aligned}
 &+ \frac{1}{c_m} \frac{4}{x^2} \sum_{l=1}^{\infty} \left[\frac{l(l+2)}{l+1} \Re\{a_l^* a_{l+1} + b_l^* b_{l+1}\} \right. \\
 &\quad \left. + \frac{2l+1}{l(l+1)} \Re\{a_l^* b_l\} \right] \quad (5.1.36)
 \end{aligned}$$

The size of the refractive-index contrast m is crucial for the sharpness of the resonances. We plot in Fig. 5.4 C_{sca} for polystyrene spheres of $r_s = 715 \text{ nm}$ in water, where $m = 1.2$ [5]. There exist many resonances in the frequency range shown (see Table 5.2), however their large amount of damping does not allow any sharp effects to be seen. Low m also means that the sums over l in Eqs. (5.1.31, 5.1.36) converge slowly: 50 terms are taken for convergence within the frequency range shown in Fig. 5.4, whereas only 9 terms sufficed for Fig. 5.2.

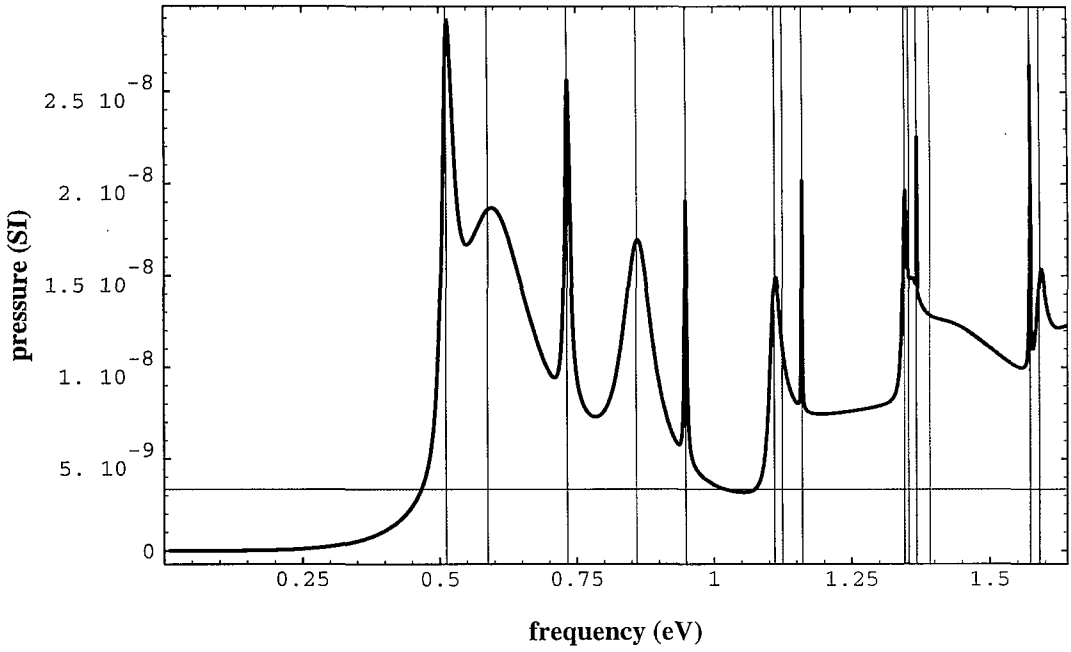


Figure 5.3: Radiation pressure on the same GaP sphere in air as of Fig. 5.2. The horizontal line shows the incident radiation pressure P_{inc} for unit intensity $I_0 = 1 \text{ W/m}^2$ (to adjust to another I_0 just multiply the pressure with the desired intensity). As expected, the pressure on the sphere approaches P_{inc} only when the scattering cross section becomes equal to the physical cross section πr_s^2 .

For dielectric spheres we find that the electric modes a_l^i are broader than the magnetic modes b_l^i of the same order. For metallic spheres, only the electric modes survive, being better known to some as ‘surface plasmons’.

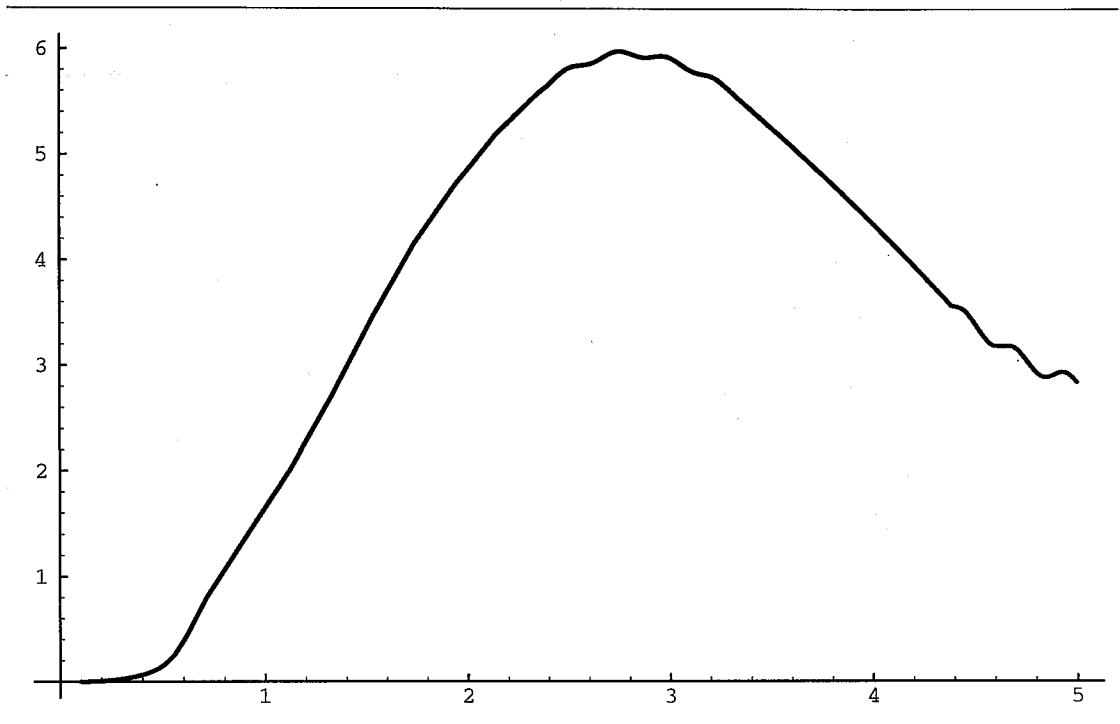


Figure 5.4: The scattering cross section C_{sca} (in units of μm^2) for polystyrene spheres in water ($r_s = 715$ nm, $m = 1.2$). The horizontal frequency axis is in eV. For simplicity, no absorption or frequency-dependence in the dielectric properties of either the sphere or water has been considered.

Type of mode n	$\Re\{\omega_n\}$ (eV)	$\Im\{\omega_n\}$ (eV)	Type of mode n	$\Re\{\omega_n\}$ (eV)	$\Im\{\omega_n\}$ (eV)
b_1	2.14301	0.197691	a_1	2.08970	0.694836
b_2	1.71397	0.200167	a_2	1.67006	0.524447
b_3	2.00891	0.201326	a_3	1.97608	0.482999
b_4	2.29617	0.202112	a_4	2.27014	0.455725
b_5	1.68293	0.198277	a_5	1.66983	0.316663
b_6	1.93504	0.192978	a_6	1.92857	0.299818
b_7	2.18592	0.187159	a_7	2.18501	0.284472
b_8	2.43589	0.181012	a_8	2.43979	0.270258

Table 5.2: To appreciate the large density of Mie resonances for the system of Fig. 5.4 (polystyrene sphere in water), we show here a non-complete list of modes located in the vicinity of a test frequency 2 eV. Many modes are actually present, here we only show the first 8 electric and magnetic multipole modes found in the frequency range 2 ± 0.4 eV (hence we do not have knowledge of their sequential order and do not provide the superscript i for each mode). The high mode density combined with the large amount of damping $\Im\{\omega_n\}$ accounts for the very broad spectrum for C_{sca} of Fig. 5.4.

5.2 Mie resonances in dielectric crystals

For most of the force plots shown in the remainder of this chapter the force calculation was done with the method of momentum-balance and not with the stress tensor calculation. The two methods give the same force away from resonances. However, for filling fractions greater than 10 % they only agree qualitatively at the occurrence of resonances, the momentum-balance calculation being correct quantitatively in these instances. The reason that the stress tensor calculation fails numerically is that it involves differences of products of *total* fields. At resonance, the evanescent components of the total fields are strongly enhanced inside the spheres, and decay away from them. Owing to the current computing limitations in the discretisation for 3D systems, there are not enough mesh points to describe the spatial decay of the resonant fields to a degree of numerical accuracy that would guarantee full correctness for the stress tensor calculation when the difference of two large quantities is taken. However for small f or when allowing at least 4 mesh layers away from the spheres to calculate the stress tensor, quantitative agreement is obtained. We show such results for $f = 0.06$.

5.2.1 Signature

Apart from effective-medium propagation, there are two distinct mechanisms providing the allowed modes for light in a dielectric crystal: scattering by the lattice structure (leading to band gaps *etc.*) provides what we shall call *lattice modes* (or *bands*); and scattering from single spheres provides the Mie modes. When many dielectric spheres are put together in a lattice to form a photonic crystal, the Mie energy levels interact with the lattice bands and both are affected: the Mie levels obtain a finite but generally small dispersion, becoming the *heavy-photon bands*, and the lattice bands are distorted, pushed into new frequency ranges or often cut in two by an intervening heavy-photon band.

Thus one way of detecting a heavy-photon band is simply to examine the photonic band structure (Fig. 4.3) and look for bands of low dispersion. This approach will track most of the low-lying (in frequency) bands having their origins in Mie scattering. However for higher frequencies, where many bands lying close together interact and further distort the bands (see Fig. 4.3.d), the certainty with which Mie-originating modes are detected with this approach decreases.

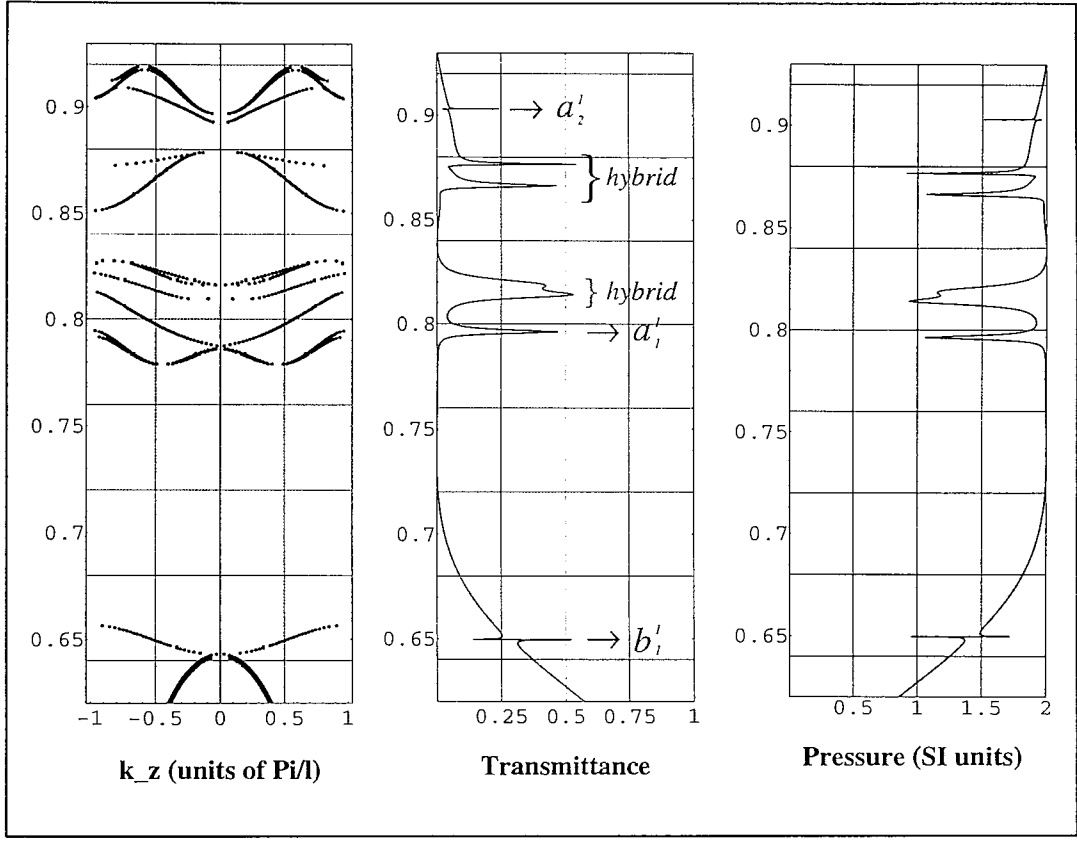


Figure 5.5: (a) Photonic band structure along the ΓX direction for a simple cubic lattice of GaP spheres ($r_s = 350$ nm, $\tilde{\epsilon} = 8.9$) in air. (b) Transmittance for a crystal monolayer and for normal incidence along $\hat{z}$. The sharp peaks correspond to the doubly-degenerate low-dispersion modes of the band structure in (a), and occur when the incident light excites one of the EM eigenmodes of isolated spheres (Mie resonances). Some peaks can be attributed to a single resonance (b'_1 : magnetic dipole mode; a'_1 : electric dipole mode; a'_2 : electric quadrupole mode) while others involve mixing from more than one Mie modes (hybrids) owing to the organisation of the spheres in a lattice. (c) Normal pressure on one unit cell for the system of (b).

Another question of experimental importance is which of the heavy-photon modes can couple to external radiation, since not all such modes are associated with sharp peaks in the transmittance spectra. To answer this question thoroughly one has to study the symmetry of each band with group theory arguments[90]. If the band structure or the symmetries are not known, one has to perform a scattering experiment and look for sharp peaks in the spectra, as

in Fig. 5.5. However, we have found that much more sensitive than the total transmittance t (or total reflectance r) is the component of the transmission (reflection) coefficient involving polarisation conversion, $T_{sp}(R_{sp})$ or $T_{ps}(R_{ps})$. Under the conditions for Mie scattering, the secondary radiation is chiefly composed of the resonant spherical wave that is elliptically polarised (see Sec. 5.1.2). In Fig. 5.6 we show that both the magnitude and the phase of T_{sp} undergo sharp changes when a resonance takes place. The same is true for R_{sp} , as shown in Fig. 5.7. Looking closely into the peaks we find that for each resonant peak the phase of T_{sp}, R_{sp} suffers an abrupt change by an angle between $\pi/4$ and π (Figs. 5.8, 5.9). Bands whose origins can be attributed to single Mie modes (especially the b_1^1 and a_2^1 bands) cause the sharpest peaks in all T_{sp}, R_{sp}, t, F , whereas hybrid modes are less likely to influence the t and F spectra.

5.2.2 Location in ω axis and mode identification

When many spheres approach each other to form the lattice, interactions occur that shift the resonances with respect to the frequencies for an isolated sphere (Fig. 5.10). Unless the coupling between neighbouring spheres is strong, each resonance in the crystal will retain its characteristic EM field distributions on the surface and the interior of each sphere. Thus the lowest-in-frequency heavy-photon resonance in our GaP crystal is the b_1^1 magnetic dipole mode [83] (Figs. 5.11, 5.12). The corresponding EM fields match exactly the expected patterns for Mie scattering from a single sphere. The next resonance is an electric dipole a_1^1 mode, with field distributions matching adequately the Mie patterns (Fig. 5.12).

For the next peak in transmittance (at ~ 0.81 eV in Fig. 5.5.b) the EM fields are similar but not identical to those for the magnetic quadrupole mode b_2^1 ; and likewise for the following two peaks (just before and after 0.87 eV), whereby the fields display partial similarity with those for the electric quadrupole mode a_2^1 [89]. Two are the origins of such modes: (a) Mixing from more than one Mie modes owing to the organisation of the spheres into a lattice. To quantify the contribution of each Mie mode into the heavy-photon resonance one has to decompose the scattered field into its constitutive spherical waves, as Ohtaka and Tanabe have done[77]. (b) Another source for such hybrid modes may be the coarsening of the spherical surfaces associated with the discretisation of space.

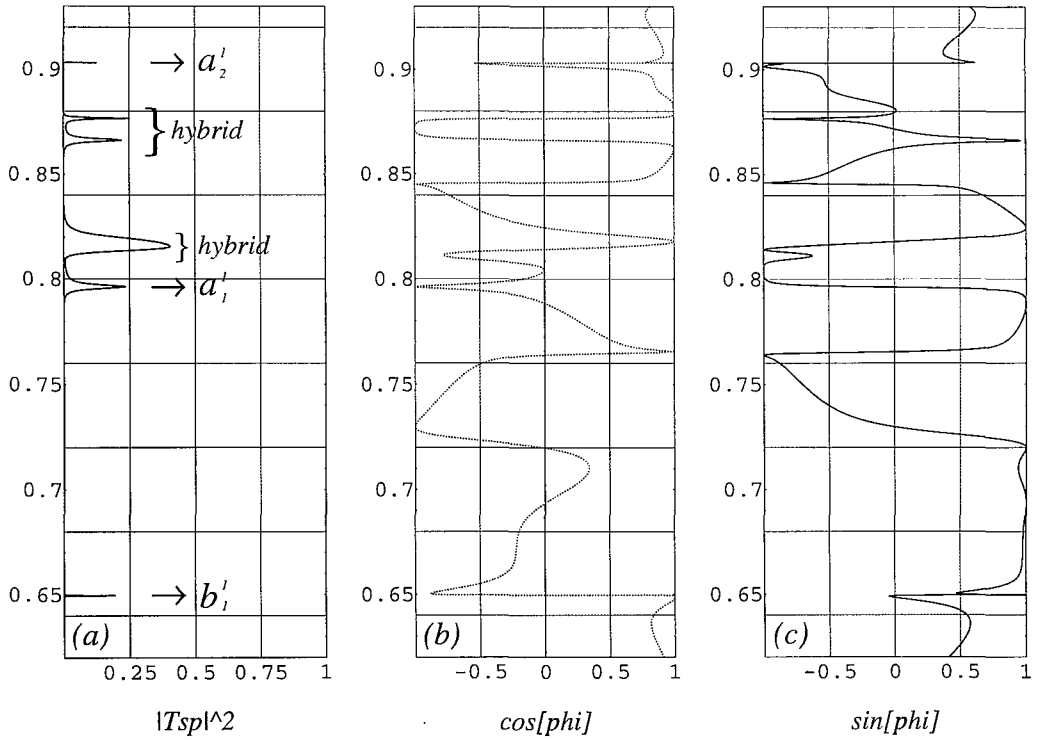


Figure 5.6: Signature of resonant scattering for the system of Fig. 5.5.b. The spherical wave excited by the resonance dominates the scattered field which therefore becomes elliptically polarised (see text), causing nonzero T_{sp} . Both the magnitude of $T_{sp} = |T_{sp}| e^{i\phi}$ and its phase ϕ suffer abrupt changes at the occurrence of the resonances.

Due to the finite number of mesh points per unit cell, the dielectric sphere is approximated by a spheroid that has a lower symmetry and a number of flat surfaces. Since any object possesses its own Mie-type modes, it follows that the spectra we obtain for the actual spheroids of our mesh (hoping they hold for true spheres) possess features that apart from being quantitatively different from their spherical counterparts, may even be non-existent for perfect spheres. In the following sections we discuss the plausibility of this argument.

However the mode that follows in frequency (at ~ 0.9 eV) is a distinct electric quadrupole, with field distributions identical to the a_2^1 Mie mode (Fig. 5.13) [89].

As the frequency increases and higher-order resonances are expected there

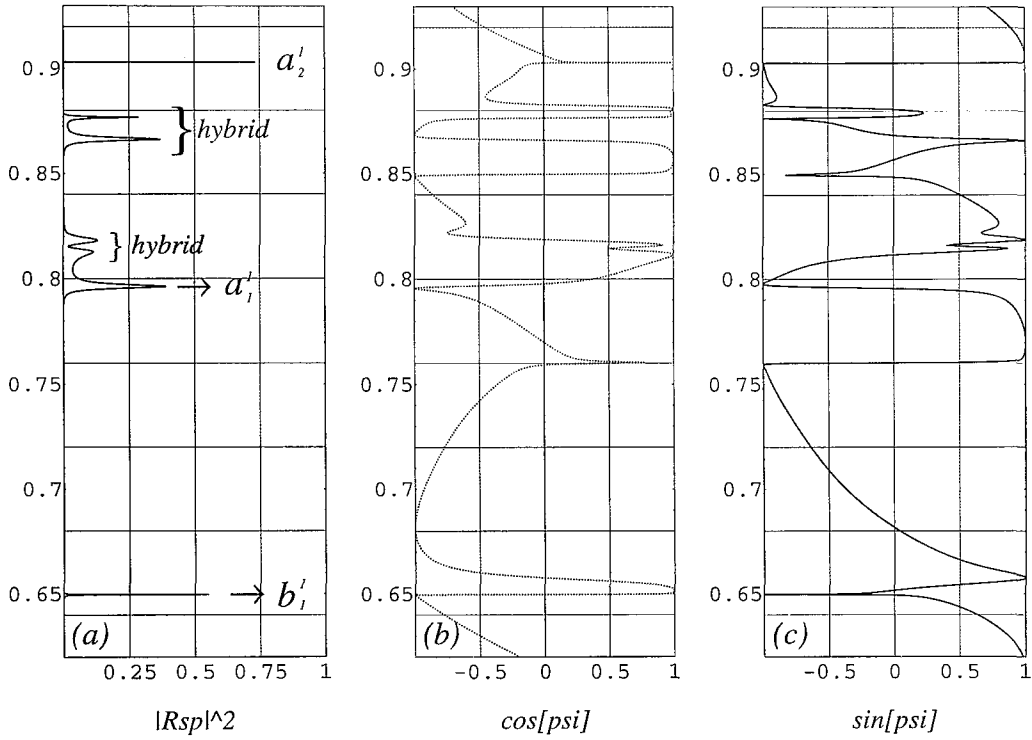


Figure 5.7: Equivalent plot to Fig. 5.6 but for the reflection coefficient $R_{sp} = |R_{sp}| e^{i\psi}$. The pure modes labelled as b_1^1, a_1^1, a_2^1 result in very sharp and narrow peaks, the hybrid modes produce broader peaks.

is a greater requirement for spatial resolution of the fields inside the sphere, calling for a larger discretisation mesh. At present we are restricted to mesh of sizes roughly up to $15 \times 15 \times 15$, and have therefore avoided looking into the higher-order resonances in detail.

Depending on ω , the incident field produces polarisation charges and/or induced currents in the spheres and orchestrates their response according to which mode is excited each time. It is interesting that spheres which are initially non-magnetic ($\tilde{\mu} = 1$) can acquire magnetic properties at the occurrence of any b_i^i resonance. An effective homogeneous medium replacing the crystal structure would have $\tilde{\mu} \neq 1$ at this instance.

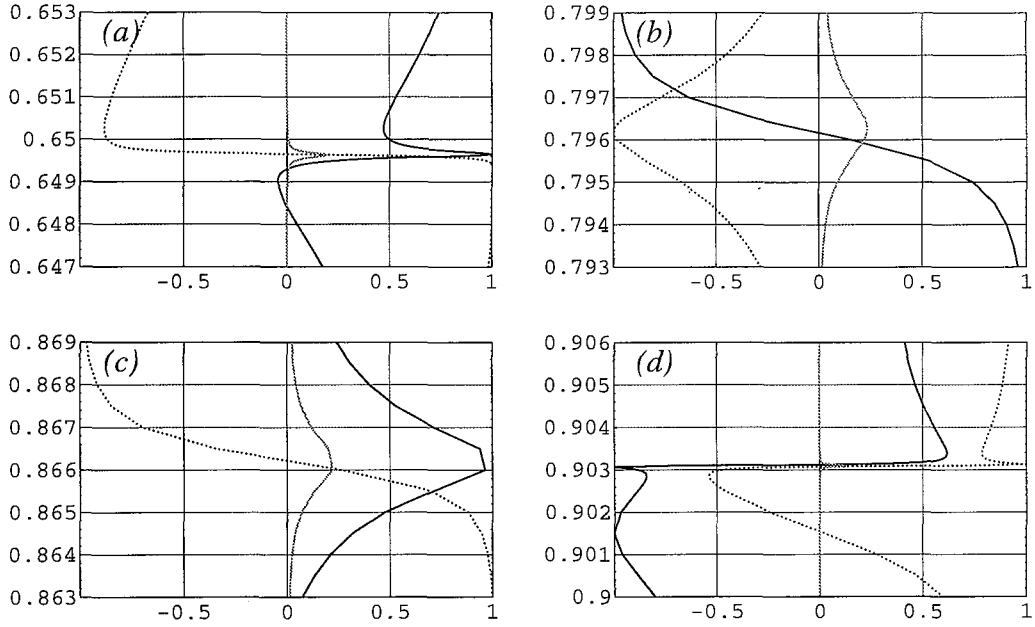


Figure 5.8: Detail from Fig. 5.6. The magnitude $|T_{sp}|^2$ is shown in grey colour, $\sin \phi$ in solid black line and $\cos \phi$ in dotted black line. (a) The b_1^1 resonance. A total change of $\delta\phi \sim 5\pi/6$ occurs at the vicinity of the peak. (b) The a_1^1 resonance. $\delta\phi \sim \pi$. (c) One of the hybrid resonances. $\delta\phi \sim \pi$. (d) The a_2^1 resonance. $\delta\phi \sim 2\pi/3$. The smallest phase change of about 50 deg occurs for the lowest hybrid resonance at ~ 0.81 eV (not shown here).

5.2.3 Influence of filling fraction f

It was mentioned in the previous section that two possible causes for the hybrid modes were the proximity of the dielectric objects in the lattice and the coarsening of the spheres into spheroids with reduced symmetry properties. As long as we are trying to model spheres, both effects will be simultaneously present in the spectra and it will be hard to distinguish the relative consequences of each one in particular. Therefore in this section we try to separate these two effects by studying cubes alone. We study three systems, each of a unit-cell length $l_0 = 900$ nm, with the edge-length of cubes increasing from (a) 360 nm ($f_a = 0.064$) to

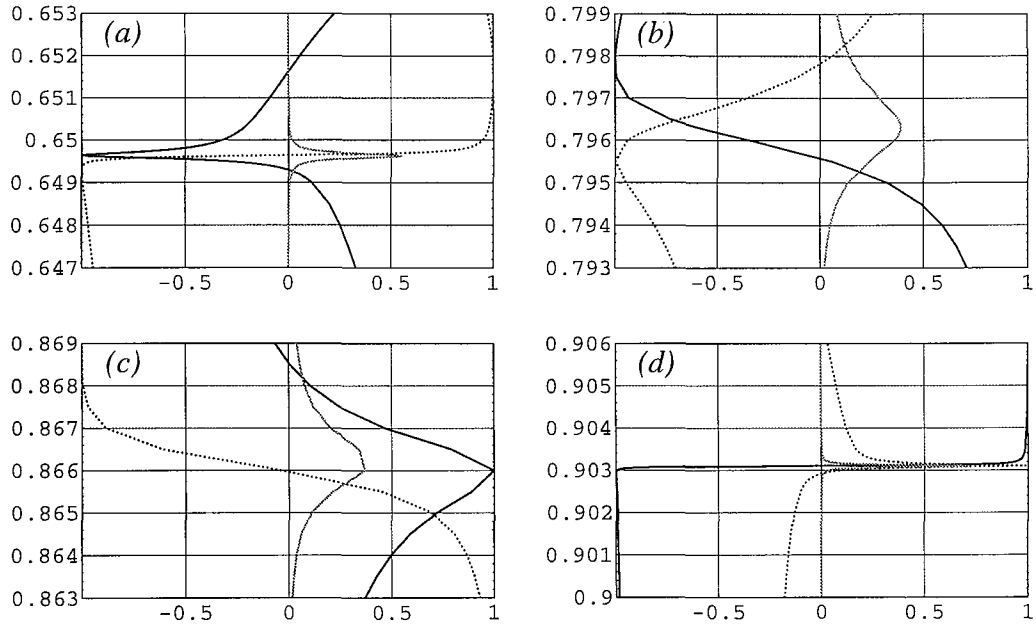


Figure 5.9: Detail from Fig. 5.7. The magnitude $|R_{sp}|^2$ is shown in grey colour, $\sin \psi$ in solid black line and $\cos \psi$ in dotted black line. (a) The b_1^1 resonance. A total change of $\delta\psi \sim \pi$ occurs at the vicinity of the peak. (b) The a_1^1 resonance. $\delta\psi \sim \pi$. (c) One of the hybrid resonances. $\delta\psi \sim \pi$. (d) The a_2^1 resonance. $\delta\psi \sim \pi$.

(b) 540 nm ($f_b = 0.216$) to (c) 720 nm ($f_c = 0.512$). We monitor their respective photonic band structures along ΓX direction (Fig. 5.14); also their transmission coefficients and force spectra (Figs. 5.15, 5.16, 5.17).

Firstly, we find that the number of bands is conserved, at least up to energies of 1.35 eV for system (a) which corresponds to 1 eV for system (b) and 0.77 eV for (c). In other words, all three systems possess 12 qualitatively similar bands above the lowest frequency for which the bands cross the $k_z = 0$ axis. This is the same number of bands shown in Fig. 5.5 for the spheroid system.

Secondly, the number of resonance peaks in the transmission coefficient $|T_{sp}|^2$ spectra is also conserved: 6 peaks are found at the frequency range cov-

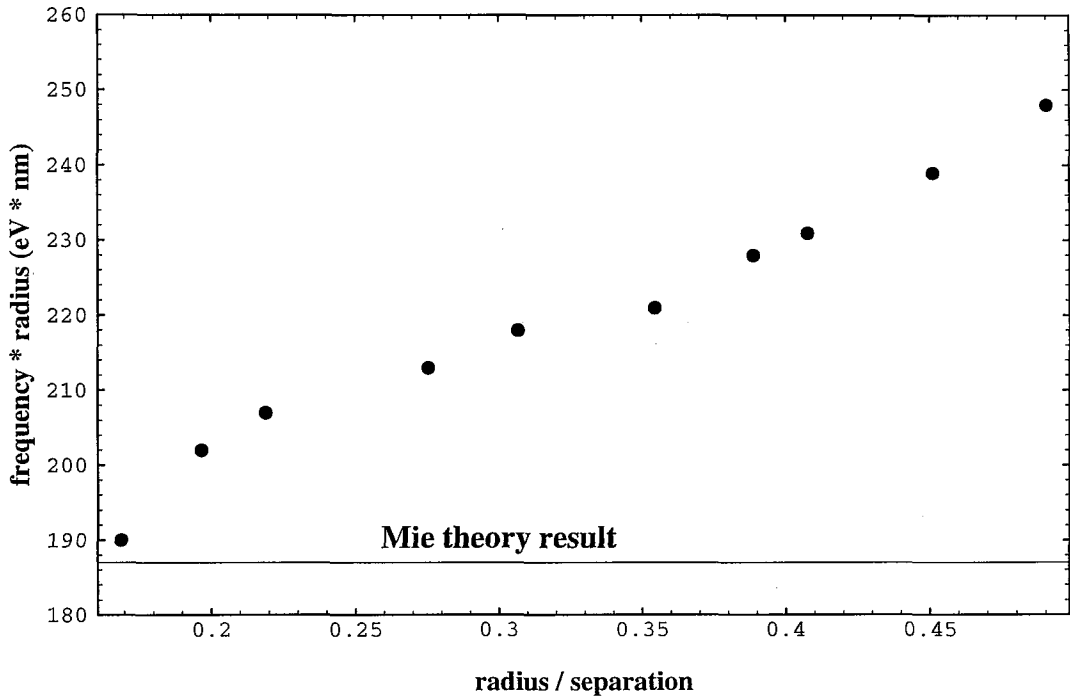


Figure 5.10: For a given Mie mode (at frequency ω) of an isolated sphere (radius r_s), the product $\omega \cdot r_s$ is independent of the sphere size. When the spheres are put together in a lattice, interactions shift the resonances. For radius small compared to the separation (l_0), the one-sphere result is recovered. Results here are for the b_1^1 mode.

ered by the 12 bands in all cubic systems; the same number of peaks as for the spheroids. The lowest two of those have been identified respectively as the exact equivalents of the magnetic (b_1^1) and electric (a_1^1) dipole bands found in spheres, as professed by their field distributions.

So it appears that the number of the hybrid resonances we discovered in Fig. 5.5 are less likely to be due to the proximity of the dielectric objects in the photonic crystal. Rather they seem to be connected to the cubic properties (*i.e.* cubic symmetry, flat faces, acute angles, etc.) of the spheroids approximating true spheres. Let us note here that it was not possible to extend our comparison of the three cubic systems to more bands because system (a) displays off-diagonal

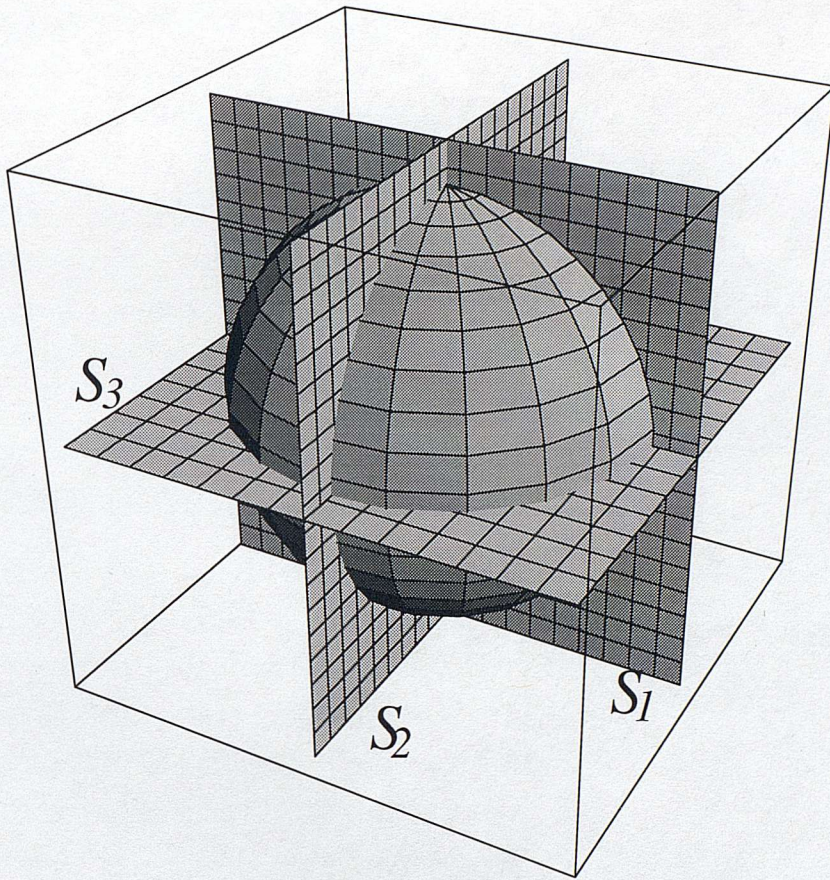


Figure 5.11: The spherical sections S_1, S_2, S_3 on which the field distributions are plotted in the following figures.

scattering beyond the 12th band and this results in additional features in the spectra that are not related to Mie resonances: for example the sharp force dip at 1.355 eV, for which, incidentally, the EM energy enhancement is nowhere near the heavy-photon resonance values: the average energy inside the cube is a mere factor of 20 larger than the incident energy (compared to at least a few hundred for the resonances).

But there also exist some differences between the three cubic systems: out of the 12 bands, some (not all) change shape and/or relative position, whereas the sequence in which they appear is also changed both from (a) to (b) as well as from

(b) to (c). The heavy photons at lowest ω are least affected. They are gradually prolonged in going from (a) to (c); in contrast, the heavy photons immediately higher up are gradually flattened. Other bands do not show a monotonic variation of their extent as f increases. Thus we notice that while some band gaps open up from f_a to f_b , all of them except one have closed by the time f reaches f_c .

Other effects found are the general tendency for packing the bands into lower energies as f increases which results from the increase of average ϵ (ϵ_{av}) of the unit cell (since ω scales roughly as $1/\sqrt{\epsilon_{av}}$). Also the sharpness of the resonance peaks, both for the transmittance and the force spectra, increases with f *i.e.* with the proximity of the artificial atoms.

5.2.4 Influence of spheroidal properties other than cubic symmetry (size, shape, mesh)

In the previous section it was shown that the spectra (band structure and force) of dielectric crystals of spheroidal and cubic objects display several similar features. Out of the several cubic properties, we try to isolate the ones responsible for the similarities: is it the cubic symmetry, or the existence of flat surfaces and acute angles? Both the former and the latter are shared between spheroids and cubes. We try to answer this question in this section.

First we compare between two different classes of spheroids, one with sharp dielectric boundaries (which we refer to as ‘binary- ϵ system’) and another with smooth borders (‘average- ϵ system’). For the binary- ϵ system the ϵ at each point in the unit cell is defined as either ϵ_s or ϵ_m , depending on whether the point lies inside or outside the sphere (hence our choice of name). For the average- ϵ system, ϵ at each mesh point is defined as the weighted average of ϵ_s and ϵ_m , such that deep inside the sphere $\epsilon = \epsilon_s$, far away from it $\epsilon = \epsilon_m$, and in the vicinity of the spherical boundary ϵ has an intermediate value. This method of defining ϵ has been explained in Ref. [64].

The two types of objects that result are shown in Fig. 5.18. Both share cubic symmetry. Their respective band structures (Fig. 5.19) do not display any telling differences: both have an equal number of bands and the gaps appear at similar frequencies for similar packing fractions. Their force (Fig. 5.20) and $|T_{sp}|^2$ spectra possess an equal number of peaks also. The same number of bands and resonant features was found for cubes earlier. The major qualitative difference

regards the two hybrid electric quadrupole modes (a_2^1): the average- ϵ system displays two *dips*, whereas the binary- ϵ system displays two *peaks*. Also upon analysing the EM fields inside the spheroids we find that for the average- ϵ system the 2nd resonance is an electric dipole mode (a_1^1) followed by a (hybrid) magnetic quadrupole mode (b_2^1), whereas for binary- ϵ it is the other way round.

Of the two systems, the behaviour of average- ϵ is less parameter-dependent. Convergence occurs quickly, *i.e.* for moderate mesh sizes: a $9 \times 9 \times 9$ mesh suffices to produce all the main qualitative features in the force spectra (Fig. 5.22.a). The same is true for a $10 \times 10 \times 10$ mesh (Fig. 5.20.b). At a quantitative level convergence is achieved for all resonances shown by the time the mesh becomes $13 \times 13 \times 13$ (compare Figs. 5.22.b,c) which is quite realistic regarding the time of calculation needed.

The averaging procedure in defining ϵ removes or reduces the acute features in the spheroid shape caused by the discretisation (Fig. 5.21). In this way small spheres may be treated also, provided one of the larger mesh sizes is used (such as $15 \times 15 \times 15$ or higher). We are thus confident that at least for the first few heavy-photon modes (energies up to ~ 1 eV for GaP spheres of $f \sim 0.25$ in a unit cell of length 900 nm), the average- ϵ definition allows reliable calculations of the force spectra.

On the other hand, for the binary- ϵ system the sequence and the strength of the resonances depends sometimes sensitively on the parameters: Thus we find that for small values of the filling fraction f (*i.e.* small spheroid sizes) and moderate mesh size ($\sim 11 \times 11 \times 11$) the a_1^1 resonance precedes b_2^1 (Fig. 5.23), contrary to what was found for larger spheroid sizes.

To conclude this section, we find that the *number* of heavy photons in the band structure and of resonant features in the force and transmission spectra are independent of the shape, size and proximity of the artificial atoms that form the photonic crystal. They must result from the *symmetry* of the system (*i.e.* the symmetries of the lattice and/or of the artificial atoms). However the *strength* (and sometimes the order) of the resonances in the force spectra depends on the specific parameters such as the filling fraction f or the shape of the artificial atoms. As a general trend proximity between spheroids increases interactions and the resonant forces are enhanced. The average- ϵ definition must be used in order to minimise unwanted effects due to the coarsening of spheres.

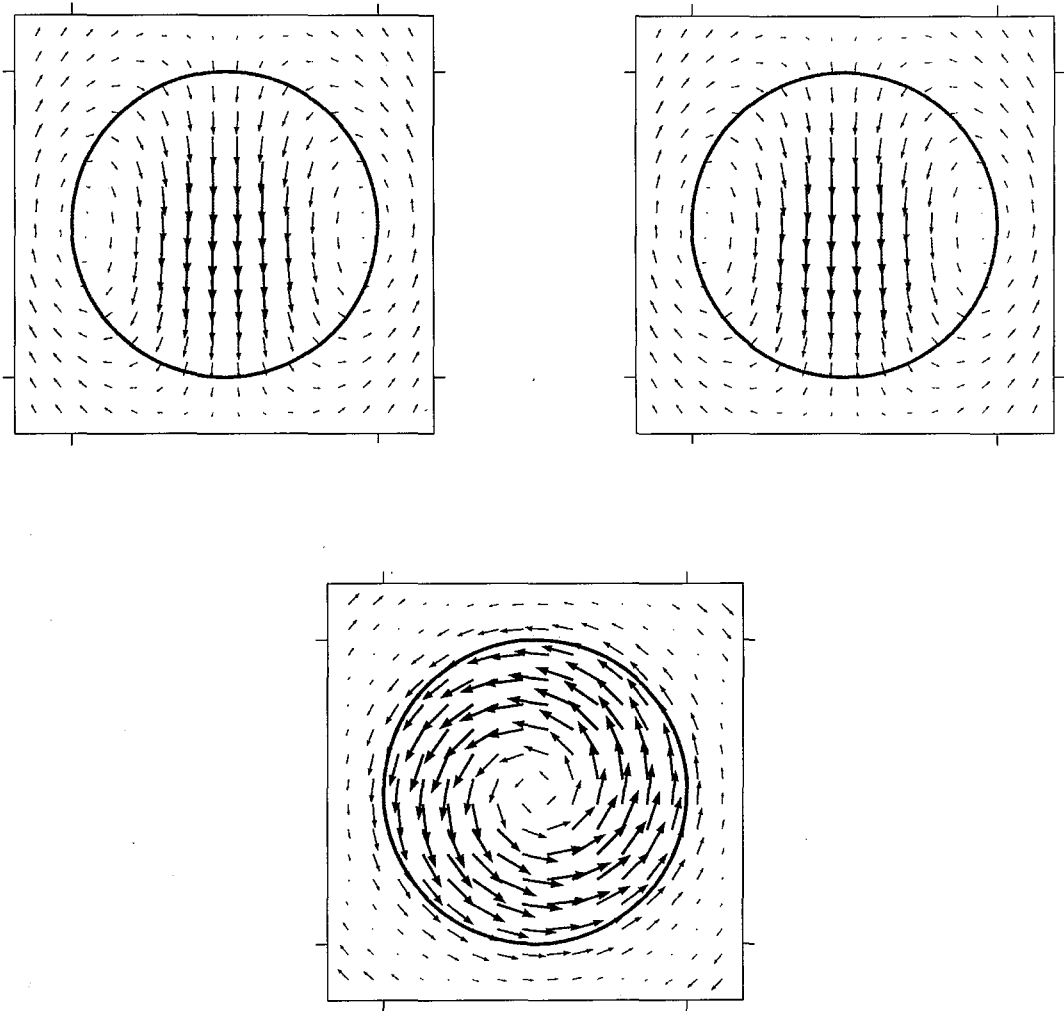


Figure 5.12: (a-c): Dipole mode. For the b_1^1 resonance (magnetic mode) the $\mathbf{H}$ field is plotted on the spherical sections S_1, S_2 , while the $\mathbf{E}$ field is shown on S_3 (see Fig. 5.11). For the a_1^1 electric mode the role of the $\mathbf{H}$ and $\mathbf{E}$ fields is interchanged.

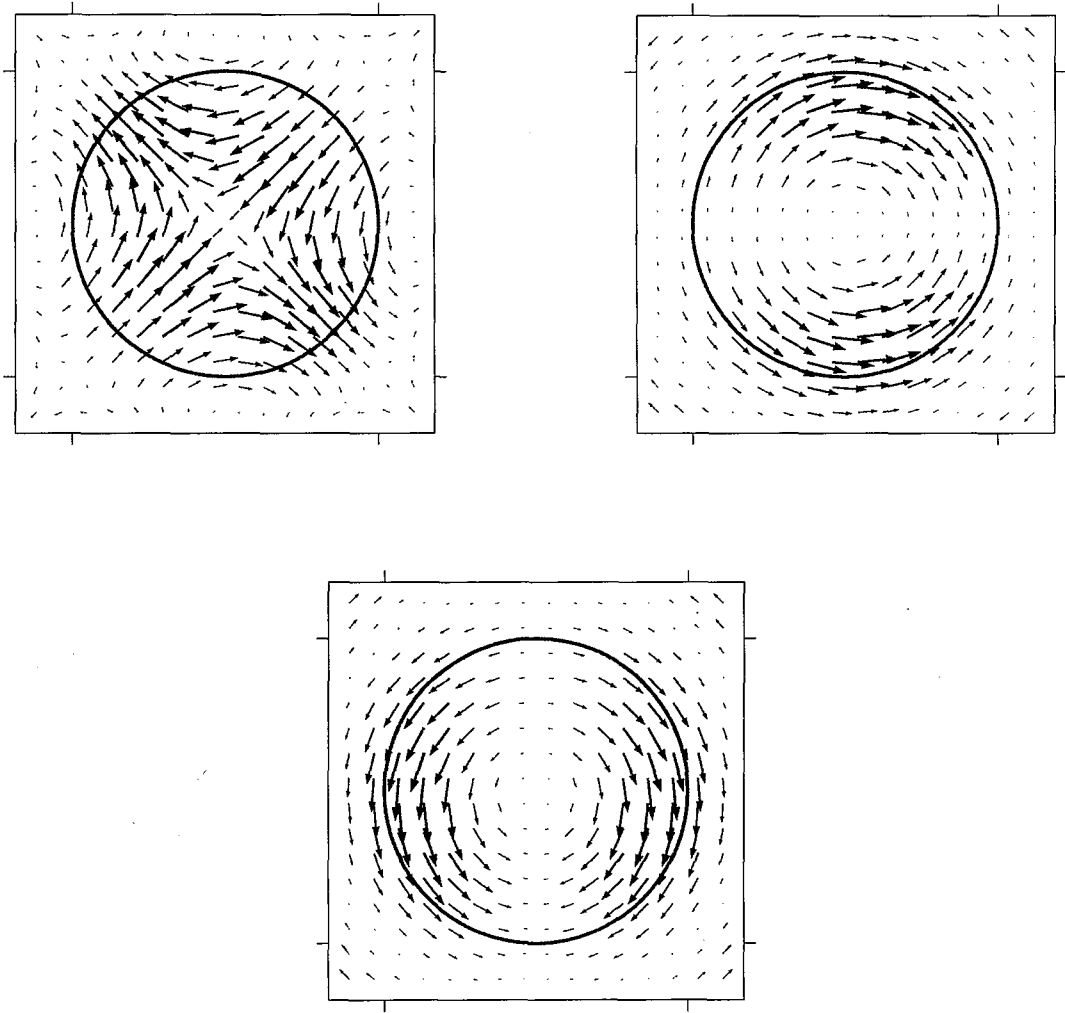


Figure 5.13: (a-c):Quadrupole mode. For the b_2^1 resonance (magnetic mode) the $\mathbf{H}$ field is plotted on the spherical section S_1 , while the $\mathbf{E}$ field is shown on S_2, S_3 . For the a_2^1 electric mode the role of the $\mathbf{H}$ and $\mathbf{E}$ fields is interchanged.

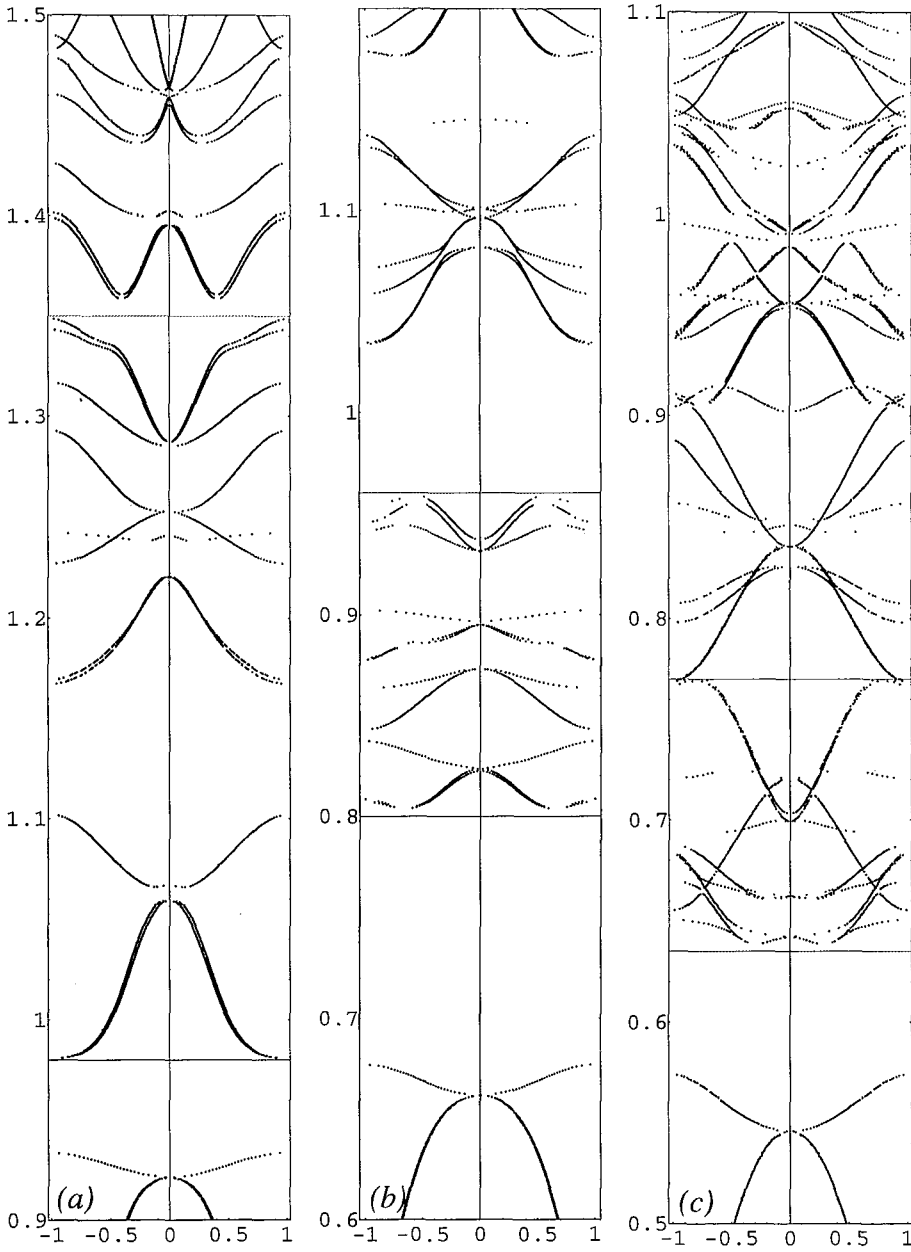


Figure 5.14: The band structure along ΓX for a SC lattice of GaP *cubes* and edge-length (a) 360 nm (filling fraction $f_a = 0.064$), (b) 540 nm ($f_b = 0.216$) and (c) 720 nm ($f_c = 0.512$). The horizontal gridlines for each plot contain the 11 bands following the b_1^1 -like resonance.

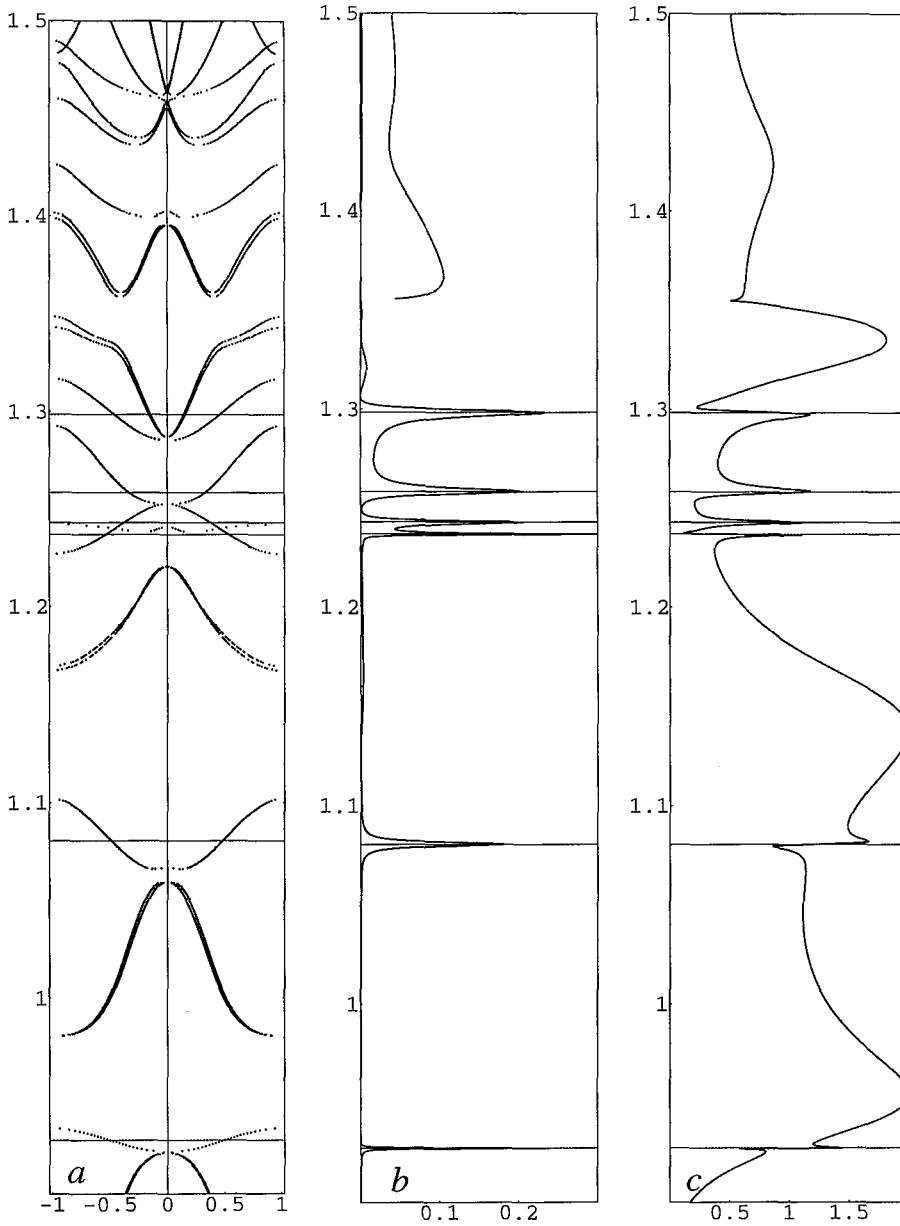


Figure 5.15: For the crystal monolayer of cubes with filling fraction $f_a = 0.064$ (Fig. 5.14.a). Comparison between (a) photonic band structure along ΓX , (b) $|T_{sp}|^2$, (c) pressure P_z on one unit cell. For (b) and (c) light is normally incident along $\hat{z}$. The horizontal gridlines denote the positions of the peaks in the $|T_{sp}|^2$ spectrum. All except one of the sharp features in $|T_{sp}|^2$ and P_z are due to heavy-photon resonances. The exception is the dip in pressure at 1.355 eV, caused by off-diagonal scattering. In fact there exist four off-diagonally scattered beams beyond this frequency, but only one dominates strongly the spectrum of $|T_{sp}|^2$.

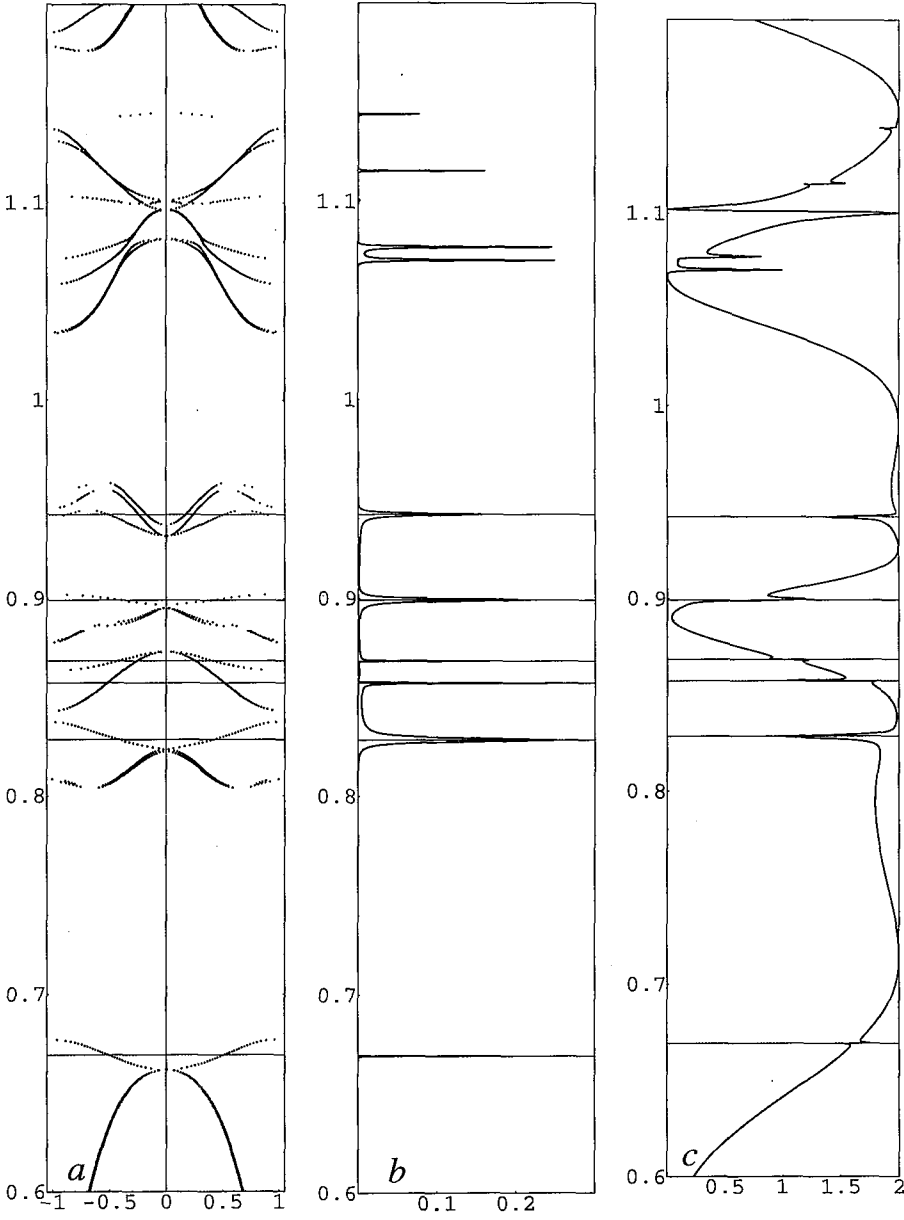


Figure 5.16: Same as Fig. 5.15 but for cubes of edge-length 540 nm ($f_b = 0.216$). All sharp effects appear both in the $|T_{sp}|^2$ and pressure spectra.

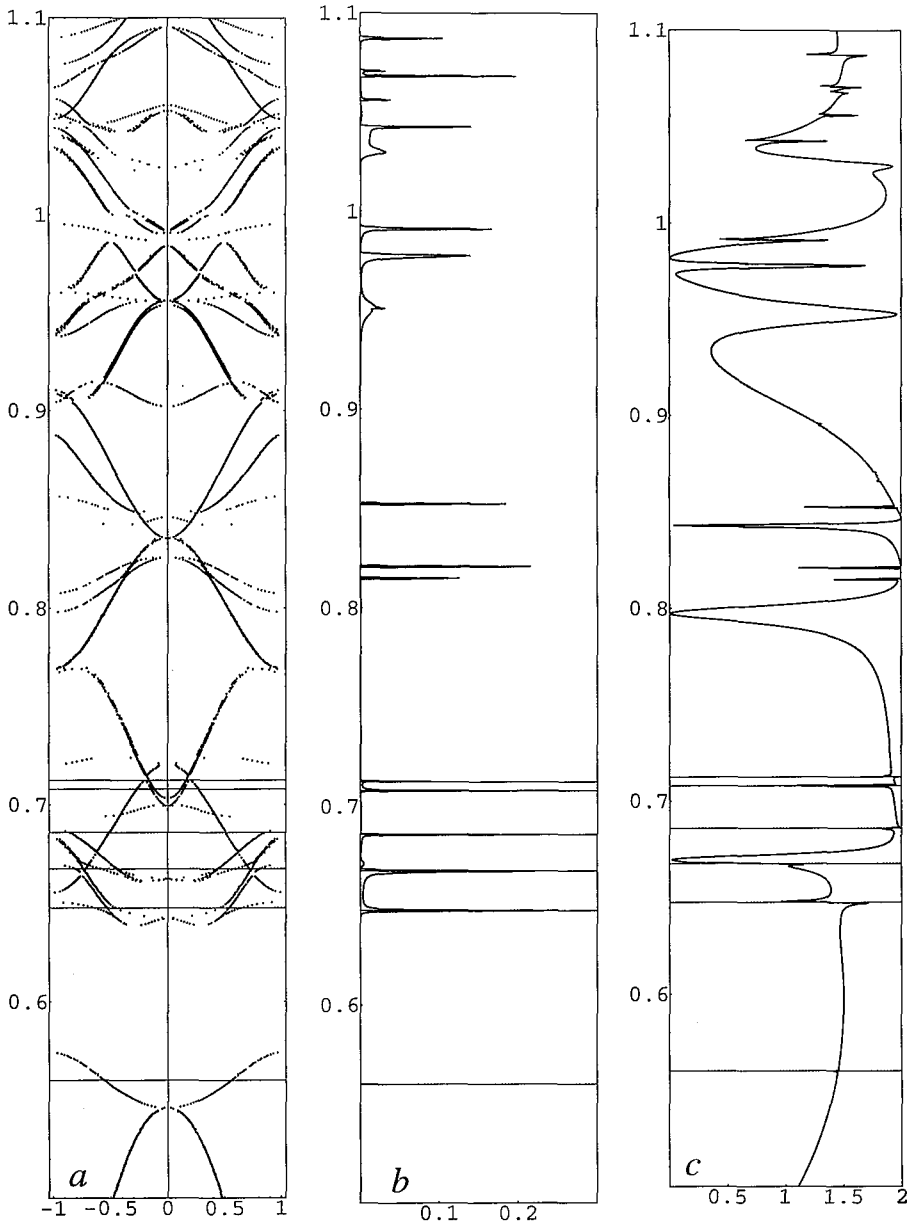


Figure 5.17: Same as Fig. 5.15 but for cubes of edge-length 720 nm ($f_c = 0.512$). The resolution of the peaks is such that up to 0.8 eV, all sharp effects appear in both the $|T_{sp}|^2$ and pressure spectra. Beyond 0.8 eV some peaks are not resolved in the $|T_{sp}|^2$ spectrum but show up in P_z .

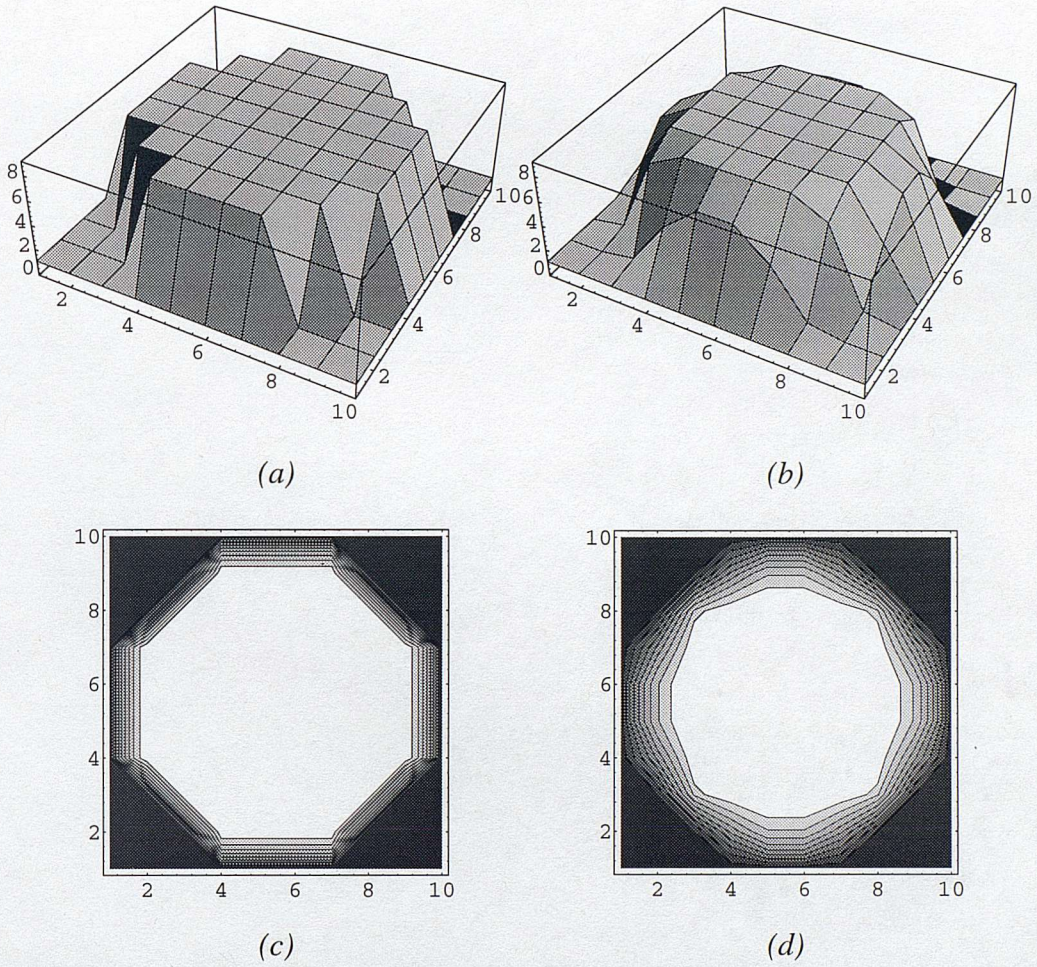


Figure 5.18: Binary- ϵ vs. average- ϵ . Profile of the dielectric function on a cross section S_3 cutting through the middle of the unit cell, for (a) binary- ϵ and (b) average- ϵ . The horizontal axes show the location in the mesh and the vertical axis the value of $\tilde{\epsilon}$, ranging from $\tilde{\epsilon}_m = 1$ to $\tilde{\epsilon}_s = 8.9$. In (c),(d) the contour plot of parts (a),(b) respectively is shown; the shading is such that black corresponds to locations outside the spheroid, white to locations inside. For both systems the discretisation mesh is $10 \times 10 \times 10$. $r_s = 365$ nm for (a), $r_s = 350$ nm for (b).

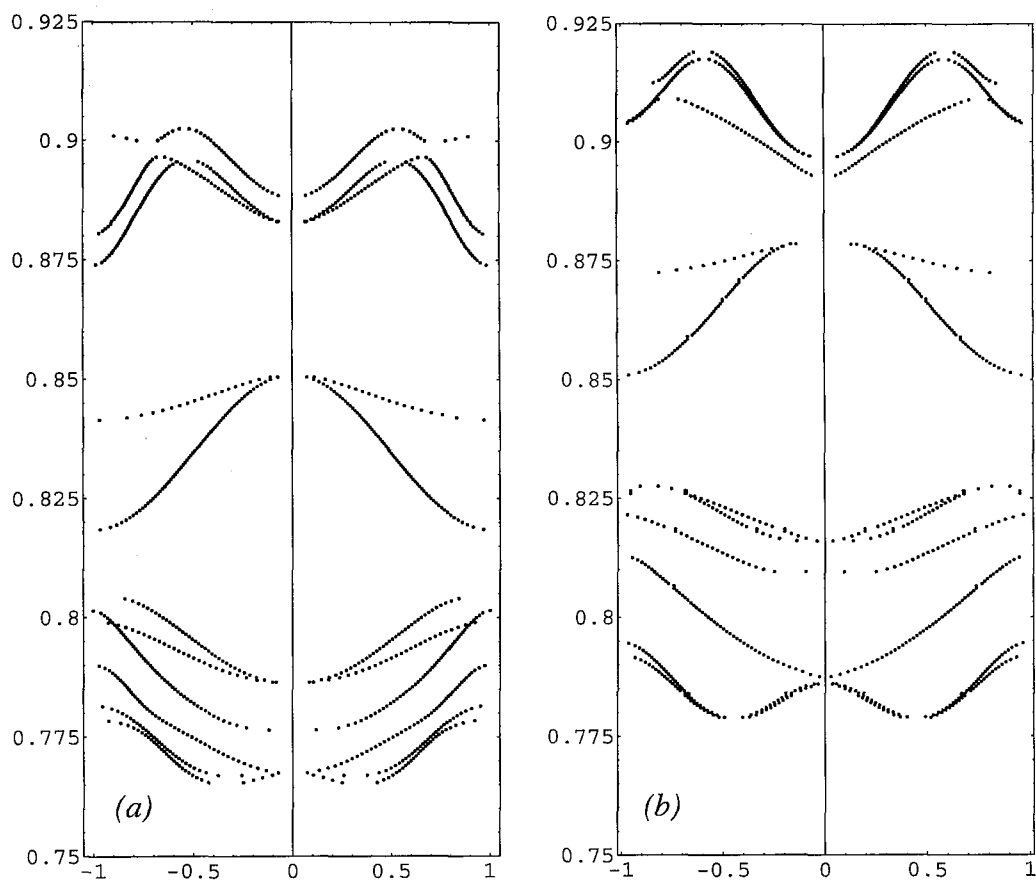


Figure 5.19: Binary- ϵ vs. average- ϵ . The band structure along ΓX for (a) the binary- ϵ and (b) the average- ϵ systems of Fig. 5.18. The 11 bands following the b_1^1 resonance are shown. No major qualitative differences are visible.

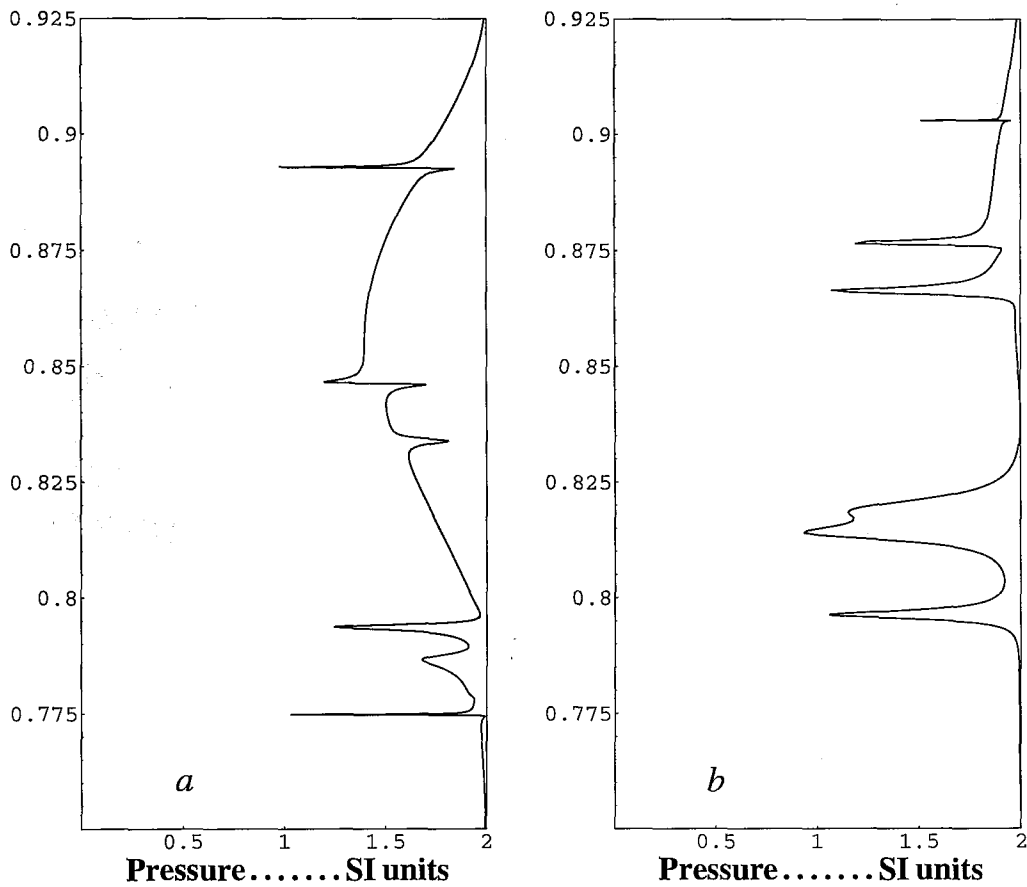


Figure 5.20: Binary- ϵ vs. average- ϵ . Normal pressure on a unit cell for (a) the binary- ϵ and (b) the average- ϵ systems of Fig. 5.18. A qualitative difference is that two resonance peaks in (a) (between 0.825 eV and 0.850 eV) appear as dips in (b). The remaining resonances present only quantitative differences between the two systems.

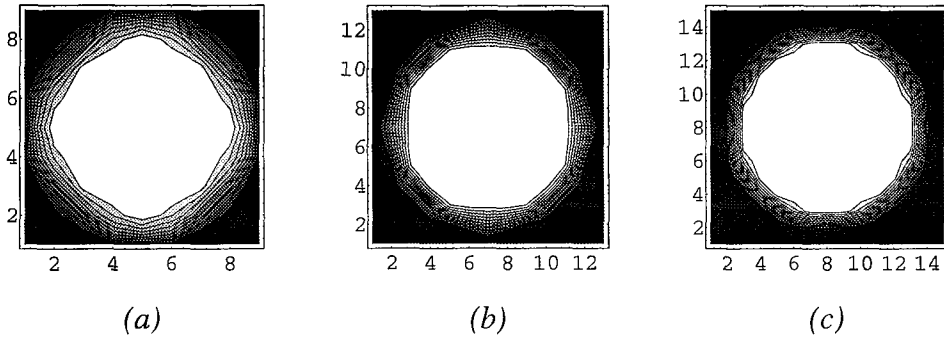


Figure 5.21: Average- ϵ for different mesh sizes. Contour plot of the profile of the dielectric function on a cross section S_3 cutting through the middle of the unit cell, for (a) $9 \times 9 \times 9$ ($r_s = 349$ nm, $f = 0.2450$), (b) $13 \times 13 \times 13$ ($r_s = 337$ nm, $f = 0.2208$), (c) $15 \times 15 \times 15$ ($r_s = 337$ nm, $f = 0.2208$).

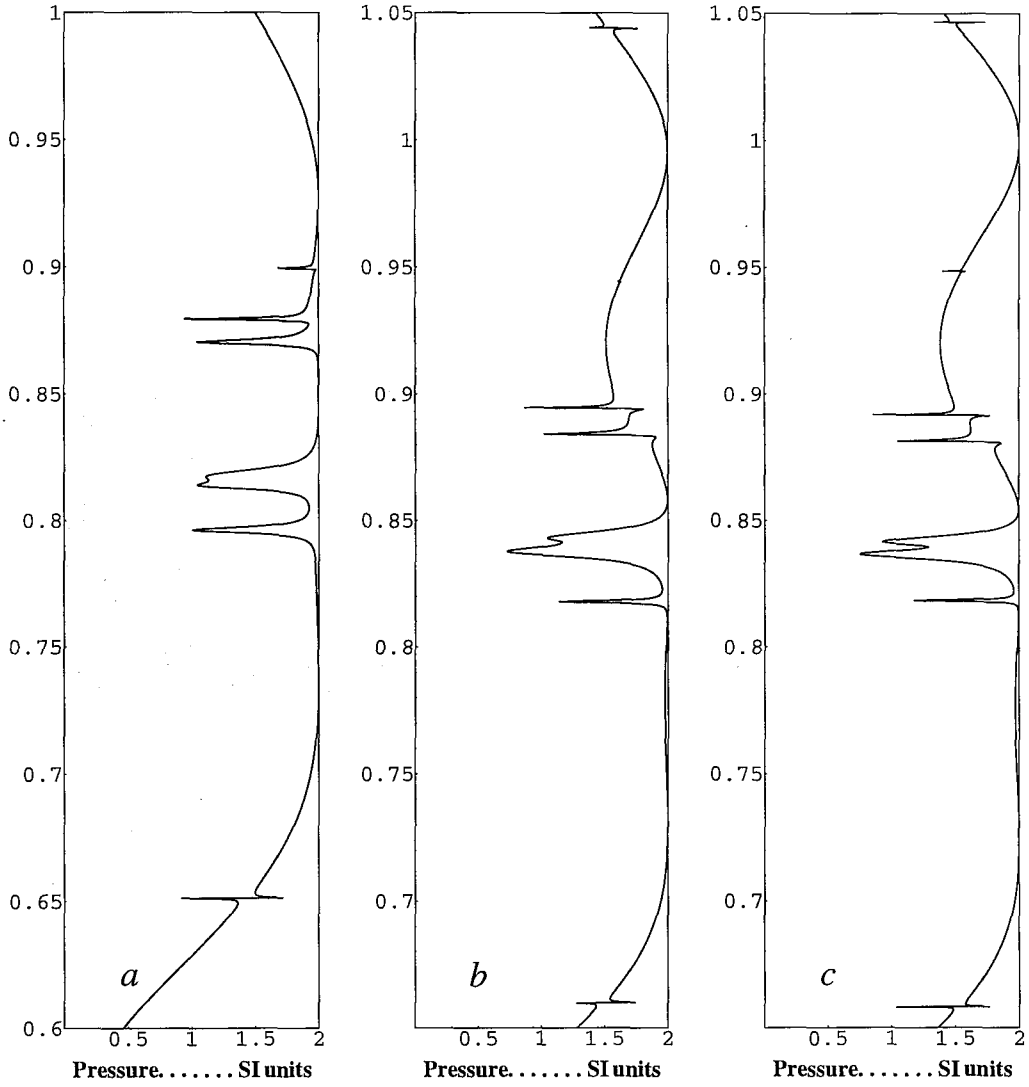


Figure 5.22: Forces for different mesh sizes and average- ϵ . Normal pressure on a unit cell for (a) $9 \times 9 \times 9$, (b) $13 \times 13 \times 13$, (c) $15 \times 15 \times 15$ mesh of Fig. 5.21. All resonances display the same qualitative behaviour.

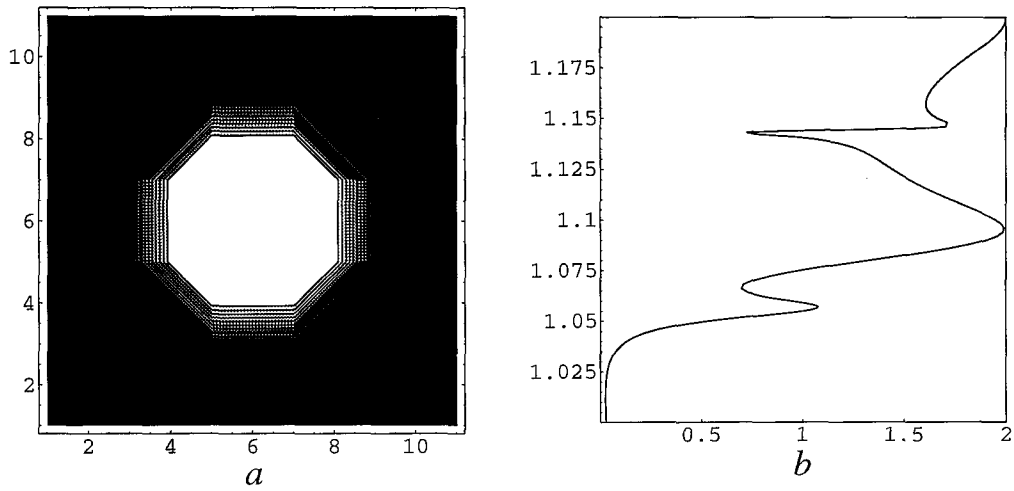


Figure 5.23: Effect of size for binary- ϵ . For a small spheroid of the *same shape* as that of Fig. 5.18.c, in a mesh $11 \times 11 \times 11$ the second lowest-in- ω resonance is an electric dipole (a_1^1) mode. The spheroid radius is $r_s = 195$ nm ($f = 0.0428$).

5.2.5 EM energy enhancement

What happens at a resonance? Each sphere acts as a cavity that traps the EM waves. The energy density internal to the spheres is 2-3 orders of magnitude larger than for the incident light beam[83, 89, 91] (Figs. 5.24, 5.25). As Van Albada *et al* have shown[92, 93], the transport of EM energy can be slowed down considerably at the conditions for Mie scattering. They derive a rigorous microscopic formula for the velocity of energy transport v_E , and subsequently propose a heuristic but physically plausible model for v_E , which they show to agree with the exact model and experimental results within reasonable accuracy, and which reads

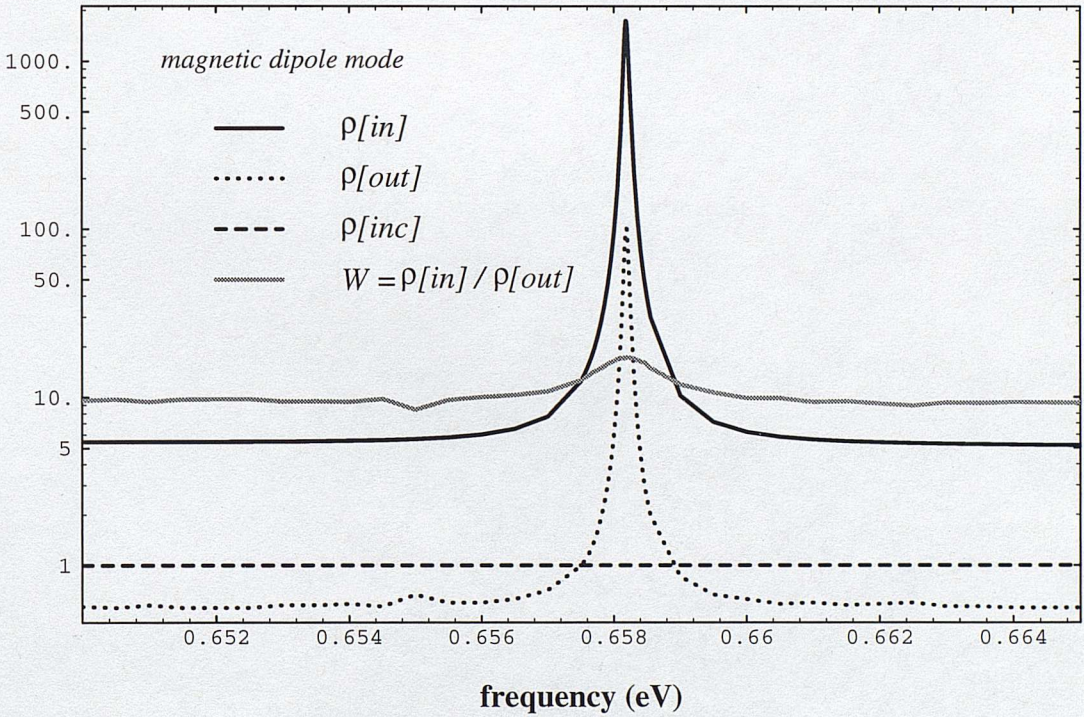


Figure 5.24: Resonant enhancement of the EM energy density $\rho = \frac{1}{2} (\mathbf{E} \cdot \mathbf{D} + \mathbf{H} \cdot \mathbf{B})$ versus ω for a monolayer of GaP spheres ($r_s = 337$ nm). The average ρ internal to the sphere for the b_1^1 mode is shown in solid black line. The average ρ in the region of the unit cell external to the sphere is shown in dotted line. The ratio of the two, defined in the text as W appears in grey. All densities scale with the incident density which is set to unity (dashed line).

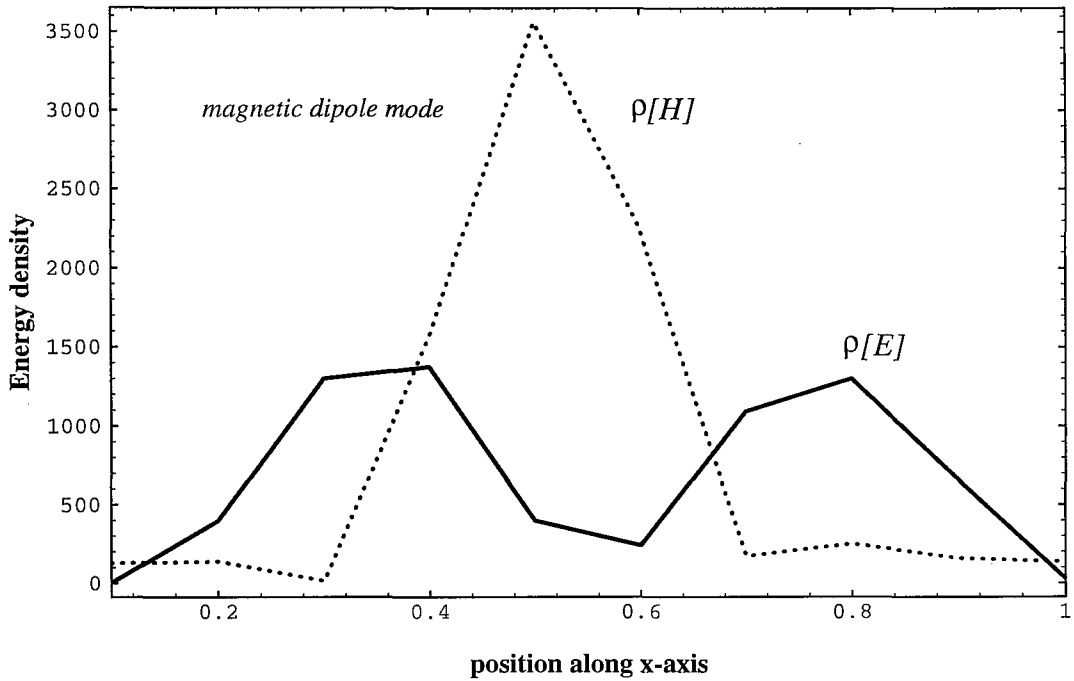


Figure 5.25: Spatial distribution of the electric ($\rho[E] = \frac{1}{2} \mathbf{E} \cdot \mathbf{D}$) and magnetic ($\rho[H] = \frac{1}{2} \mathbf{H} \cdot \mathbf{B}$) energy densities inside a unit cell at the magnetic dipole resonance (b_1^1). The $\mathbf{H}$ field forms a hyperboloid of one sheet around which the $\mathbf{E}$ field circulates in a ring (Fig. 5.12). Thus the dipole points along the $\hat{\mathbf{z}}$ axis and the electric-field ring lies chiefly on the xy plane. Here we plot the densities along an equatorial diameter on the $\hat{\mathbf{x}}$ axis (*i.e.* along the intersection between surfaces S_1, S_3 of Fig. 5.11). Ten calculation points are used, spaced out in tenths of the unit cell length, from 0 to 1. We see that relative to the (unit) EM energy density of the incident radiation, $\rho[E]$ and $\rho[H]$ are enhanced up to 3 orders of magnitude at certain regions inside the sphere. $r_s = 365$ nm.

$$v_E = \frac{c_0}{f(W-1) + 1}, \quad (5.2.1)$$

where c_0 the speed of light in vacuum, f the volume fraction and W the energy density internal to the spheres relative to the surrounding medium (vacuum in our case). Our computations suggest that $W \sim 20$ for the first few heavy-photon modes, resulting in a velocity v_E that is much lower than one expects from a

straightforward estimate for the average velocity away from resonances,

$$v_{av} = \frac{c_0}{\sqrt{\tilde{\epsilon}_{av}}} = \frac{c_0}{\sqrt{f(\tilde{\epsilon} - 1) + 1}}, \quad (5.2.2)$$

or even from the velocity in the effective medium à la Garnett,

$$v_{eff} = \frac{c_0}{\sqrt{\tilde{\epsilon}_{eff}}}. \quad (5.2.3)$$

Thus for $f = 0.22$ the equations above give $v_E/c_0 \sim 0.19$, whereas $v_{av}/c_0 \sim 0.60$ and $v_{eff}/c_0 \sim 0.78$. For comparison, the group velocity $v_g = d\omega/dk$ varies with frequency and ranges from $0.02 c_0$ to $0.25 c_0$ for these resonances. The accumulation of energy in the system at the resonance condition means that the overall EM fields are enhanced[†], and this results in large forces when the stress-tensor integration is carried out.

5.2.6 Force strength

The magnitude of the photo-induced force is proportional to I_0 , the intensity of the incident beam. Provided the spheres are in a liquid medium which facilitates heat evacuation (so that melting is avoided), the photo-induced force can dominate gravitational, thermal-fluctuation and Van der Waals forces. Assuming a moderate laser of $I_0 \sim 3.5 \times 10^8 \text{ W/m}^2$, the incident radiation pressure is $\sim 1 \text{ pN}$ per unit cell ($900 \times 900 \text{ nm}^2$). For a crystal of one layer thickness, with $\tilde{\epsilon}_m = 1, \tilde{\epsilon}_s = 8.9, r_s = 350 \text{ nm}$, and when ω lies inside the second band gap (centred at $\sim 0.72 \text{ eV}$ in Fig. 5.5.a) the force on a unit cell is $\sim 2 \text{ pN}$ (the ‘natural limit’ for full reflection). In resonances too the total force on the crystal may not exceed this natural limit, and varies typically between 1 pN and 2 pN . However when more crystal layers are put together then the light-induced resonant interactions between the layers may easily be enhanced to 10 times or more the incident radiation pressure. Other forces present are Van der Waals, gravitational and thermal fluctuation forces due to Brownian motion of the spheres. The influence of thermal forces is negligible, as can be demonstrated by comparison between the thermal energy, $\frac{3}{2}k_B T \sim 6 \times 10^{-21} \text{ J}$, and the energy of each sphere in a b_1^1 Mie resonance, for which a rough under-estimate is $F_{z,min} \times D_s \sim 1 \text{ pN} \times 200 \text{ nm}$

[†]This mechanism has been known to explain the enhancement of the field on the surface of GaP particles used in SERS (Surface Enhanced Raman Scattering) experiments [94].

$\sim 2 \times 10^{-19}$ J. Gravitational effects are also small: $mg \sim 8 \times 10^{-3}$ pN for GaP. As for the Van der Waals attraction between two of our GaP spheres in air, an upper bound for the force, calculated in the basis of pairwise additivity [95], is ~ 0.2 pN. This value is clearly an overestimate, since it is obtained in the limit $D_s \ll r_s$ and ignoring retardation effects [95, 96]. Even the force between two GaP half-spaces (flat surfaces) separated by D_s (~ 0.4 pN per unit cell, according to the macroscopic Lifshitz theory incorporating retardation and non-additivity [69]) is at least an order of magnitude lower than the photo-induced force. So the photo-induced forces can be dominant. In fact both the magnitude and sharpness of the resonant forces increase when more crystal layers are added, as we shall see later (Figs. 5.29, 5.35).

5.2.7 Dependence on angle of incidence, absorption, polydispersion

The sharp force features shift slightly with the angle of incidence (Fig. 5.26). More importantly, they are sensitive to the amount of absorption present in the spheres or the surrounding medium: a moderate $\tilde{\epsilon}'' = 0.1$ is enough to suppress them. When both the spheres and the reference medium have low enough absorption the bonding effects are observable. Liquid media facilitate crystallisation by allowing the spheres to move slowly into an equilibrium structure; also by carrying away excess charges as well as heat which could otherwise accumulate and melt the spheres. For GaP spheres in water, we have found that the absorption is too small to blur the sharp features in forces, within the frequency range of interest (Fig. 5.27). Finally, one does not need perfect, identical spheres: objects of any shape possess their own Mie-like resonances and we have checked that the bonding effect can withstand a polydispersion of at least a few percent.

5.2.8 Bonding / anti-bonding effects

The effect of increasing thickness is of particular importance with reference to resonances. Let us look into the magnetic dipole resonance for the moment. When the number of layers is doubled, the number of sharp features doubles also (Fig. 5.28), the two new peaks being located slightly below (at $\omega_<$) and slightly above (at $\omega_>$) the original frequency.

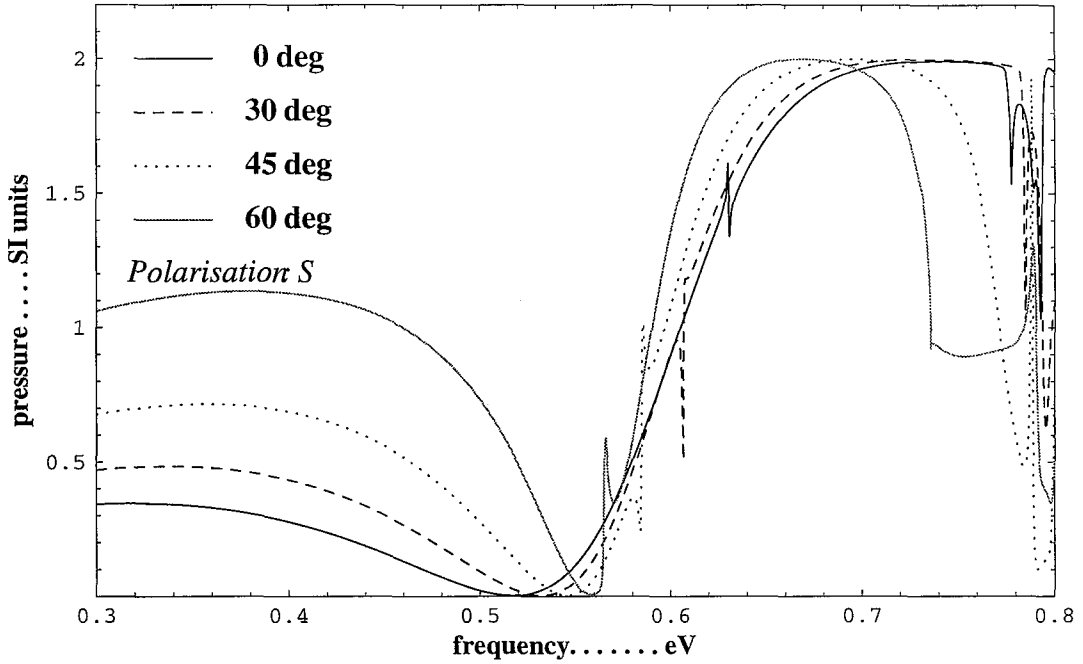


Figure 5.26: Influence of the angle of incidence θ on the normal force ($\hat{z}$ component). The resonance positions shift little with θ .

The total force on the structure is positive, so the light pushes the structure as it comes along, however the forces on an individual sphere of either layer alternate sign, exhibiting a bonding/anti-bonding behaviour[83, 89]: at $\omega_<$ the two layers attract each other, whereas at $\omega_>$ they repel each other (see the two first peaks at ~ 0.65 eV in Fig. 5.29; this calculation is made with the stress tensor method and the results are qualitatively correct, but at resonances quantitatively incorrect). The magnetic fields inside each sphere indicate that at $\omega_<$ the magnetic dipoles in the top layer are parallel to the dipoles in the bottom layer, whereas at $\omega_>$ they are antiparallel (Fig. 5.30). Thus it is possible to interpret the forces between any two vertically adjacent spheres in the following manner. The incident EM field populates the b_1^1 Mie mode of each sphere, which is thus made into a magnetic dipole. Due to symmetry, the magnetic dipoles of a given layer are oriented vertically (upwards or downwards) for a normally inci-

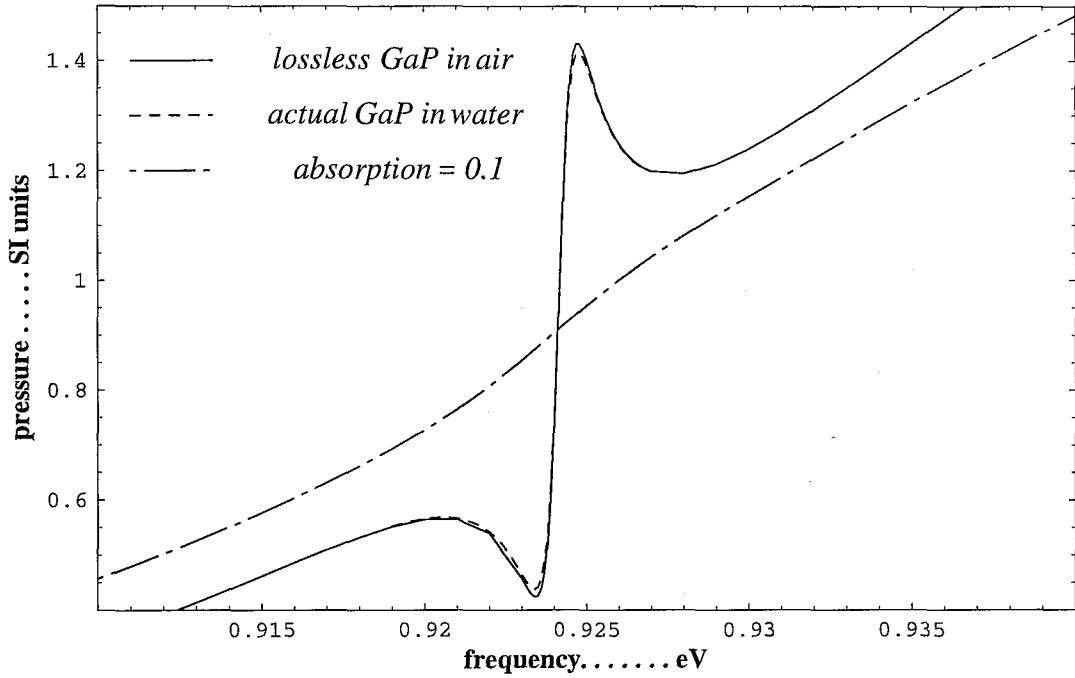


Figure 5.27: Dependence of the force on absorption. Away from resonances the forces do not suffer a sizeable change with moderate absorption. The resonances are sensitive to losses, but the actual amount of absorption present in GaP spheres and in an experimentally suitable liquid medium (in this case water) is small enough so the resonance effects can be clearly seen. Here we plot the normal pressure for the b_1^1 resonance on a crystal monolayer (GaP spheres, $r_s = 225$ nm) in water (dashed black line) for normal incidence. For comparison, the force on a hypothetical loss-free GaP sphere in air is also shown (solid black line). Although an amount of absorption $\epsilon_s'' = 0.1$ would suffice to mask the resonances (dash-dotted line), it is clear that GaP is a material with very low absorption in this frequency range and is therefore highly suitable for displaying the effects of resonant scattering.

dent beam, and they are all parallel to each other (since our system is periodic in the xy plane). When a second layer is added below the first, the dipoles of the new layer have the option to orient themselves parallel or anti-parallel to the dipoles of the top layer. In the former instance there is an attraction, same in nature as the attraction between the opposite poles of two magnets (although the

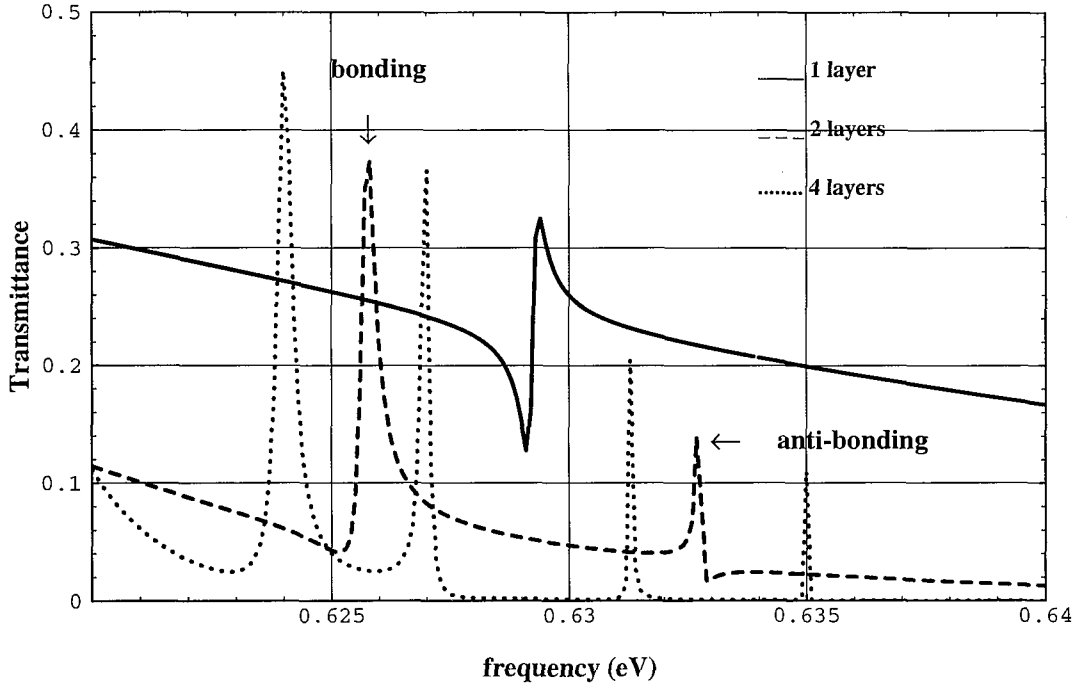


Figure 5.28: Transmittance for three crystal samples of increasing thickness from one to four unit cells in the z direction. The frequency spans the b_1^1 resonance. Normal incidence. $r_s = 365$ nm.

dipoles oscillate at the light frequency they remain always parallel and attract each other). Consequently the energy of the whole system decreases. In a similar manner when the dipoles of the bottom layer become anti-parallel to those of the top layer, repulsive interactions occur and the energy increases.

The same state of affairs occurs for the higher modes, whereby we see a series of alternating signs in the force on a sphere as different resonances are encountered (Fig. 5.29) for a crystal two layers thick. Thus we find that for the electric dipole (a_1^1) mode, all dipoles are parallel at $\omega = 0.7915$ eV, anti-parallel at $\omega = 0.8025$ for the system of Fig. 5.29. Similarly, the multipoles of the higher-energy modes can be oriented constructively, in which case an attraction occurs, or destructively, leading to repulsion between neighbouring spheres (Fig. 5.31). This argument holds also for the mixed hybrid states that are made out of more

than one Mie modes, for example at 0.8584 eV and 0.8744 eV of Fig. 5.29, as is shown below (Figs. 5.32, 5.33).

In Fig. 5.34 we show the same effects but for small spheres ($r_s = 220$ nm, $f = 0.06$), so the stress tensor calculation is in good shape. The pressure is in SI units and we verify that at resonances the pressure on the individual layers may well exceed the natural limit of $2P_{inc}$ ($= 2\text{N/m}^2$ for $I_0 = 3 \times 10^8 \text{ W/m}^2$), rising to 8 N/m^2 for the a_1^1 bonding mode for example (at 1.08 eV). The pressure for the b_1^1 modes (at ~ 0.94 eV) is down to $\sim 1\text{N/m}^2$, but for increasing filling fraction f it rises also. The reason for this is that at resonance there are strong evanescent fields excited within each sphere. At large distances these fields decay to zero and do not give rise to forces, but when another sphere exists nearby they may couple to it and produce a sizeable effect on the force, in a similar manner to what we found for the force between two half-spaces separated by a layer of air upon the incidence of evanescent waves.

Incidentally, the broad features around 1.01 eV and 1.2 eV in the forces on each layer of Fig. 5.34 are *not* resonances. They form as photons scatter multiply between the layers, banging as tennis balls against each layer and exiting on the lower side eventually with a unit transmittance. Hence they cause repulsion between the layers, although the total force on the crystal is zero. Being interference and not resonance peaks they are rather resistant to absorption: they survive to 60 % of their strength when $\tilde{\epsilon}'' = 0.1$.

It is therefore evident that degeneracy splitting of the sharp resonance features is directly analogous to the formation of bonding and anti-bonding states of the electronic orbitals in atomic physics. Our system of two oscillating multipoles (GaP spheres in a heavy photon mode) is the EM analogue of two Hydrogen atoms in the same state, brought close to each other. As they approach, hybridisation occurs and the energy levels split into a bonding state at slightly lower energy ($\omega_<$, multipoles ‘parallel’), and an anti-bonding state at slightly higher energy ($\omega_>$, multipoles ‘antiparallel’) than that of the un-hybridised levels.

The splitting of the energy levels and the force strength increase as the spheres of two vertically adjacent layers approach each other (Fig. 5.35). The nature of the sharp forces between adjacent layers therefore lies in the sphere-sphere interactions *induced* by the light beam, when the latter populates the various Mie resonances.

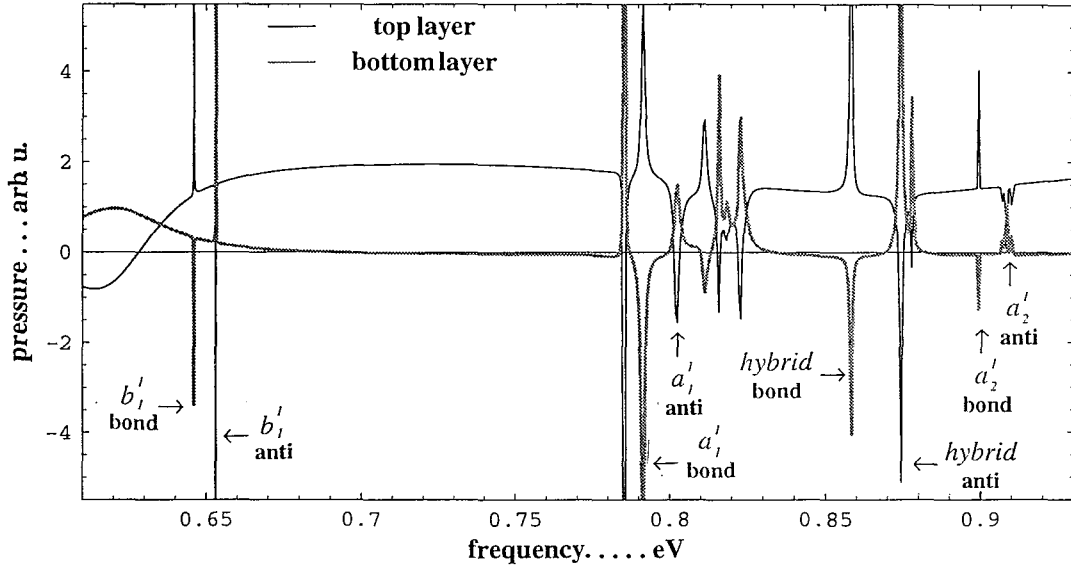


Figure 5.29: Normal pressure in arbitrary units on a two-layer crystal of GaP spheres ($\tilde{\epsilon} = 8.9$, $r_s = 350$ nm) in air for normal incidence along $\hat{z}$. The pressure on the top (bottom) layer is shown in solid black (grey) line; on both layers (*i.e.* total pressure) it is shown in a dotted line. The resonant forces on either layer are generally opposite in direction, peak more sharply than the total force and their sign alternates with frequency, exhibiting a series of bonding/anti-bonding interactions (*i.e.* layers attract/repel each other). At the bonding modes the spheres of the two layers behave as oscillating EM multipoles oriented parallel to each other, at the anti-bonding modes they are anti-parallel. Various modes are seen: magnetic dipole (b_1^1), electric dipole (a_1^1), electric quadrupole (a_2^1) and hybrids. Compare with the force on one layer of Fig. 5.5.

The analogy between resonant, light-induced, sphere-sphere interactions and the bonding of atoms that we just described is not the only one to be made. An analogy exists also with the case of cutting a metal in two along a plane, whereby the bulk plasmons reorganise into surface plasmons of attractive and repelling nature at frequencies below and above the original frequency[97]. The similarities between these three very different systems prompts us to acknowledge

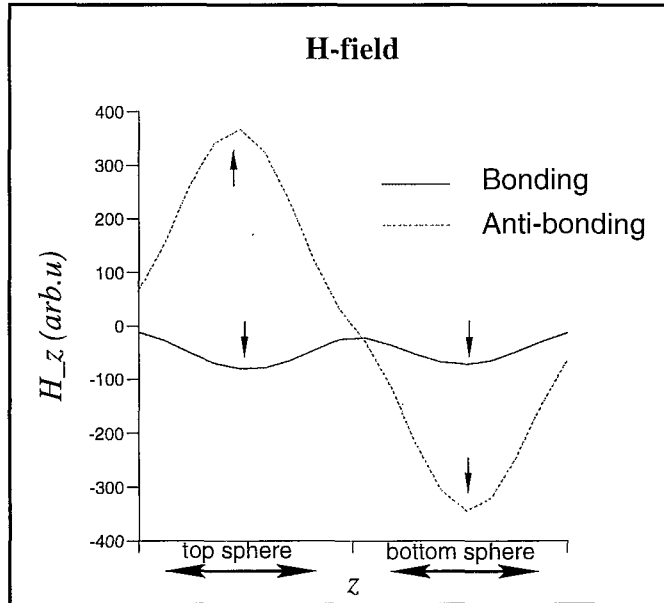


Figure 5.30: Magnetic fields plotted along the z axis through the centre of the spheres for the b_1^1 resonance on the two-layer crystal of Fig. 5.29. Light is s polarised, normally incident from the z direction at frequency $\omega_<$ (bonding, dipoles parallel), $\omega_>$ (anti-bonding, dipoles anti-parallel). For p polarisation the directions of the fields are reversed.

a general tendency in nature for the formation of energetically favourable EM bonds between two systems in the same state that come into proximity.

The rest of the force spectrum displays mostly smooth, attractive interactions due to the band gaps between the heavy-photon bands. This off-resonance attraction is produced as the band-gap condition for high reflectivity causes the top layer to absorb almost all the shock from the reflection of the incident light and be therefore pushed towards the bottom layer which remains relatively inert.

The occurrence of resonant bonding effects between spheres may add to the current methods for engineering 3D photonic crystals out of assemblies of colloidal spheres in liquid solutions. Experiments on gold nanospheres (radius $r_s \sim 10$ nm) by Kimura have shown fast formation of aggregates upon illumi-

nation at the surface-plasmon frequency (*i.e.* the electric-dipole mode a_1^1), in agreement with calculations of the photo-induced Van der Waals dipole-dipole interaction[98, 99]. Burns, Fournier and Golovchenko on the other hand have concentrated on dielectric microspheres ($r_s \sim 700$ nm)[5, 6]. They have experimentally demonstrated the presence of long-range attractive forces, induced by laser light, between two polystyrene spheres in water. They argue that these are magnetic-dipole interactions, owing their existence to induced currents on the spheres. They calculate the interaction energy between two magnetic dipoles based on a simplified model that works in the long-distance limit $R_{dip} \gg \lambda$, where R_{dip} the distance between the dipoles and λ the wavelength of light. From the perspective of Mie resonances this approach is justified because although the spheres Burns *et al* use possess many different resonances at or near the frequency of their laser light (see Fig. 5.4 and Table 5.2), the large inter-sphere distance ensures that the dipole approximation works to leading order. Also the electric-dipole solutions are of secondary importance since they are damped compared to the magnetic dipoles.

In contrast to the work of Kimura (metallic spheres) and that of Burns *et al* ($R_{dip} \gg \lambda$), we are dealing with *dielectric* spheres at *close proximity* ($R_{dip} \sim \lambda$). In general, the sphere-sphere interactions in our system display a *mixing* of EM-multipole behaviour, unless a specific condition for an isolated Mie resonance is met. For example two spheres of $r_s = 350$ nm interact at ~ 0.65 eV as oscillating magnetic dipoles, at ~ 0.80 eV as oscillating electric dipoles, at ~ 0.9 eV as oscillating electric quadrupoles and so on. Although the model of Burns, Fournier and Golovchenko does not apply to our system (due to the close proximity between the spheres), we believe that their physical explanation for the spatially oscillatory shear forces is basically correct: the oscillation is due to the changes in the phase shifts associated with retardation between spheres of different layers.

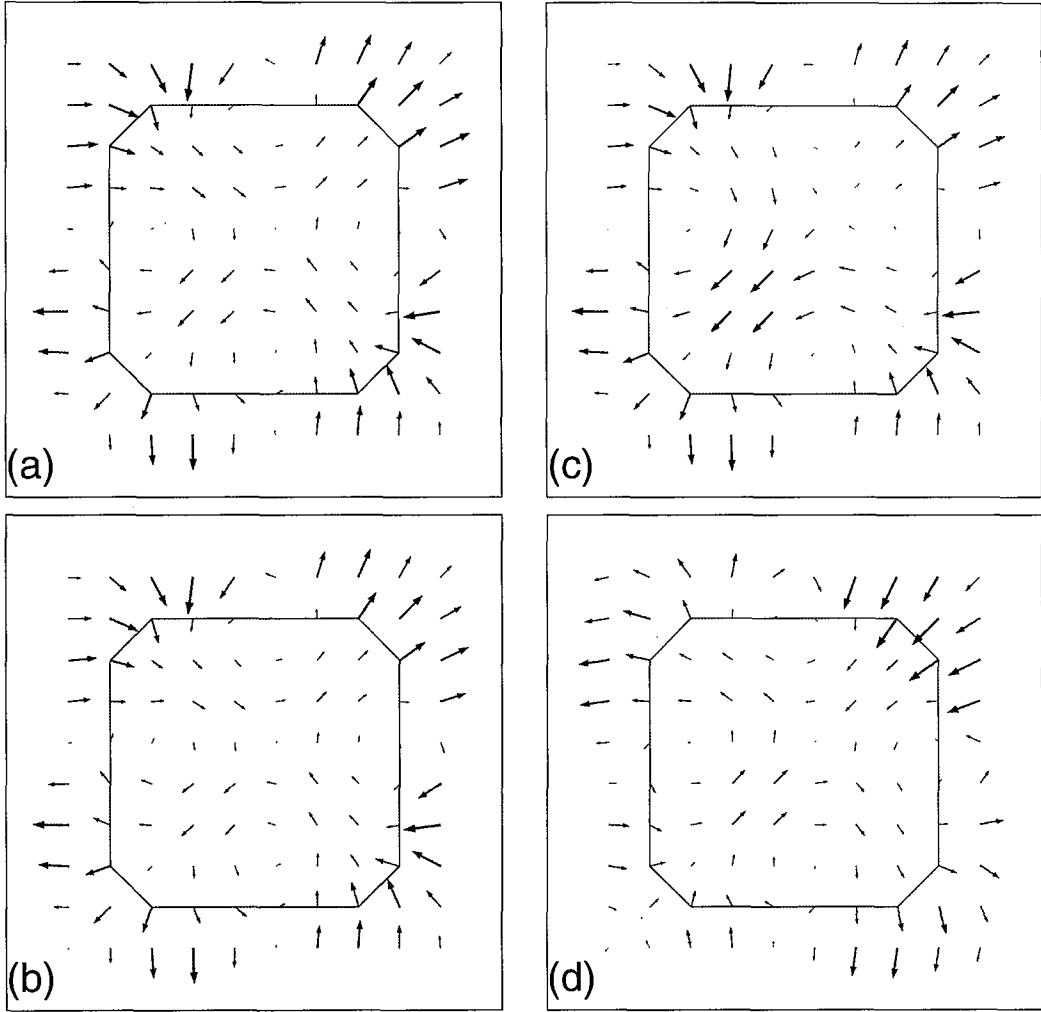


Figure 5.31: Bonding and anti-bonding for the electric quadrupole mode a_2^1 of Fig. 5.29. In all parts the $\mathbf{E}$ field is shown on the cross section S_3 . (a) Top sphere, bonding mode (0.8995 eV). (b) Bottom sphere, bonding mode. (c) Top sphere, anti-bonding mode (0.9068 eV). (d) Bottom sphere, anti-bonding mode. At the bonding mode the quadrupoles of the top and bottom spheres are ‘parallel’, at the anti-bonding mode anti-parallel. The boundaries of the dielectric object (mimicking a sphere) inside the unit cell are drawn for a mesh size $10 \times 10 \times 10$. An average- ϵ definition has been used.

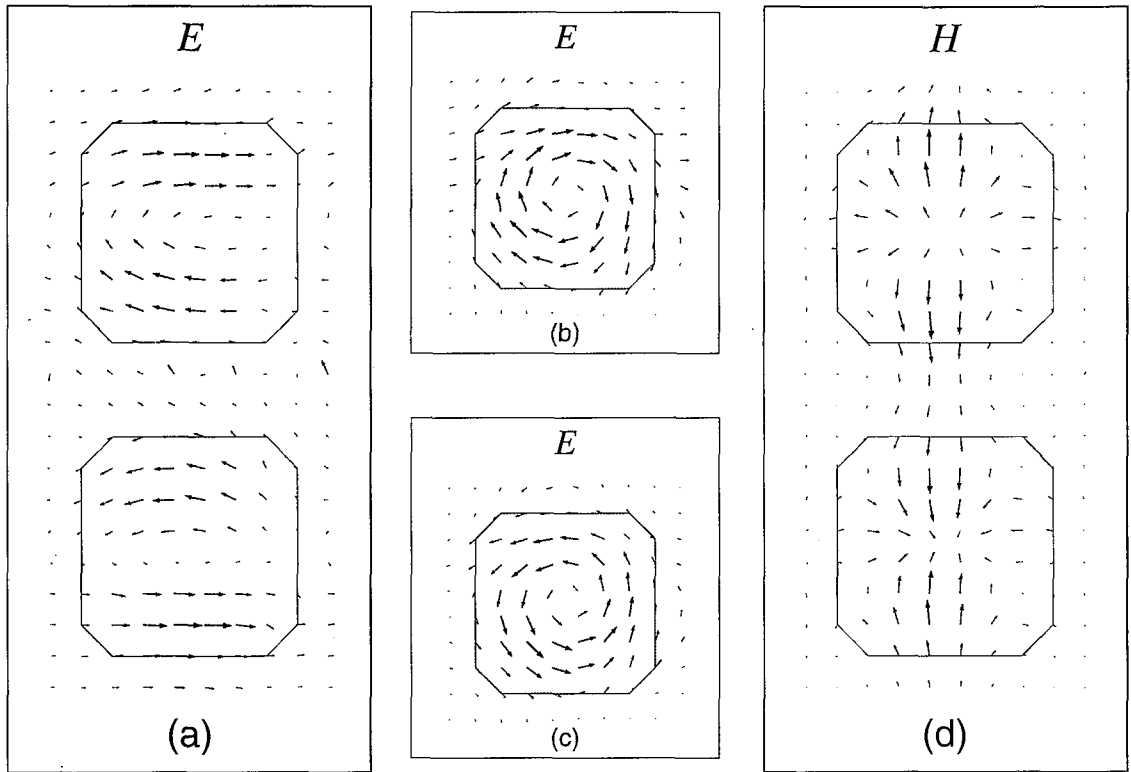


Figure 5.32: Hybrid bonding mode for the two-layer system of Fig. 5.29. $\omega_< = 0.8584$ eV, s polarisation. (a) $\mathbf{E}$ field along section S_1 . The field in the lower part of the top sphere is parallel to the field in the upper part of the bottom sphere. Hence constructive interference occurs. (b) $\mathbf{E}$ field along section S_3 for the top sphere. (c) $\mathbf{E}$ field along section S_3 for the bottom sphere. (d) $\mathbf{H}$ field along section S_2 . Again, it is favourable for the spheres to lie close together. The $\mathbf{E}$ fields in (b,c) rotate in opposite directions due to the fact that the $\mathbf{H}$ fields along the vertical axis are diverging from the sphere centre for the top layer, converging to the centre for the bottom layer.

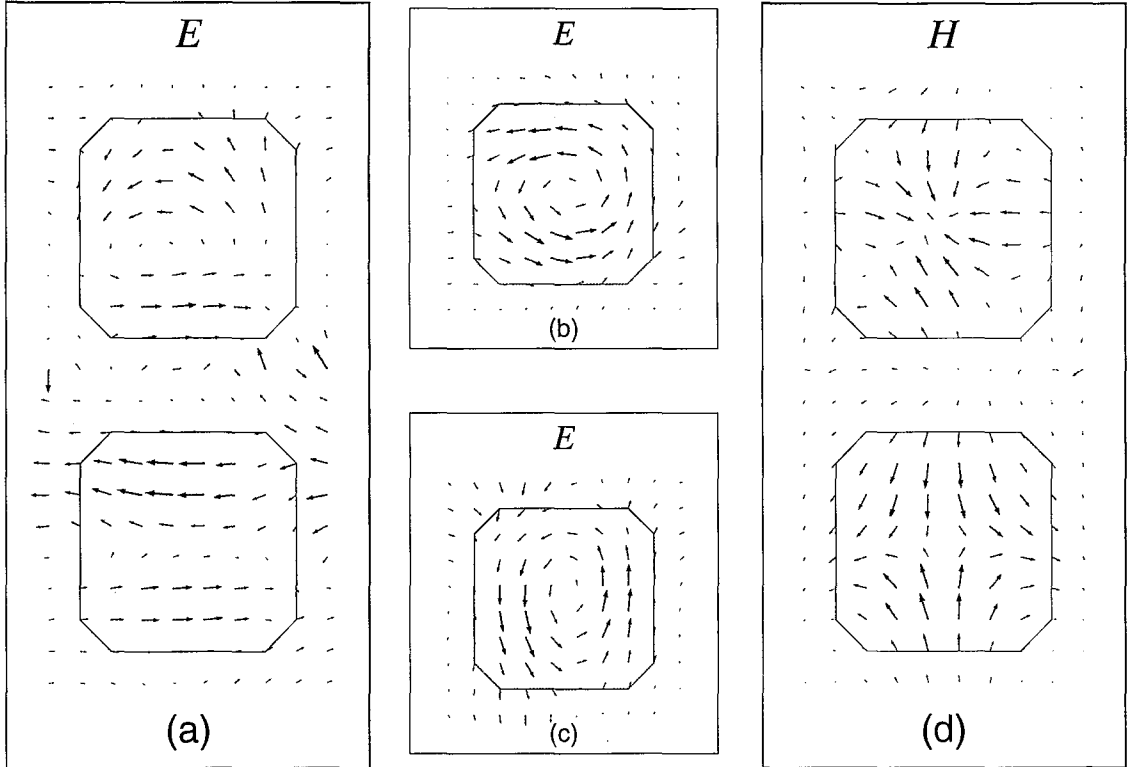


Figure 5.33: Hybrid anti-bonding mode for the two-layer system of Fig. 5.29. $\omega_{>} = 0.8744$ eV, p polarisation. (a) $\mathbf{E}$ field along section S_1 . The field in the lower part of the top sphere is anti-parallel to the field in the upper part of the bottom sphere. Hence destructive interference occurs. (b) $\mathbf{E}$ field along section S_3 for the top sphere. (c) $\mathbf{E}$ field along section S_3 for the bottom sphere. (d) $\mathbf{H}$ field along section S_2 . Due to the field orientations, it is unfavourable for the spheres to lie close together.

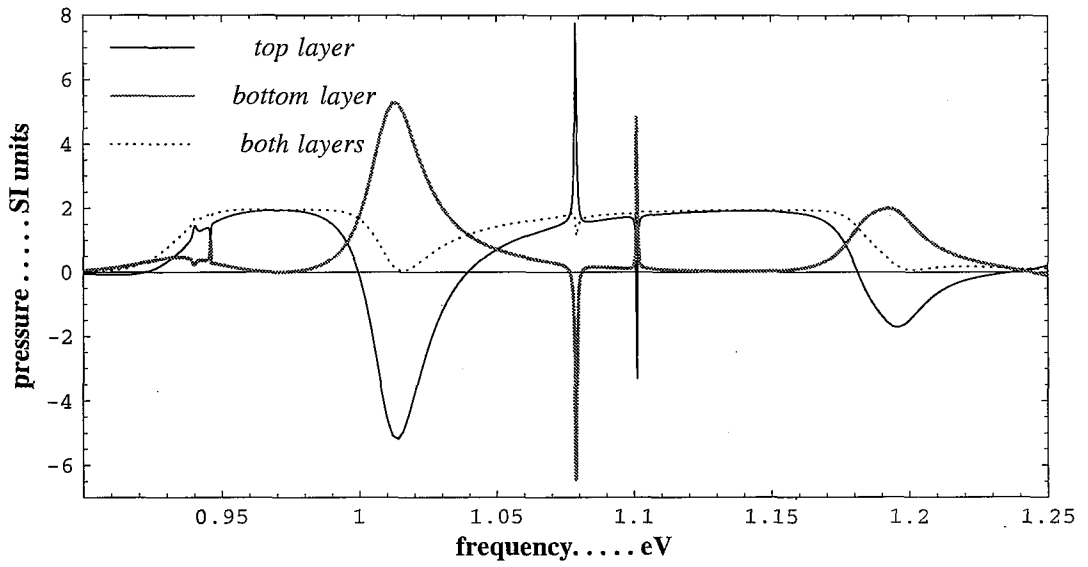


Figure 5.34: Normal pressure on a two-layer crystal of GaP spheres ($\tilde{\epsilon} = 8.9$, $r_s = 220$ nm) in air for normal incidence along $\hat{z}$. The bonding and anti-bonding b_1^1 modes are seen at ~ 0.94 eV, and the a_1^1 modes at 1.08 eV and 1.1 eV. The broad peaks of repulsion are due to multiple scattering between the layers. The units of pressure are in SI.

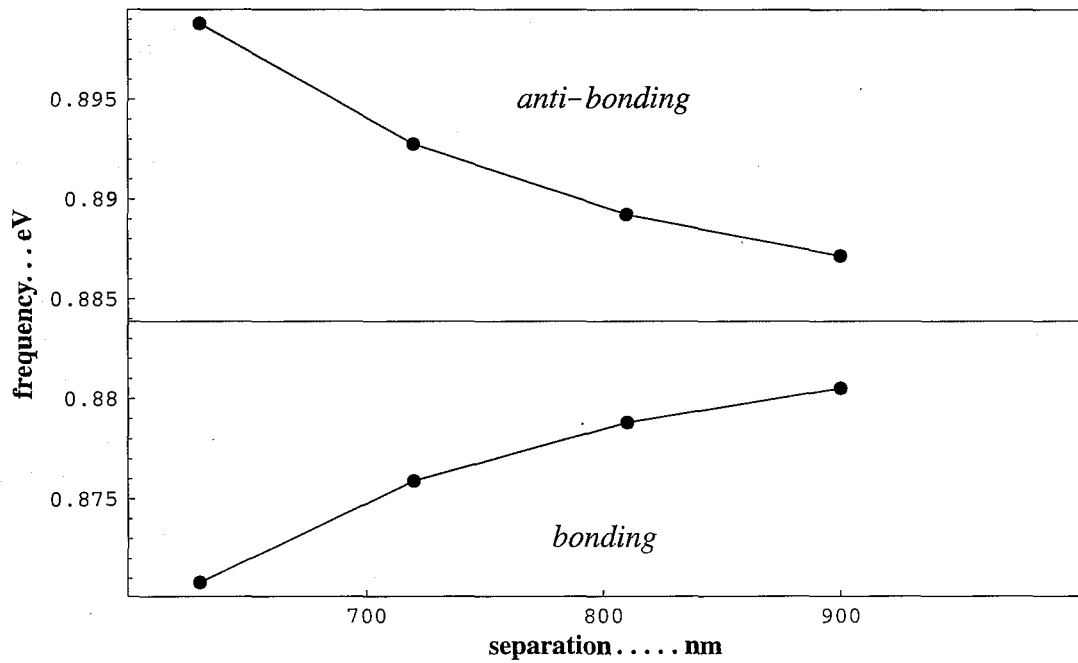


Figure 5.35: Resonant energies of a two-layer crystal for normal incidence along $\hat{z}$ and for the magnetic dipole mode. As the two crystal layers approach each other the bonding/anti-bonding energy levels of the Mie modes on the spheres split further. The dots denote the energies of the bonding and anti-bonding modes as a function of the distance of the layers. $r_s = 240$ and $l_0 = 900$ nm for each layer.

“Vain is the word of a philosopher which does not heal any suffering of man. For just as there is no profit in medicine if it does not expel the diseases of the body, so there is no profit in philosophy either, if it does not expel the suffering of the mind.”

Epicurus

Chapter 6

Summary and future work

6.1 Summary

In conclusion, we have presented a general methodology for accurate and efficient computation of EM fields and forces in matter, based on the Maxwell Stress Tensor formalism and on the Transfer Matrix approach to calculating reflection coefficients.

Our goal has been to understand light-induced forces in photonic crystals, with a view to self-assembly of colloidal systems.

In chapter 2 we outline our theoretical and numerical approach. We explain how the need for a stress tensor arises in calculating EM forces, and adopt a particular form for the tensor appropriate for the systems we have in mind: photonic crystals in which both the artificial atoms and the reference medium are homogeneous, isotropic, non-pyroelectric and non-piezoelectric and ignoring electrostriction and magnetostriction effects. Mechanical and thermal equilibrium is also assumed. Under these restrictions the stress tensor that originates from static or time-averaged harmonic fields yields the full EM force on a macroscopic body when integrated over the body’s external surface.

We present some necessary technical details of the numerical calculation of EM forces, including the normalisation of the fields and the conversion from atomic units to SI. We relate the forces to experimental parameters when the fields are induced by the incidence of external laser radiation.

In chapter 3 we study the EM forces induced by a monochromatic light beam on homogeneous systems (dielectric and metallic): (i) a half-space and (ii) a slab. The simplicity of these systems allowed us to understand the nature of

forces at interfaces and the role of evanescent fields in producing attraction across a cavity separating two bodies. We analytically solve for the fields and forces in these systems, testing the accuracy of our numerical approach as well. We identify the natural limits of the force strength for non-resonant scattering. We find that in passing from a medium ϵ_1 to a medium ϵ_2 , light induces an attractive surface force on the interface when $\epsilon_2 > \epsilon_1$. But for total forces there is a distinction between the type of light-wave used: travelling waves always produce positive pressure on both the slab and the half-space. Evanescent waves, however, under specific conditions cause the two half-spaces (on either side of the slab) to attract each other with a force strength even greater than the Van der Waals attraction between them. Evanescent waves look promising in the application to cohesive forces in nanostructures.

In chapters 4 and 5 we concentrate on the forces induced by a laser beam of intensity I_0 on a dielectric crystal of GaP spheres arranged in a SC lattice. Chapter 4 deals with non-resonant effects: the long wavelength effective-medium approach, the influence of band gaps on total forces on the crystal, the onset of scattering into multiple outgoing beams. In chapter 5 we explore the large resonant forces at frequencies corresponding to the EM eigenmodes of isolated spheres (Mie resonances). After explaining the nature of the Mie modes for one sphere, we study the properties of the corresponding modes in the crystal (heavy photon resonances). We show how they may be identified in the reflectance spectra. Next we study how they are affected by the filling fraction, the size and shape of the artificial atoms, the size of the discretisation mesh and the absorption in the crystal. These resonances cause a large enhancement of the EM energy inside and around the spheres (of 2-3 orders of magnitude). Generally they are electric or magnetic multipoles (or hybrids) oscillating at the light frequency. An analogy to atomic physics proves fruitful as the resonant EM fields on the spheres arrange into bonding and anti-bonding states, causing highly attractive and repelling interactions which may become dominant over gravitational, thermal and Van der Waals forces, for large enough (but realistic) values of I_0 . Although most of our results are for GaP spheres suspended in air, we have checked that the finite absorption of a more realistic medium such as water does not obliterate these resonant effects which we believe therefore to be experimentally observable and relevant to the formation of stable crystals of colloidal nanospheres.

6.2 Future problems to be addressed

The subject of EM forces in nanostructures is a very wide one. Although some analytical work on Van der Waals forces has been carried out over the past decades, there is still considerable difficulty in solving for interactions between two macroscopic bodies with arbitrary geometry and dielectric properties; and a lot remains to be done on assemblies of bodies (such as a colloidal crystals) beyond the weak-interacting limit. Experimentally, there is a large selection of systems where EM forces play a dominant role, awaiting proper theoretical understanding. We think that extending the work we presented here may have a significant impact on the subject, since we are in principle able to calculate numerically the full EM force on both dielectric and metallic systems of frequency-dependent dielectric function; as well as to objects of any size and geometrical properties.

A short-term list of potentially interesting problems to be addressed follows:

- Adapting the codes to calculate accurately the surface forces on the artificial atoms. How are these forces influenced as the photonic band gap is traversed in frequency ?
- Treating metallic or metallodielectric photonic crystals.
- The influence of many laser beams on forces. What is the combined effect of standing wave patterns and resonant scattering ?
- Developing a time-dependent picture of the force field, following the artificial atoms until they assume their equilibrium positions under the influence of external radiation.

All the above involve one (or a discrete set of) frequencies. For Van der Waals calculations, integration over all frequencies is needed. Having developed the appropriate formalism and computer codes one could tackle the following (long-term) issues:

- The force-distance law in the AFM, especially with application to lateral resolution as a function of distance. The theory can be formulated in exact parallel to earlier STM theory with electron reflection coefficients replaced by photon reflection coefficients.
- Forces in systems where resonant enhancement is extreme: for example metal tips on rough silver surfaces as in the giant Raman resonance experiment. What parallels can we find in the AFM problem?

- The influence of illumination by powerful laser beams on forces in the AFM, especially in highly resonant systems.
- Investigation of whether the AFM/STM in conjunction with laser illumination can provoke strong non-linear optical phenomena.
- Theory of the light microscope and study of whether resolution can be enhanced by exploiting resonant phenomena to compress the range of the electromagnetic wavefield in the vicinity of the tip.

Appendix A

List of symbols

Symbol	Name
a_l^{mi}	electric multipole natural mode
a_l^i	electric multipole Mie mode
a_B	Bohr magneton
$\hat{A}$	complex amplitude of incident beam on slab
b_l^{mi}	magnetic multipole natural mode
b_l^i	magnetic multipole Mie mode
$\mathbf{B}$	total magnetic field
$\hat{B}$	complex amplitude of upward beam in region I (for slab)
c	speed of light
c_0	speed of light in vacuum
$\hat{C}$	complex amplitude of downward beam in region II (for slab)
C_{sca}	scattering cross section
C_{ext}	extinction cross section
$\mathbf{D}$	displacement field
$\hat{D}$	complex amplitude of upward beam in region II (for slab)
D_s	distance between two nearest spherical surfaces
e	electronic charge
$\hat{E}$	complex amplitude of downward beam in region III (for slab)
$\hat{\mathbf{e}}_{\mathbf{k}'\sigma'}$	unit vector of beam with $\mathbf{k}'$, σ'
$\mathbf{E}$	electric field
E_0	amplitude of incident electric field
f	volume fraction, packing fraction or filling fraction
$\mathbf{f}_v$	volume force
$\mathbf{F}$	total force
$\mathbf{g}$	EM momentum density

Symbol	Name
$\mathbf{g}_{\parallel}$	2D reciprocal lattice vector
$\mathbf{G}$	3D reciprocal lattice vector
$\hbar$	Planck's constant
$\mathbf{H}$	magnetic field
H_0	amplitude of incident magnetic field
Hartree	13.6 eV or one-half of the atomic unit for energy
I_0	light intensity of incident beam
$\mathbf{J}_{ext}$	volume density of external (<i>i.e.</i> free) currents
$\mathbf{J}_{tot}$	volume density of total (<i>i.e.</i> free + induced) currents
c_0	speed of light in vacuum
$\mathbf{k}$	wavevector of incident beam on a crystal
$\mathbf{k}'$	wavevector of scattered beam off a crystal
$\mathbf{k}_{\parallel}$	component of $\mathbf{k}$ parallel to xy plane
$\mathbf{k}_{inc}$	incident wavevector on interface between two homogeneous media
$\mathbf{k}_{ref}$	reflected wavevector on interface between two homogeneous media
$\mathbf{k}_{tr}$	transmitted wavevector on interface between two homogeneous media
$\mathbf{k}_1$	wavevector in region I (with z-component > 0)
$\mathbf{k}_2$	wavevector in region II (with z-component > 0)
$\mathbf{k}_3$	wavevector in region III (with z-component > 0)
l	principal quantum number
l_0	lattice constant (unit-cell length)
m	refractive index ratio between sphere and reference medium
m_l	azimuthal quantum number
$\mathbf{p}$	EM momentum
$\mathbf{p}_{r;k'}$	EM momentum of reflected beam with $\mathbf{k}'$
$\mathbf{p}_{t;k}$	EM momentum of transmitted beam with $\mathbf{k}'$
$\mathbf{p}_{i;k}$	EM momentum of incident beam with $\mathbf{k}$
P	radiation pressure on a sphere for Mie scattering
P_{inc}	radiation pressure of incident light beam
P_z	normal pressure (along $\hat{\mathbf{z}}$)
$P_{z,vol}$	normal pressure (along $\hat{\mathbf{z}}$) on a volume element
$P_{z,int}$	normal pressure (along $\hat{\mathbf{z}}$) at interface
$P_{z,tot}$	normal pressure (along $\hat{\mathbf{z}}$) on a body
r	intensity of reflected light
$r_{\rightarrow}$	intensity of reflected light travelling rightwards
$r_{\leftarrow}$	intensity of reflected light travelling leftwards
r_s	radius of sphere
R_{dip}	dipole-dipole distance
$R_{\mathbf{k}'\sigma'}$	complex reflection coefficient
$R_{\sigma\sigma'}$	complex reflection coefficient for scattering from polarisation σ to σ'

Symbol	Name
$\mathbf{S}_P$	Poynting vector
$\mathbf{S}_{sca}$	Poynting vector of the scattered radiation
$\mathbf{S}_{ext}$	Poynting vector of the extinct (scattered + absorbed) radiation
$\mathbf{S}$	integration surface for force
S_1	xz section through the centre of sphere
S_2	xz section through the centre of sphere
S_3	xy section through the centre of sphere
t	intensity of transmitted light
$t_{\rightarrow}$	intensity of transmitted light travelling rightwards
$t_{\leftarrow}$	intensity of transmitted light travelling leftwards
T_{ij}	Maxwell Stress Tensor
$\langle T_{ij} \rangle$	time-averaged Maxwell Stress Tensor
$T_{\sigma\sigma'}$	complex transmission coefficient for scattering from polarisation σ to σ'
U	EM energy
v_E	velocity of energy transport
W	energy density ratio between interior and exterior of spheres
x	order parameter
α	phase shift between E_x and E_z for p scattering from a half-space
β	component of wavevector parallel to xy plane
γ_D	coefficient of damping for GaP
$\gamma_1 = \gamma'_1$	component of $\mathbf{k}_1$ normal to xy plane
$\gamma_2 = \gamma'_2 + i\gamma''_2$	component of $\mathbf{k}_2$ normal to xy plane
$\gamma_3 = \gamma'_3 + i\gamma''_3$	component of $\mathbf{k}_3$ normal to xy plane
δ	displacement variable
δ_{ij}	Kronecker delta function
$\epsilon = \epsilon' + i\epsilon''$	permittivity
$\tilde{\epsilon}$	relative permittivity
ϵ_0	permittivity of vacuum
ϵ_1	permittivity of medium I
ϵ_2	permittivity of medium II
ϵ_3	permittivity of medium III
ϵ_{av}	average permittivity in a unit cell
ϵ_{eff}	permittivity of effective medium
ϵ_m	permittivity of reference medium
ϵ_s	permittivity of sphere
θ_0	angle for total internal reflection between media I and III
θ_1	angle of incidence from medium I to medium II
θ_2	angle of refraction from medium I to medium II
θ_3	angle of refraction from medium II to medium III
$\cos \theta$	asymmetry factor

Symbol	Name
λ	wavelength of light
μ	permeability
$\tilde{\mu}$	relative permeability
μ_0	permeability of vacuum
ρ	EM energy density
$\rho[E]$	electric energy density
$\rho[H]$	magnetic energy density
ρ_{ext}	volume density of external (<i>i.e.</i> free) charges
ρ_{tot}	volume density of total (<i>i.e.</i> free + induced) charges
σ	polarisation of incident beam (<i>s</i> or <i>p</i>)
σ'	polarisation of scattered beam (<i>s</i> or <i>p</i>)
σ_c	electrical conductivity
ϕ	phase of T_{sp}
ϕ_1	phase of reflected wave from a half-space
ϕ_2	phase of transmitted wave into a half-space
ψ	phase of R_{sp}
ω	light frequency
$\omega_<$	light frequency at bonding mode
$\omega_>$	light frequency at anti-bonding mode
ω_p	plasma frequency

Appendix B

Derivation of the Stress Tensor

We start from Eq. (2.1.6) for the the EM force acting on the body:

$$\langle \mathbf{F} \rangle = \int \left\langle \rho_{ext} \mathbf{E} - \frac{1}{2} E^2 \nabla \epsilon + \mathbf{J}_{ext} \times \mathbf{B} - \frac{1}{2} H^2 \nabla \mu \right\rangle d^3 r, \quad (\text{B.0.2})$$

having ignored the time-dependent term since we are interested in time-averaged forces caused by harmonic wavefields. In this case all terms with a time-derivative give zero when averaged *i.e.* we are in the quasi-stationary regime (electrostatics and magnetostatics combined). We use two of Maxwell's equations,

$$\nabla \cdot (\epsilon \mathbf{E}) = \rho_{ext} \quad (\text{B.0.3})$$

and

$$\nabla \times \mathbf{H} = \frac{\partial(\epsilon \mathbf{E})}{\partial t} + \mathbf{J}_{ext} \quad (\text{B.0.4})$$

to rewrite the force in terms of the fields only:

$$\langle \mathbf{F} \rangle = \int \left\langle \left[\nabla \cdot (\epsilon \mathbf{E}) \right] \mathbf{E} - \frac{1}{2} E^2 \nabla \epsilon + (\nabla \times \mathbf{H}) \times \mathbf{B} - \frac{1}{2} H^2 \nabla \mu \right\rangle d^3 r, \quad (\text{B.0.5})$$

where again we left out the time-dependent term that averages to zero. To find the form of the stress tensor we need to bring the integrand above into the form of a divergence of a quantity which we may then identify T_{ij} . We re-express some of the terms above,

$$\left\langle E^2 \nabla \epsilon \right\rangle = \left\langle \nabla(\epsilon E^2) - \epsilon \nabla E^2 \right\rangle \quad (\text{B.0.6})$$

$$= \left\langle \nabla(\epsilon E^2) - \epsilon \left[2(\mathbf{E} \cdot \nabla) \mathbf{E} + 2\mathbf{E} \times (\nabla \times \mathbf{E}) \right] \right\rangle, \quad (\text{B.0.7})$$

having used the identity

$$\nabla(\mathbf{A} \cdot \mathbf{B}) = (\mathbf{A} \cdot \nabla)\mathbf{B} + \mathbf{A} \times (\nabla \times \mathbf{B}) + (\mathbf{B} \cdot \nabla)\mathbf{A} + \mathbf{B} \times (\nabla \times \mathbf{A}). \quad (\text{B.0.8})$$

We may use one of Maxwell's equations,

$$\nabla \times \mathbf{E} = -\frac{\partial(\mu\mathbf{H})}{\partial t}, \quad (\text{B.0.9})$$

to bring out the time-dependence in the last term of Eq. (B.0.7):

$$\left\langle E^2 \nabla \epsilon \right\rangle = \left\langle \nabla(\epsilon E^2) - 2(\epsilon \mathbf{E} \cdot \nabla)\mathbf{E} + 2\epsilon \mathbf{E} \times \frac{\partial(\mu\mathbf{H})}{\partial t} \right\rangle \quad (\text{B.0.10})$$

$$= \left\langle \nabla(\epsilon E^2) - 2(\epsilon \mathbf{E} \cdot \nabla)\mathbf{E} \right\rangle, \quad (\text{B.0.11})$$

where we have invoked the time-averaging process to drop the time-dependent term. Hence for the j -th component we have

$$\left\langle [\nabla \cdot (\epsilon \mathbf{E})] \mathbf{E} - \frac{1}{2} E^2 \nabla \epsilon \right\rangle_j = \left\langle [\nabla \cdot (\epsilon \mathbf{E})] \mathbf{E} - \frac{1}{2} \nabla(\epsilon E^2) + (\epsilon \mathbf{E} \cdot \nabla)\mathbf{E} \right\rangle_j \quad (\text{B.0.12})$$

$$= \left\langle \sum_i \left\{ [\nabla_i(\epsilon E_i)] E_j + \epsilon E_i [\nabla_i E_j] \right\} + (-)\frac{1}{2} \nabla_j(\epsilon E^2) \right\rangle \quad (\text{B.0.13})$$

$$= \left\langle \sum_i \nabla_i(\epsilon E_i E_j) - \frac{1}{2} \nabla_j(\epsilon E^2) \right\rangle \quad (\text{B.0.14})$$

$$= \sum_i \nabla_i \left\langle \epsilon E_i E_j - \frac{1}{2} \delta_{ij} \epsilon E^2 \right\rangle. \quad (\text{B.0.15})$$

Now for the magnetic part we re-express

$$\left\langle H^2 \nabla \mu \right\rangle = \left\langle \nabla(\mu H^2) - \mu \nabla H^2 \right\rangle \quad (\text{B.0.16})$$

$$= \left\langle \nabla(\mu H^2) - \mu [2(\mathbf{H} \cdot \nabla)\mathbf{H} + 2\mathbf{H} \times (\nabla \times \mathbf{H})] \right\rangle, \quad (\text{B.0.17})$$

so that

$$\left\langle (\nabla \times \mathbf{H}) \times \mathbf{B} - \frac{1}{2} H^2 \nabla \mu \right\rangle_j = \left\langle (\nabla \times \mathbf{H}) \times \mu \mathbf{H} - \frac{1}{2} \nabla(\mu H^2) + \right.$$

$$+(\mu \mathbf{H} \cdot \nabla) \mathbf{H} + \mu \mathbf{H} \times (\nabla \times \mathbf{H}) \Big\rangle_j \quad (\text{B.0.18})$$

$$= \left\langle (\mu \mathbf{H} \cdot \nabla) \mathbf{H} - \frac{1}{2} \nabla_j (\mu H^2) \right\rangle_j \quad (\text{B.0.19})$$

$$= \sum_i \left\langle \mu H_i [\nabla_i H_j] - \frac{1}{2} \delta_{ij} \nabla_i (\mu H^2) \right\rangle. \quad (\text{B.0.20})$$

We can bring this into the desired form by adding on both sides the divergence of $\mathbf{B}$ which is zero:

$$\begin{aligned} \left\langle (\nabla \times \mathbf{H}) \times \mathbf{B} - \frac{1}{2} H^2 \nabla \mu \right\rangle_j + 0 &= \left\langle \sum_i \left\{ \mu H_i [\nabla_i H_j] - \frac{1}{2} \delta_{ij} \nabla_i (\mu H^2) \right\} + \right. \\ &\quad \left. + \sum_i [\nabla_i (\mu H_i)] H_j \right\rangle \end{aligned} \quad (\text{B.0.21})$$

$$= \sum_i \left\langle \nabla_i (\mu H_i H_j) - \frac{1}{2} \delta_{ij} \nabla_i (\mu H^2) \right\rangle \quad (\text{B.0.22})$$

$$= \sum_i \nabla_i \left\langle \mu H_i H_j - \frac{1}{2} \delta_{ij} \mu H^2 \right\rangle. \quad (\text{B.0.23})$$

Adding the electrostatic and magnetostatic part we therefore obtain

$$\langle F_j \rangle = \int \left\langle \rho_{ext} \mathbf{E} - \frac{1}{2} E^2 \nabla \epsilon + \mathbf{J}_{ext} \times \mathbf{B} - \frac{1}{2} H^2 \nabla \mu \right\rangle_j d^3 r \quad (\text{B.0.24})$$

$$= \int \sum_i \nabla_i \left\langle \epsilon E_i E_j + \mu H_i H_j - \frac{1}{2} \delta_{ij} (\epsilon E^2 + \mu H^2) \right\rangle d^3 r, \quad (\text{B.0.25})$$

which is expressed as a volume integral of the divergence of a quantity that we identify as the stress tensor

$$\langle T_{ij} \rangle = \left\langle \epsilon E_i E_j + \mu H_i H_j - \frac{1}{2} \delta_{ij} (\epsilon E^2 + \mu H^2) \right\rangle. \quad (\text{B.0.26})$$

Appendix C

Mie resonances

The vector spherical harmonics are given here:

$$\mathbf{M}_{ol1} = \cos \phi \pi_l(\cos \theta) z_l(\rho) \hat{\mathbf{e}}_\theta - \sin \phi \tau_l(\cos \theta) z_l(\rho) \hat{\mathbf{e}}_\phi \quad (\text{C.0.2})$$

$$\mathbf{M}_{el1} = -\sin \phi \pi_l(\cos \theta) z_l(\rho) \hat{\mathbf{e}}_\theta - \cos \phi \tau_l(\cos \theta) z_l(\rho) \hat{\mathbf{e}}_\phi \quad (\text{C.0.3})$$

$$\begin{aligned} \mathbf{N}_{ol1} = & \sin \phi l(l+1) \sin \theta \pi_l(\cos \theta) \frac{z_l(\rho)}{\rho} \hat{\mathbf{e}}_r \\ & + \sin \phi \tau_l(\cos \theta) \frac{[\rho z_l(\rho)]'}{\rho} \hat{\mathbf{e}}_\theta \\ & + \cos \phi \pi_l(\cos \theta) \frac{[\rho z_l(\rho)]'}{\rho} \hat{\mathbf{e}}_\phi \end{aligned} \quad (\text{C.0.4})$$

$$\begin{aligned} \mathbf{N}_{el1} = & \cos \phi l(l+1) \sin \theta \pi_l(\cos \theta) \frac{z_l(\rho)}{\rho} \hat{\mathbf{e}}_r \\ & + \cos \phi \tau_l(\cos \theta) \frac{[\rho z_l(\rho)]'}{\rho} \hat{\mathbf{e}}_\theta \\ & - \sin \phi \pi_l(\cos \theta) \frac{[\rho z_l(\rho)]'}{\rho} \hat{\mathbf{e}}_\phi, \end{aligned} \quad (\text{C.0.5})$$

where $\rho = k r$ and the π_l, τ_l functions are defined below. The spherical coordinates basis vectors are of course

$$\hat{\mathbf{e}}_r = \hat{\mathbf{x}} \sin \theta \cos \phi + \hat{\mathbf{y}} \sin \theta \sin \phi + \hat{\mathbf{z}} \cos \theta \quad (\text{C.0.6})$$

$$\hat{\mathbf{e}}_\theta = \hat{\mathbf{x}} \cos \theta \cos \phi + \hat{\mathbf{y}} \cos \theta \sin \phi - \hat{\mathbf{z}} \sin \theta \quad (\text{C.0.7})$$

$$\hat{\mathbf{e}}_\phi = -\hat{\mathbf{x}} \sin \phi + \hat{\mathbf{y}} \cos \phi. \quad (\text{C.0.8})$$

Superscripts in $\mathbf{M}, \mathbf{N}$ denote the kind of the spherical Bessel function $z_l(\rho)$: (1) denotes $j_l(\rho)$ (Bessel function of the first kind), (3) denotes $h_l^{(1)}(\rho)$ (Bessel function of the third kind). It is seen from these relations that $\mathbf{M}$ has no radial part,

whereas for sufficiently large ρ the radial part of $\mathbf{N}$ is negligible compared to the transverse part. The angular functions are defined as

$$\pi_l(\cos \theta) = \frac{P_l^1(\cos \theta)}{\sin \theta} \quad (\text{C.0.9})$$

$$\tau_l(\cos \theta) = \frac{dP_l^1(\cos \theta)}{d\theta}, \quad (\text{C.0.10})$$

and can be computed from the recurrent relations

$$\pi_l = \frac{2l-1}{l-1} \cos \theta \pi_{l-1} - \frac{l}{l-1} \pi_{l-2} \quad (\text{C.0.11})$$

$$\tau_l = l \cos \theta \pi_l - (l+1) \pi_{l-1}, \quad (\text{C.0.12})$$

with $\pi_0 = 0, \pi_1 = 1$. The scattering coefficients appearing in the expressions for the fields are

$$a_l = \frac{\mu_{med} m^2 j_l(mx) [x j_l(x)]' - \mu_s j_l(x) [mx j_l(mx)]'}{\mu_{med} m^2 j_l(mx) [x h_l^{(1)}(x)]' - \mu_s h_l^{(1)}(x) [mx j_l(mx)]'} \quad (\text{C.0.13})$$

$$b_l = \frac{\mu_s j_l(mx) [x j_l(x)]' - \mu_{med} j_l(x) [mx j_l(mx)]'}{\mu_s j_l(mx) [x h_l^{(1)}(x)]' - \mu_{med} h_l^{(1)}(x) [mx j_l(mx)]'} \quad (\text{C.0.14})$$

$$c_l = \frac{\mu_s j_l(x) [x h_l^{(1)}(x)]' - \mu_s h_l^{(1)}(x) [x j_l(x)]'}{\mu_s j_l(mx) [x h_l^{(1)}(x)]' - \mu_{med} h_l^{(1)}(x) [mx j_l(mx)]'} \quad (\text{C.0.15})$$

$$d_l = \frac{\mu_s m j_l(x) [x h_l^{(1)}(x)]' - \mu_s m h_l^{(1)}(x) [x j_l(x)]'}{\mu_{med} m^2 j_l(mx) [x h_l^{(1)}(x)]' - \mu_s h_l^{(1)}(x) [mx j_l(mx)]'}. \quad (\text{C.0.16})$$

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...μά τό φῶς, τέλος, δέν βιώνεται
μέ τήν κατανόηση κβαντικῶν μεταβάσεων
ἀλλά μέσ' ἀπό τά μάτια της σάν τοῦ γελαῖ
ἤ κι ἀπό τίς σκιές στόν τοῖχο τῶν σωμάτων
ὅταν τήν κάνει δική του τίς νύχτες
πλάι στό μάτι τοῦ κεριού.
Κι ἀκόμα ἐτοῦτο·
πώς ἡ ζωή πληρώνεται
μέ τό γαλάζιο τοῦ ἀνοιχτοῦ πελάγου
πού βγάζει στόν ἥλιο
ἰδωμένο κάτω ἀπ' τά κύματα
τήν ὥρα τοῦ μικροῦ ἀπογεύματος
ἐνῶ τά μέλη μου γυμνά ἀφήνονται
νά ταυτίζουν τή μοίρα τους
μέ τήν ἀνάσα τῆς θάλασσας
ὅπως κι ἡ θάλασσα ἀντικρύζεται
μέ φθινοπωρινή ἢ ἀνοιξιάτικη ὄψη, ἀναλόγως
ὅταν κοιτῶ ψηλά
στίς φυλλωσιές μεγάλων δέντρων
τρέχοντας ἀπό κάτω μέ ποδήλατο
ἐνῶ κάποιοι ἄλλοι λογαριάζουν
τά δέντρα δέντρα
καί τίς ἀκτές οἰκόπεδα
πού τόσο κάνει, κύριοι, ἂν θέτε νά χαλάσετε
δικαίωμά σας, ἀφοῦ εἴστε ἐλεύθεροι
ἄρκεϊ προηγουμένως ν' ἀγοράσετε.

Μ. ΠΥΘΕΑΣ