Resonance collisions between three-level systems in an intense laser field

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Theory relevant to the treatment of resonance collisions between two-level dressed atoms has been extended to the three-level case in a self-contained calculation of field-dependent collision rates. With the laser field tuned close to the $|e_1\rangle \leftrightarrow |e_2\rangle$ transition, dressing of the upper two-level system is achieved. We solve coupled equations of motion for two-atom states that describe one atom in the ground state and the other in one of two dressed states. Dressed-state collision rates are calculated within the impact approximation using S-matrix theory. The coupled equations of motion are simplified in the limit of intense field. We have applied the theory to describe approximately collisions between Ne atoms in fields intense enough to dress the collision complex.

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I. INTRODUCTION

Considerable theoretical research has been carried out into the effect of *strong* laser radiation on atomic collisions [1]. In particular, the modification of collision dynamics by a strong laser field [2,3] has been of much interest. The theory used to describe this must consider the coupling of the atom with the field for the case where the effect of the field is too large to be treated using perturbation theory. The "dressed-atom" approach [4,5] treats the atom in the field to all orders of perturbation and provides the most appropriate description of strong-field systems. In this paper we shall use this approach to calculate field-dependent collision rates for the particular case of two neon atoms undergoing pure resonance collisions. The neon atom provides a three-level system. Collisions couple the $2\,^1S_0$ ground state of one atom with the $3\,^1P_1$ "first excited" (resonance) state of the other atom and a strong laser field couples the latter with the $3\,^1S_0$ "second excited" state in each atom. This three-level atom is to relate to an experimental system in which the $|g\rangle \leftrightarrow |e_1\rangle$ transition is in the vacuum ultraviolet and, for rare gases, beyond the scope of available tunable lasers. The laser field is tuned close to the $|e_1\rangle \leftrightarrow |e_2\rangle$ transition. Such a scheme helps us to avoid the effects of stimulated Raman scattering that occur when a ground state is coupled in by a strong field. The field therefore dresses the upper two level system. We solve coupled equations of motion for two-atom states that describe one atom in the ground state and the other in one of two dressed states. The treatment is simplified by using the binary collision [6–8] and secular [9] approximations. Calculation within the impact approximation means that the S-matrix theory can be used to find the dressed state collision rates. The coupled equations of motion are simplified in the limit of intense field.

II. THE THREE-LEVEL ATOM-LASER FIELD COMPOUND SYSTEM

The atoms are assumed to follow classical rectilinear paths and undergo only binary collisions. Spontaneous emission during a collision is ignored and we neglect the effect of degenerate substates.

We are interested in an atom consisting of a ground state, $|g\rangle$ and two, nondegenerate excited states, $|e_1\rangle$ and $|e_2\rangle$ which correspond to the three coupled states shown in Fig. 1.

The (neutral) atoms interact via the dipole-dipole resonance interaction between the ground state of one atom and the first excited state of the other. The leading term in the long-range analytical form of this interatomic potential is

$$\hat{H}_R(a,b)=\frac{1}{r^3}\left[\mathbf{d}_a\cdot\mathbf{d}_b-3\frac{(\mathbf{d}_a\cdot\mathbf{r})(\mathbf{d}_b\cdot\mathbf{r})}{r^2}\right], \tag{1}$$

where a and b denote the atoms with electric dipole moments $\mathbf{d}_a$ and $\mathbf{d}_b$, respectively, and internuclear vector $\mathbf{r}$. To consider the interaction of two dressed atoms it is convenient to use two-atom states. An undressed two-atom wave function $|\Phi(t)\rangle$ can be expanded in a basis of single-atom product eigenstates:

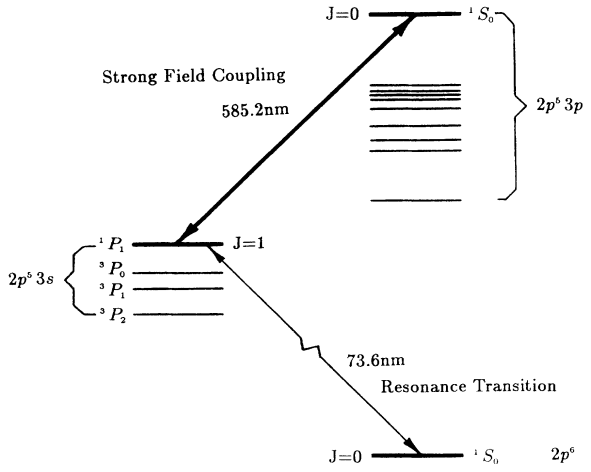

FIG. 1. Lower-energy levels of neon.

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$$|\Phi(t)\rangle = c_1(t)|gg\rangle + c_2(t)|ge_1\rangle + c_3(t)|ge_2\rangle + c_4(t)|e_1g\rangle + c_5(t)|e_1e_1\rangle + c_6(t)|e_1e_2\rangle + c_7(t)|e_2g\rangle + c_8(t)|e_2e_1\rangle + c_9(t)|e_2e_2\rangle \ . \quad (2)$$

The two, free, three-level atom system has a Hamiltonian $\hat{H}_0$ with the following energies:

$$\begin{aligned} E_1 &= \langle gg|\hat{H}_0|gg\rangle = 2\hbar\omega_g \ , \\ E_2 &= \langle ge_1|\hat{H}_0|ge_1\rangle = \langle e_1g|\hat{H}_0|e_1g\rangle = \hbar(\omega_{e_1}+\omega_g) \ , \\ E_3 &= \langle e_1e_1|\hat{H}_0|e_1e_1\rangle = 2\hbar\omega_{e_1} \ , \\ E_4 &= \langle ge_2|\hat{H}_0|ge_2\rangle = \langle e_2g|\hat{H}_0|e_2g\rangle = \hbar(\omega_{e_2}+\omega_g) \ , \\ E_5 &= \langle e_1e_2|\hat{H}_0|e_1e_2\rangle = \langle e_2e_1|\hat{H}_0|e_2e_1\rangle = \hbar(\omega_{e_2}+\omega_{e_1}) \ , \\ E_6 &= \langle e_2e_2|\hat{H}_0|e_2e_2\rangle = 2\hbar\omega_{e_2} \ . \end{aligned} \quad (3)$$

The two-atom system can be dressed by adding these basis states to the field states and allowing the atom-field interaction Hamiltonian, $\hat{V}$, to couple the two systems. $\hat{V}$ couples the upper two states in each atom whereas the resonance interaction Hamiltonian, $\hat{H}_R$ couples the first excited state of one atom with the ground state of the other. Figure 2 illustrates the atom-atom and atom-field interactions.

Not all of the states participate in the collision. Both atoms in the ground state represent a "null" two-atom state since it has no open channels; we need not be concerned with $|gg\rangle$. Similarly there are no matrix elements for resonance interactions between $|e_{1,2}\rangle$ and $|e_{2,1}\rangle$ and the effects of $\hat{V}$ and $\hat{H}_R$ cannot combine to leave both atoms excited. We are able to neglect the $|e_{1,2}e_{1,2}\rangle$ states. The addition of the two atom "bare" states to the laser field results in the system shown in Fig. 3.

The total Hamiltonian for the *undressed* system is

$$\hat{H}_0' = \hat{H}_0 + \hat{H}_L \ , \quad (4)$$

such that

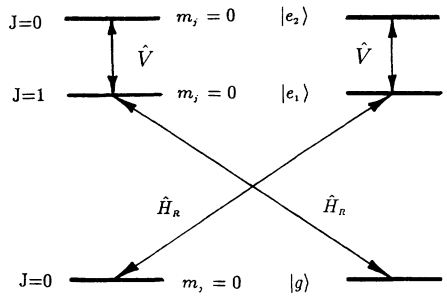

FIG. 2. Schematic of the individual three-level atoms showing the coupling of the laser field and atomic interaction operators.

$$\langle ge_1,n+1|\hat{H}_0'|ge_1,n+1\rangle = E_2 + (n+1)\hbar\omega_L \ , \quad (5)$$

$$\langle ge_2,n|\hat{H}_0'|ge_2,n\rangle = E_4 + n\hbar\omega_L \ . \quad (6)$$

We can write the total Hamiltonian for the *dressed* two-atom system (including atomic interaction) as

$$\hat{H} = \hat{H}_0 + \hat{H}_L + \hat{V} + \hat{H}_R \ , \quad (7)$$

where

$$\hat{V}(t) = -\mathbf{d}\cdot\mathbf{E}(t) \ . \quad (8)$$

$\hat{H}_R$ is defined by Eq. (1) in which r, varies with time through the collision.

The time-dependent wave function $|\Phi(t)\rangle$ is now represented on a basis of only four states and can be reexpressed as

$$|\Phi(t)\rangle = a_1(t)|ge_1,n+1\rangle + a_2(t)|ge_2,n\rangle + a_3(t)|e_1g,n+1\rangle + a_4(t)|e_2g,n\rangle \quad (9)$$

Substitution of the undressed wave function into the time-dependent Schrödinger equation with Hamiltonian defined by Eq. (7) followed by projection onto the undressed basis yields four coupled equations of motion for the amplitudes $a(t)$. Using the rotating wave approximation these equations can be written in matrix form as

$$i\hbar\frac{\partial}{\partial t}\begin{bmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{bmatrix} = \begin{bmatrix} E_2+(n+1)\hbar\omega_L & V & R(t) & 0 \\ V & E_4+n\hbar\omega_L & 0 & 0 \\ R(t) & 0 & E_2+(n+1)\hbar\omega_L & V \\ 0 & 0 & V & E_4+n\hbar\omega_L \end{bmatrix}\begin{bmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{bmatrix} , \quad (10)$$

where V and $R(t)$ represent the matrix elements of the interaction operators $\hat{V}$ and $\hat{H}_R$, respectively. It can be shown that R is given by

$$\pm R = \pm\frac{e^2\hbar}{8\pi\epsilon_0}\frac{f_{ge_1}}{m\omega_{ge_1}}\frac{1}{r^3} = \pm\frac{C_3}{r^3} \ . \quad (11)$$

Given the form of the undressed wave function $|\Phi(t)\rangle$ in Eq. (9), we can write down a 4×4 dressing transformation for the two atom system, thus,

$$\hat{T}_1 = \begin{bmatrix} \cos(\theta/2) & \sin(\theta/2) & 0 & 0 \\ -\sin(\theta/2) & \cos(\theta/2) & 0 & 0 \\ 0 & 0 & \cos(\theta/2) & \sin(\theta/2) \\ 0 & 0 & -\sin(\theta/2) & \cos(\theta/2) \end{bmatrix} , \quad (12)$$

where θ is defined by

$$\tan\theta=\frac{2V}{\hbar(\omega_0-\omega_L)}=\frac{\Omega_1}{\delta} . \tag{13}$$

$\hat{T}_1$ transforms the Hamiltonian and the Schrödinger equation for the dressed two-atom system can be written as

$$i\hbar\frac{\partial|\Psi(t)\rangle}{\partial t}=i\hbar\frac{\partial}{\partial t}\begin{bmatrix}d_1\\d_2\\d_3\\d_4\end{bmatrix}=\hat{T}_1\hat{H}\hat{T}_1^{\dagger}\begin{bmatrix}d_1\\d_2\\d_3\\d_4\end{bmatrix} . \tag{14}$$

Use of Eq. (10) and (12) then yields

$$i\hbar\frac{\partial|\Psi(t)\rangle}{\partial t}=\begin{bmatrix}E_{\rm I}&0&0&0\\0&E_{\rm II}&0&0\\0&0&E_{\rm I}&0\\0&0&0&E_{\rm II}\end{bmatrix}|\Psi(t)\rangle+\begin{bmatrix}0&0&R(t)&0\\0&0&0&0\\R(t)&0&0&0\\0&0&0&0\end{bmatrix}|\Psi(t)\rangle$$
$$+\frac{R(t)}{2\Omega}\begin{bmatrix}0&0&\delta-\Omega&-2V/\hbar\\0&0&-2V/\hbar&\Omega-\delta\\\delta-\Omega&-2V/\hbar&0&0\\-2V/\hbar&\Omega-\delta&0&0\end{bmatrix}|\Psi(t)\rangle , \tag{15}$$

where the dressed two-atom Hamiltonian has been separated into three matrices and

$$E_{\rm I}=\frac{\hbar}{2}(2\omega_g+\omega_{e_1}+\omega_{e_2}+(2n+1)\omega_L+\Omega) , \tag{16}$$

$$E_{\rm II}=\frac{\hbar}{2}(2\omega_g+\omega_{e_1}+\omega_{e_2}+(2n+1)\omega_L-\Omega) , \tag{17}$$

so that

$$E_{\rm I}-E_{\rm II}=\hbar\Omega . \tag{18}$$

To distinguish between the four dressed states they are denoted by $|{\rm I}\rangle$, $|{\rm II}\rangle$, $|{\rm III}\rangle$, and $|{\rm IV}\rangle$; the photon number, n, has been omitted for simplicity. The eigenvalues $E_{\rm I}$ and $E_{\rm II}$ show that the four dressed states form two pairs of degenerate levels.

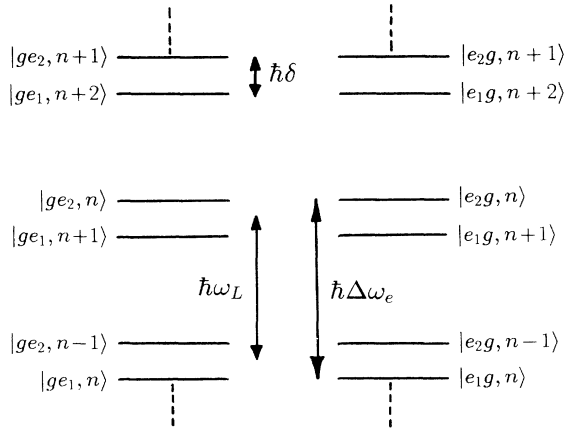

FIG. 3. The undressed states of two, three-level atom–laser field compound system. Three manifolds are shown.

The states $|{\rm I}\rangle$ and $|{\rm II}\rangle$ both correspond to the "first" atom being in the ground state and the "second" atom being in one or other excited states. States $|{\rm III}\rangle$ and $|{\rm IV}\rangle$ correspond to the *second* atom being in the ground state. It is important to remember that this system involves one excited (and hence dressed) atom and one ground (undressed) atom at any instant.

The consequence of the off-diagonal submatrices in the third matrix in Eq. (15) is that the dressed pair $|{\rm I}\rangle$ and $|{\rm II}\rangle$ are coupled to the dressed pair $|{\rm III}\rangle$ and $|{\rm IV}\rangle$ resulting in eight possible transitions. These are shown in Fig. 4 and have rates denoted by Γ_{ij}. We shall now use Eq. (15) to calculate the transition rates between the dressed states.

It is interesting to note that transitions $|{\rm I}\rangle\leftrightarrow|{\rm III}\rangle$ and $|{\rm II}\rangle\leftrightarrow|{\rm IV}\rangle$ correspond to excitation transfer between atoms with no net change in the energy of the two-atom system. The transitions $|{\rm I}\rangle\leftrightarrow|{\rm IV}\rangle$ and $|{\rm II}\leftrightarrow|{\rm III}\rangle$ correspond to collisions in which the two-atom system changes its thermal energy by $\pm\hbar\Omega$. Such collisions are a new phenomenon in the two, three-level atom interaction and are made possible by the intense laser field causing ac Stark shifts of the atomic energy levels.

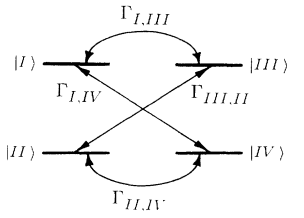

FIG. 4. Transitions induced between the dressed states with rates Γ_{ij}.

III. SOLUTION OF THE COUPLED EQUATIONS OF MOTION

To study the effect of a collision on the state vector, $|\Psi(t)\rangle$, we seek the probability that a system in initial state $|i\rangle$ is left in a final state $|f\rangle$ after the collision. This is given by the square modulus of the collision S-matrix element, S_{if}, coupling the two states. The S matrix is obtained by integrating the equations of motion through the collision. The effect of a single collision is expressed, thus,

$$|\Psi(\infty)\rangle = S(\beta,\theta,v,\delta,V)|\Psi(-\infty)\rangle \ , \tag{19}$$

where the S matrix depends on the collision geometry (impact parameter β, collision angle θ, and relative velocity v) and the laser field (detuning δ and Rabi frequency Ω_1). To obtain the field-dependent excitation transfer rates $\Gamma_{if}(V,\delta)$ we average over all possible collisions and find

$$\Gamma_{if}(V,\delta) = \mathcal{N}\int_{\beta_{\min}}^{\beta_{\max}} 2\pi\beta\, d\beta |S_{if}|^2 \int_{v_{\min}}^{v_{\max}} v f(v) dv \ , \tag{20}$$

where $\mathcal{N}$ is the number density. Note that for the two-level system coupled by the linearly polarized laser we can ignore the average over collision angles θ.

For an intense laser field $V \gg \delta$ so that $\Omega \to 2V/\hbar$. Equation (15) can then be rewritten as

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle = \begin{bmatrix} E_{\rm I} & 0 & R(t)/2 & -R(t)/2 \\ 0 & E_{\rm II} & -R(t)/2 & R(t)/2 \\ R(t)/2 & -R(t)/2 & E_{\rm I} & 0 \\ -R(t)/2 & R(t)/2 & 0 & E_{\rm II} \end{bmatrix} |\Psi(t)\rangle \ . \tag{21}$$

We further note that for large field strengths the difference $E_{\rm I} - E_{\rm II} = \Omega$ becomes large. It should therefore be valid to treat the terms coupling the states with this separation using perturbation theory. To do this, however, we first have to take the coupling between degenerate states to all orders. The coupling between the degenerate pairs is described by

$$i\hbar\frac{\partial}{\partial t}|\Psi_0(t)\rangle = \hat{H}_0|\Psi_0(t)\rangle \ , \tag{22}$$

where

$$\hat{H}_0 = \begin{bmatrix} E_{\rm I} & 0 & R(t)/2 & 0 \\ 0 & E_{\rm II} & 0 & R(t)/2 \\ R(t)/2 & 0 & E_{\rm I} & 0 \\ 0 & R(t)/2 & 0 & E_{\rm II} \end{bmatrix} . \tag{23}$$

The two pairs of coupled equations of motion are easily solved to yield time-dependent amplitudes for states in a new basis. This basis is defined by the eigenvectors of the Hamiltonian in Eq. (22) so that in this basis the Hamiltonian is diagonal. We find

$$(d_1 \pm d_3)(t) = D_{\rm I\pm}(t) = D_{\rm I\pm}(0)\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t} E_{\rm I} \pm R(t')/2\, dt'\right] , \tag{24}$$

$$(d_2 \pm d_4)(t) = D_{\rm II\pm}(t) = D_{\rm II\pm}(0)\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t} E_{\rm II} \pm R(t')/2\, dt'\right] . \tag{25}$$

The new basis vectors are denoted by $|\xi_k\rangle$, where $k = {\rm I}+, {\rm II}+, {\rm I}-, {\rm II}-$. The unperturbed wave function is written as

$$\begin{aligned} |\Xi_0(t)\rangle &= D_{\rm I+}(t)|\xi_{\rm I+}\rangle + D_{\rm II+}(t)|\xi_{\rm II+}\rangle + D_{\rm I-}(t)|\xi_{\rm I-}\rangle + D_{\rm II-}(t)|\xi_{\rm II-}\rangle \\ &= \sum_k D_k(0)\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t} E_k(t')dt'\right]|\xi_k\rangle \ . \end{aligned} \tag{26}$$

The transformation effected by diagonalizing the matrix in Eq. (22) can be written as

$$\hat{T}_2 = \frac{1}{\sqrt{2}}\begin{bmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 \\ 1 & 0 & -1 & 0 \\ 0 & 1 & 0 & -1 \end{bmatrix} . \tag{27}$$

It is easy to verify that

$$\hat{T}_2\hat{H}_0\hat{T}_2^{\dagger}=\begin{bmatrix} E_{\rm I}+R(t)/2 & 0 & 0 & 0 \\ 0 & E_{\rm II}+R(t)/2 & 0 & 0 \\ 0 & 0 & E_{\rm I}-R(t)/2 & 0 \\ 0 & 0 & 0 & E_{\rm II}-R(t)/2 \end{bmatrix} . \tag{28}$$

Perturbation theory is now used to include the effect of the collision couplings initially neglected. The matrix elements excluded from Eq. (22) constitute the perturbation matrix $\hat{H}'$. Use of $\hat{T}_2$ transforms the perturbation into the basis of $|\xi_k\rangle$:

$$\hat{T}_2\begin{bmatrix} 0 & 0 & 0 & -R(t)/2 \\ 0 & 0 & -R(t)/2 & 0 \\ 0 & -R(t)/2 & 0 & 0 \\ -R(t)/2 & 0 & 0 & 0 \end{bmatrix}\hat{T}_2^{\dagger}=\begin{bmatrix} 0 & -R(t)/2 & 0 & 0 \\ -R(t)/2 & 0 & 0 & 0 \\ 0 & 0 & 0 & R(t)/2 \\ 0 & 0 & R(t)/2 & 0 \end{bmatrix} . \tag{29}$$

The couplings represented by the elements of $\hat{H}'$ in each basis are shown in Fig. 5.

Following Eq. (26), the general solution of the perturbed Schrödinger equation,

$$i\hbar\frac{\partial}{\partial t}|\Xi(t)\rangle=(\hat{H}_0+\hat{H}')|\Xi(t)\rangle , \tag{30}$$

can be written as

$$|\Xi(t)\rangle=\sum_k D_k(t)\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t}E_k(t')dt'\right]|\xi_k\rangle . \tag{31}$$

The rate of change of the probability amplitude to first order for a collisional transition from $|\xi_a\rangle$ to $|\xi_b\rangle$ is given by

$$\frac{\partial}{\partial t}D_b^{(1)}(t)=-\frac{i}{\hbar}\langle\xi_b|\hat{H}'|\xi_a\rangle \times\exp\left[\frac{i}{\hbar}\int_{-\infty}^{t}E_b(t')-E_a(t')dt'\right] . \tag{32}$$

From the form of the matrix $\hat{H}'$ we see that the matrix element in Eq. (32) can take four values corresponding to the collisional couplings in Fig. 5(b). The four time-dependent amplitudes $D_b^{(1)}(t)$ are found by integrating Eq. (32) over all time. We find that the energy difference between $|\xi_a\rangle$ and $|\xi_b\rangle$ is always constant at $\pm\hbar\Omega$ as a consequence of the perturbation and Eq. (18). The simplified transition amplitudes are given by

$$D_{\rm I\pm}^{(1)}(t)=\pm\frac{i}{2\hbar}\int_{-\infty}^{\infty}R(t)e^{i\Omega t}dt , \tag{33}$$

$$D_{\rm II\pm}^{(1)}(t)=\pm\frac{i}{2\hbar}\int_{-\infty}^{\infty}R(t)e^{-\Omega t}dt , \tag{34}$$

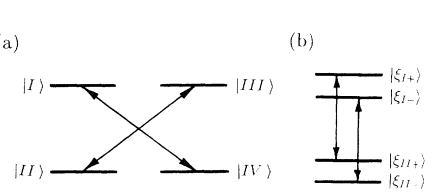

FIG. 5. Collisional coupling between Stark-shifted states. For a large shift the couplings are weak and can be applied as a perturbation. (a) Dressed-state basis $|i\rangle$. (b) Stationary unperturbed basis $|\xi_k\rangle$.

where the lower limit of the integral over t' has been effectively set to zero since it will have no effect when the square modulus of the amplitude is calculated. The S-matrix elements are thus given by the Fourier transforms of the dressed atomic interaction matrix elements. The Fourier components of $R(t)$ represent the frequencies of (dressed state) transitions that can be induced by the collision; the harder (i.e., shorter) the collision, the greater the energy separation it can overcome.

We make the approximation of a straight-line classical path in which the perturber moves past the radiating atom on a classical trajectory. The atom is considered to be perturbed during the collision only. r is a function of t and, for impact parameter β and mean relative velocity v, is given by $r=(\beta^2+\overline{v}^2t^2)^{1/2}$. Equation (11) shows that $R(t)$ is proportional to r^{-3} with a constant, C_3. Inserting this into Eqs. (33) and (34) we find that the S-matrix elements are given by the following Fourier transforms:

$$D_{\rm I\pm}^{(1)}(t)=\pm\frac{iC_3}{2\hbar}\int_{-\infty}^{\infty}\frac{e^{i\Omega t}}{(\beta^2+\overline{v}^2t^2)^{3/2}}dt , \tag{35}$$

$$D_{\rm II\pm}^{(1)}(t)=\pm\frac{iC_3}{2\hbar}\int_{-\infty}^{\infty}\frac{e^{-i\Omega t}}{(\beta^2+\overline{v}^2t^2)^{3/2}}dt . \tag{36}$$

The Fourier transform of the even function, $R(t)$ was taken to be twice the Fourier cosine transform of $R(t)$. The cosine transform was taken from Ref. [10] and the S-matrix elements $S_{\rm if}$ are found to contain modified Bessel functions of first order:

$$D_k^{(1)}(\Omega,\beta,v)=S_{\rm if}=\pm\left[\frac{2iC_3}{\hbar}\right]\frac{\Omega}{v^2\beta}K_1(\Omega\beta/v) , \tag{37}$$

where $k={\rm I,II}\pm$. To obtain the collision rates $\Gamma_{\rm if}$ we average $|S_{\rm if}|^2$ over all possible collisions according to Eq. (20).

Shah *et al.* [11] have carried out a closely related calculation for the resonance collision of two dressed mercury atoms where the intense field drives the resonance transition. To establish the qualitative effect of laser radiation on the resonance interaction the field-dependent excitation transfer rates were calculated for the $6\,^1S_0\leftrightarrow 6^3P_1$ transition in mercury. To see if it was necessary to perform an average over relative velocity, Shah *et al.* compared [11]

$$\int_{v_{\min}}^{v_{\max}} v f(v) |S_{if}|^2 dv$$

with $\bar{v}|S_{if}|^2$ over a range of temperatures from 100 to 1500 K. It was found that the two expressions were the same to within 3% and $\bar{v}$ was used for a given temperature. It was also shown that the depopulation rate of a dressed state was negligible for $\beta > 10$ nm thus setting $\beta_{\max}$. Furthermore, for $\beta > 0.3$ nm rapid oscillations in the depopulation rate suggested a strong collision "cutoff" at this impact parameter. Contributions to the rate from collisions for which $\beta < 0.3$ nm were found to be less than 0.25% of the final values.

Assuming a mean velocity will work here we can rewrite Eq. (20) as

$$\Gamma_k(\Omega) = 2\pi \mathcal{N} \left[\frac{4C_3^2}{\hbar^2} \right] \frac{\Omega^2}{\bar{v}^3} \int_{\beta_{\min}}^{\beta_{\max}} \frac{|K_1(\Omega\beta/\bar{v})|^2}{\beta} d\beta , \quad (38)$$

where we have used Eq. (37) and C_3 is given by Eq. (11).

Numerical Algorithms Group (NAG) Fortran routine S18ADF was used to generate the modified Bessel function as a function of Ω for given β and $\bar{v}$. The function was also generated as a function of β for $0.3 \leq \beta \leq 10$ nm at a given Ω and $\bar{v}$ to check continuity of the integrand in Eq. (38). NAG Fortran routine D01AHF was used to perform the integration over the impact parameter at a mean relative velocity $\bar{v}$. Our intention was to plot Γ_k as a function of Ω. The above analysis is valid in the limit that the perturbation $\hat{H}'$ is small, i.e., the Stark shift, $\hbar\Omega$ is large. The minimum impact parameter for which the analysis remains valid, $\beta_{\min}$, was found by calculating the transition probability $|S_{if}|^2$ as a function of β for various values of Ω and $\bar{v} = 589$ ms^{-1} ($\equiv 75$°C). The dressed-state depopulation rate should oscillate rapidly for $\beta < \beta_{\min}$. However, in this simple treatment the rate simply approaches infinity asymptotically as the impact parameter approaches zero ($|S_{if}|^2 \to \infty$ as $\beta \to 0$). Clearly the perturbation is breaking down at the impact parameter for which $|S_{if}|^2 > 1$. We assumed that the rapid oscillations in the transition probability between 0 and 1 would give a mean probability of $\frac{1}{2}$. We were thus able to deduce the value of the impact parameter, $\beta_{\min}$, (for a given value of Ω) for which $|S_{if}|^2 = \frac{1}{2}$. Values of Ω between 10 and 300 cm^{-1} were taken with $\bar{v} = 589$ ms^{-1} and the corresponding transition probabilities yielded by the above analysis are shown as a function of β in Fig. 6.

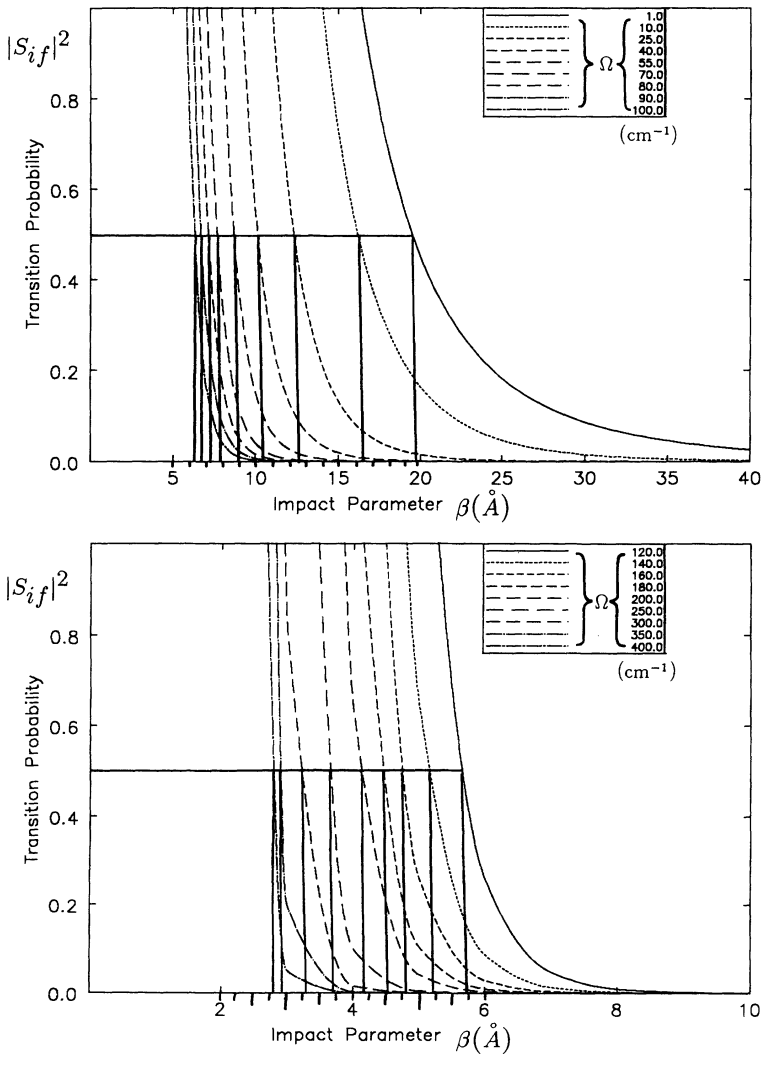

FIG. 6. Transition probability to state $|\xi_k\rangle$ as a function of impact parameter at $\bar{v} = 589$ ms^{-1} for various Ω.

The values of $\beta_{\min}$ yielded by Fig. 6 were then fed into Eq. (38) to calculate the rates of excitation transfer between two neon atoms at 75°C for various values of the generalized Rabi frequency.

IV. RESULTS AND CONCLUSIONS

With $\mathcal{N}$ taken as 10^{16} cm^{-3} and $4C_3^2/\hbar^2$ calculated to be 2.57×10^{-3} m^3 s^{-1} rad^{-1} ($f_{ge}=0.162$ for the λ-73.6-nm transition) the excitation transfer rates for the states $|\xi_k\rangle$ are shown in Fig. 7.

How would the effect of such collisions be observed? In an experiment we would first produce population in the first excited resonance levels using a discharge. We would then turn on an intense filed and observe the amount of collision-induced fluorescence from the upper level which would then be dressed. The amount of fluorescence would be determined by the rate of collisions that cause transitions between the dressed two sets of degenerate states $|\mathrm{II}\rangle$, $|\mathrm{IV}\rangle$, and $|\mathrm{I}\rangle$, $|\mathrm{III}\rangle$. Collisional transitions between $|\mathrm{I}\rangle$ and $|\mathrm{III}\rangle$ or $|\mathrm{II}\rangle$ and $|\mathrm{IV}\rangle$ will have no effect on the light intensity emitted (they do broaden the lines but that is much more difficult to observe). In terms of observables, then, all we are interested in is the field-dependent transition rate, Γ_{12} ($=\Gamma_{21}$). It is clear that

$$\Gamma_{12}=\Gamma_{\mathrm{I,IV}}+\Gamma_{\mathrm{III,II}}\propto\Gamma_{\mathrm{I+II+}}+\Gamma_{\mathrm{I-II-}}\ . \tag{39}$$

We have calculated $\Gamma_{\mathrm{I\pm II\pm}}$ ($\equiv\Gamma_k$). To relate these to the above rates we can return to the S-matrix elements and project $|\xi_k\rangle$ onto $|i\rangle$. We write

$$S_{\mathrm{I,IV}}=\langle \mathrm{I}|\xi_{\mathrm{I+}}\rangle\langle\xi_{\mathrm{I+}}|\hat{S}|\xi_{\mathrm{II+}}\rangle\langle\xi_{\mathrm{II+}}|\mathrm{IV}\rangle\ , \tag{40}$$

$$S_{\mathrm{III,II}}=\langle \mathrm{III}|\xi_{\mathrm{I-}}\rangle\langle\xi_{\mathrm{I-}}|\hat{S}|\xi_{\mathrm{II-}}\rangle\langle\xi_{\mathrm{II-}}|\mathrm{II}\rangle\ . \tag{41}$$

It then follows that

$$\Gamma_{12}\propto|\langle \mathrm{I}|\xi_{\mathrm{I+}}\rangle S_{\mathrm{I+II+}}\langle\xi_{\mathrm{II+}}|\mathrm{IV}\rangle|^2 + |\langle \mathrm{III}|\xi_{\mathrm{I-}}\rangle S_{\mathrm{I-II-}}\langle\xi_{\mathrm{II-}}|\mathrm{II}\rangle|^2\ . \tag{42}$$

The scalar products each yield a factor of $1/\sqrt{2}$ and we find that

$$\Gamma_{12}=\tfrac{1}{4}(\Gamma_{\mathrm{I+II+}}+\Gamma_{\mathrm{I-II-}})=\tfrac{1}{2}\Gamma_{\mathrm{if}}\ . \tag{43}$$

The rates displayed in Fig. 7 are thus easily converted into rates that could be observed experimentally in the fluorescence intensities. The actual intensities in a specific experiment will also depend on details of the temporal an spatial profiles of the laser pulse. The rates we have calculated can be used in the modeling of an experiment based on such specific knowledge.

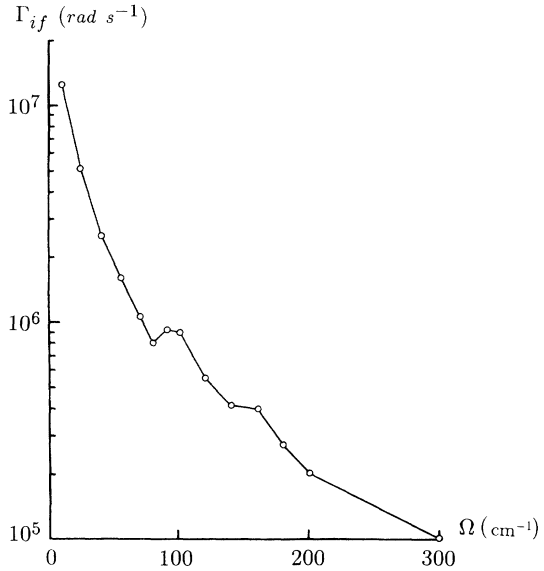

FIG. 7. Excitation transfer rates for states $|\xi_k\rangle$ as a function of Ω at $\bar{v}=589$ ms^{-1} and $\mathcal{N}=10^{16}$ cm^{-3}.

The strong-field-dependent collision rates can now be compared with the field-free rate, Γ^C. Measurements of the pressure width of the λ-585.2-nm line in self-broadened neon have been made by, for example, Stacey and Thompson [12]. The lower level in this transition is predominantly resonance broadened by the 73.6-nm transition down to the ground state. Stacey and Thompson report a pressure width of 1.6×1.0^9 rad s^{-1} at 1 Torr. (N.B. natural width $=0.719\times10^8$ rad s^{-1}.) Even at the lowest Rabi frequency considered by the calculation (10 cm^{-1}) the collision rate has fallen to just over 10^7 rad s^{-1}.

This analysis has shown why and by how much the excitation transfer rates decrease when the intensity of the laser field reaches the regime of $\Omega\tau_c\approx1$. Although we have chosen the case of Neon the analysis given here can be extended quite straightforwardly to other systems. At present two-laser experiments are underway in mercury that will exploit the advantages offered by the three level scheme described here.

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