

Collective atomic effects in resonance fluorescence: Dipole-dipole interaction

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The resonance fluorescence of a two-atom system interacting collectively with an intense driving field is studied. The first-order dispersion forces, or dipole-dipole interaction, between the atoms is included as well. The scattered-light spectrum is found to differ considerably from the single-atom spectrum even for very intense driving fields, in direct contrast with the results obtained by Agarwal *et al.*, who studied the two-atom system but neglected the dipole-dipole interaction. The effects of collective scattering are calculated on the fluorescence spectra of idealized beams of atoms moving with uniform velocity, for a number of different number densities $\langle N \rangle$.

I. INTRODUCTION

In recent months, interest has arisen in multi-atom effects in resonance fluorescence. Agarwal *et al.*¹ have calculated the fluorescence spectrum and the intensity-correlation function for systems of two and three identical atoms interacting collectively with the radiation field, Kimble *et al.*² have studied the effects on the intensity-correlation function of fluctuations in the number of radiating atoms and of their finite transit time in the field of view of the photodetector(s), and Mollow³ has studied the effects on the fluorescence spectrum of collisions with foreign gas atoms. In this article we consider again the case of collective atomic scattering, which becomes important when high number densities $\langle N \rangle$ occur in the atomic beam. We study the effect on the scattering of a phenomenon which has hitherto been neglected, namely, the first-order dispersion forces or the dipole-dipole interaction. We find that the inclusion of this interaction changes the spectrum of the two-atom system considerably from that of the single-atom system, even for intense driving fields,⁴ and, for an idealized atomic beam consisting of a random distribution of atoms moving with uniform velocity, we calculate the net effect of collective scattering on the fluorescence spectrum for several densities $\langle N \rangle$. The integrated scattered intensity, on the other hand, is found to be independent of the dipole-dipole interaction, so that the latter's inclusion does not alter the results obtained by Agarwal *et al.*¹ for the intensity-correlation function of the collective two-atom system.

We neglect in this article the fluctuation and transit-time corrections of Kimble *et al.*,² and concentrate solely on the effects of collective scattering. We reduce the problem to its bare

essentials by considering two, two-level atoms,⁵ separated by a distance R , under the influence of an external driving field of frequency ω_0 , at resonance with the atomic transition. It is easily verified that the separations R for which the dipole-dipole interaction is appreciable are $R \lesssim c/\omega_0$ [see Eq. (4)], so that the dipole approximation or small sample model is used for the interaction of the field with the atomic system. Finally, we neglect single-atom radiative shifts, and the contribution to the spectrum of the transient response of the system compared to that of the steady-state response,⁶ and confine our attention to intense fields only. We are thus able to study transitions of the system using the dressed-atom description and master-equation approach of Cohen-Tannoudji and Reynaud⁷ (to be referred to as CR) and obtain an analytical expression for the fluorescence spectrum.⁸

It should be noted that the collective scattering problem studied here is completely equivalent to a study of the effects on the fluorescence spectrum of resonance broadening⁹ by collisions of the radiating atom with other atoms from the beam. The idealization to uniform atomic beam velocities corresponds to making the quasistatic approximation in pressure-broadening theory.⁹ To study a real beam, whose atoms travel with nonuniform velocities, more sophisticated applications of pressure-broadening theory are required.

In Sec. II, we introduce our model and calculate the energy levels and eigenstates of the dressed two-atom system. In Sec. III, we study the evolution of the density matrix elements and in Sec. IV obtain an expression for the fluorescence spectrum as a function of the dipole-dipole interaction. Finally, in Sec. V, we present curves which illustrate the effects of collective atomic scattering on the fluorescence spectra of atomic beams having different number densities $\langle N \rangle$.

II. DRESSED TWO-ATOM SYSTEM

We consider two atoms, $a=1, 2$, having lower levels $|g_a\rangle$ and upper levels $|e_a\rangle$ separated by energy $\hbar\omega_0$, driven by an intense resonant single-mode laser field in the state $|n\rangle$. In the absence of any coupling between atoms and field, the energy level $E_n = n\hbar\omega_0$ is fourfold degenerate, corresponding to the zero-order states $|g_1g_2n\rangle$, $|e_1g_2n-1\rangle$, $|g_1e_2n-1\rangle$, and $|e_1e_2n-2\rangle$ of the two-atom-plus-field system. In the small sample model, the interaction Hamiltonian \mathcal{H}^1 between the atoms and the driving field has nonvanishing elements

$$\begin{aligned} \langle g_1g_2n|\mathcal{H}^1|e_1g_2n-1\rangle &= \langle g_1g_2n|\mathcal{H}^1|g_1e_2n-1\rangle \\ &= \langle e_1g_2n-1|\mathcal{H}^1|e_1e_2n-2\rangle \\ &= \langle g_1e_2n-1|\mathcal{H}^1|e_1e_2n-2\rangle \\ &= i(2\pi n\omega_0\hbar/V)^{1/2}\hat{e}\cdot\vec{d}_{ge} = i\hbar\epsilon, \quad (1) \end{aligned}$$

connecting the zero-order states, where \hat{e} is the polarization vector of the laser, \vec{d}_{ge} is the dipole matrix element $\vec{d}_{ge} = \langle g_a|\vec{D}_a|e_a\rangle$, V is the quantization volume, and 2ϵ is the Rabi nutation frequency. The dipole-dipole interaction U between the atoms has nonvanishing elements

$$\begin{aligned} \langle e_1g_2n-1|U|g_1e_2n-1\rangle &= |\vec{d}_{ge}|^2(1-3\cos^2\theta)/R^3, \\ &\equiv \alpha\hbar\epsilon \quad (2) \end{aligned}$$

connecting the zero-order states, where θ is the angle between \vec{d}_{ge} and \vec{R} , and the dimensionless coefficient α measures the relative strengths of the atom-atom and atom-driving-field interactions. The dipole-dipole matrix elements are conveniently rewritten in terms of γ , half the Einstein A coefficient for a single atom,

$$2\gamma = A = 4|\vec{d}_{ge}|^2\omega_0^3/3\hbar c^3, \quad (3)$$

$$\alpha\hbar\epsilon = \frac{3}{2}(1-3\cos^2\theta)(c/\omega_0 R)^3\hbar\gamma. \quad (4)$$

We assume the driving field to be sufficiently intense that $\epsilon \gg \gamma$, and that the variation of ϵ with n may be neglected.

To obtain the energy levels and eigenstates of the dressed two-atom system, we diagonalize the 4×4 matrix

$$\hbar \begin{pmatrix} n\omega_0 & i\epsilon & i\epsilon & 0 \\ -i\epsilon & n\omega_0 & \alpha\epsilon & i\epsilon \\ -i\epsilon & \alpha\epsilon & n\omega_0 & i\epsilon \\ 0 & -i\epsilon & -i\epsilon & n\omega_0 \end{pmatrix}. \quad (5)$$

The resulting energy levels and eigenstates are the following:

$$E_{1n} = n\hbar\omega_0, \quad |1n\rangle = 2^{-1/2}(|g_1g_2n\rangle + |e_1e_2n-2\rangle),$$

$$E_{2n} = n\hbar\omega_0 + (\frac{1}{2}\alpha + \beta)\hbar\epsilon,$$

where

$$\beta^2 = 4 + \frac{1}{4}\alpha^2,$$

$$\begin{aligned} |2n\rangle &= (\beta^2 + \frac{1}{2}\alpha\beta)^{-1/2} [|g_1g_2n\rangle - \frac{1}{2}i(\frac{1}{2}\alpha + \beta) \\ &\quad \times (|e_1g_2n-1\rangle + |g_1e_2n-1\rangle) \\ &\quad - |e_1e_2n-2\rangle], \end{aligned}$$

$$E_{3n} = n\hbar\omega_0 + (\frac{1}{2}\alpha - \beta)\hbar\epsilon,$$

$$\begin{aligned} |3n\rangle &= (\beta^2 - \frac{1}{2}\alpha\beta)^{-1/2} [|g_1g_2n\rangle - \frac{1}{2}i(\frac{1}{2}\alpha - \beta) \\ &\quad \times (|e_1g_2n-1\rangle + |g_1e_2n-1\rangle) \\ &\quad - |e_1e_2n-2\rangle], \end{aligned}$$

$$E_{4n} = n\hbar\omega_0 - \alpha\hbar\epsilon,$$

$$|4n\rangle = 2^{-1/2}(|e_1g_2n-1\rangle - |g_1e_2n-1\rangle). \quad (6)$$

For all values of α , in the small sample model, level $|4n\rangle$ is optically inactive. In the limit of $\alpha \rightarrow 0$, it is easily verified that the energy levels can be divided into an equally spaced triplet,

$$E_{1n} = n\hbar\omega_0, \quad |1n\rangle = 2^{-1/2}(|g_1g_2n\rangle + |e_1e_2n-2\rangle),$$

$$E_{2n} = n\hbar\omega_0 + 2\hbar\epsilon,$$

$$\begin{aligned} |2n\rangle &= \frac{1}{2}(|g_1g_2n\rangle - i|e_1g_2n-1\rangle \\ &\quad - i|g_1e_2n-1\rangle - |e_1e_2n-2\rangle), \end{aligned}$$

$$E_{3n} = n\hbar\omega_0 - 2\hbar\epsilon,$$

$$\begin{aligned} |3n\rangle &= \frac{1}{2}(|g_1g_2n\rangle + i|e_1g_2n-1\rangle \\ &\quad + i|g_1e_2n-1\rangle - |e_1e_2n-2\rangle), \quad (7) \end{aligned}$$

and a singlet

$$E_{4n} = n\hbar\omega_0, \quad |4n\rangle = 2^{-1/2}(|e_1g_2n-1\rangle - |g_1e_2n-1\rangle). \quad (8)$$

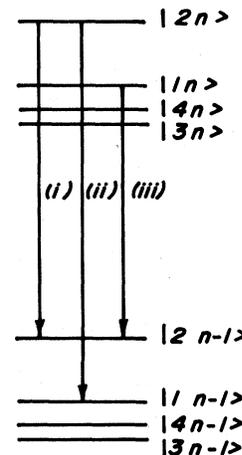


FIG. 1. Schematic energy-level diagram showing the n and $n-1$ multiplets of the dressed two-atom system for $\alpha=1$, together with some of the possible downward spontaneous transitions. Transition (i) contributes to the central frequency component of the spectrum. Transitions (ii) and (iii) correspond to the sidebands at $\omega_0 \pm (\frac{1}{2}\alpha + \beta)\epsilon$.

The energy-level diagram for two adjacent multiplets is represented schematically in Fig. 1 for $\alpha=1$. Considering only this diagram, it is apparent that the possibility of fluorescence exists at 13 different frequencies, $\omega_{ij} \equiv \hbar^{-1}(E_{jn} - E_{i, n-1})$. The optical inactivity of level $|4n\rangle$ reduces this number to 7 for $\alpha \neq 0$; when $\alpha=0$, the lines at frequencies ω_{23} and ω_{32} disappear and the pair at frequencies ω_{21} and ω_{13} merge into a single line, as do the pair at frequencies ω_{12} and ω_{31} , producing a spectrum identical with the familiar single-atom spectrum. These results will be derived systematically in Secs. III and IV.

III. EVOLUTION OF THE DENSITY MATRIX ELEMENTS

The average time behavior of the system coupled now to the remaining modes of the electromagnetic field is determined by the equations of evolution of the density matrix $\sigma(t)$, denoted by

$$\langle in|\sigma(t)|jn-p\rangle = \sigma_{ij}^p(t). \quad (9)$$

To these can be related the evolution of the mean raising and lowering dipole-moment operators

$$\begin{aligned} \bar{D}^\dagger &= \sum_{ij} \bar{D}_{ij}^\dagger = \sum_{ij} d_{ij}^\dagger |in\rangle \langle jn \mp 1|, \\ d_{ij}^\dagger &= \langle in| \sum_{a=1}^2 \bar{D}_a |jn \mp 1\rangle, \end{aligned} \quad (10)$$

and of their mean two-time correlation functions, from which the spectrum is determined.

As in CR,⁷ we simplify the discussion by making the secular approximation: We restrict our calculations to driving fields sufficiently high that $\epsilon \gg \gamma$, allowing us to neglect in the master equation coupling between the diagonal elements of σ (populations) and the off-diagonal elements. The equations for the diagonal elements can then be written in terms of the spontaneous rates of transition Γ_{ij} from $|jn\rangle$ to $|in-1\rangle$,

$$\Gamma_{ij} = |d_{ij}^-|^2 \gamma / 2 |d_{ge}^-|^2, \quad (11)$$

and the total spontaneous rates of transition Γ_j from $|jn\rangle$,

$$\Gamma_j = \sum_i \Gamma_{ij}; \quad (12)$$

$$\dot{\sigma}_{iin}^0(t) = -\Gamma_i \sigma_{iin}^0(t) + \sum_{j=1}^4 \Gamma_{ij} \sigma_{jj, n+1}^0(t). \quad (13)$$

The singlet level $|4n\rangle$ is optically inactive, since $\Gamma_{i4} = \Gamma_{4i} = 0$, and its population remains constant—zero if both atoms are in the ground state on entering the field. The equations of evolution of the triplet populations are solved by the method

of CR.⁷ For both atoms in the ground state on entering the field, the populations at time t are given by the expressions

$$\begin{aligned} \begin{bmatrix} \rho_{11}^0(t) \\ \rho_{22}^0(t) \\ \rho_{33}^0(t) \end{bmatrix} &= \begin{bmatrix} \sum_n \sigma_{11n}^0(t) \\ \sum_n \sigma_{22n}^0(t) \\ \sum_n \sigma_{33n}^0(t) \end{bmatrix} \\ &= \frac{1}{3} \Pi_1 + \frac{4 - \alpha^2 - 2S}{24S(4 + \alpha^2)} \Pi_2 \exp[-\gamma t(8 + \alpha^2 + S)/\beta^2] \\ &\quad + \frac{4 - \alpha^2 + 2S}{24S(4 + \alpha^2)} \Pi_3 \exp[-\gamma t(8 + \alpha^2 - S)/\beta^2], \end{aligned} \quad (14)$$

where $\{\Pi_i\}$ are the vectors

$$\Pi_1 = \begin{bmatrix} 1 \\ 1 \\ 1 \end{bmatrix}, \quad \Pi_{2,3} = \begin{bmatrix} -\frac{64 + 12\alpha^2 \pm 16S \pm 2\alpha^2 S + \alpha^4}{\pm 2S + \alpha^2} \\ 4 \pm S + (8 + \alpha^2 \pm S)\alpha/2\beta \\ 4 \pm S - (8 + \alpha^2 \pm S)\alpha/2\beta \end{bmatrix}, \quad (15)$$

and $S^2 = 16 + \alpha^2 + \frac{1}{4}\alpha^4$. Thus, in the steady state, the populations of the triplet sublevels are equal, and, since $\Gamma_{ij} = \Gamma_{ji}$, satisfy the detailed balance conditions¹⁰

$$\Gamma_{ij} \rho_{jj}^0(t) = \Gamma_{ji} \rho_{ii}^0(t). \quad (16)$$

We next consider the evolution of the off-diagonal elements $\sigma_{iin}^\dagger(t) \equiv \sigma_{iin}^\dagger(t)$, $i=1-3$ [$\sigma_{44n}^\dagger(t) = 0$ due to the inactivity of level $|4n\rangle$], which are related to the central component of the fluorescence spectrum. These elements are coupled to the matrix elements $\sigma_{jj, n+1}^\dagger(t)$ which have the same Bohr frequency ω_0 as $\sigma_{iin}^\dagger(t)$, and are easily verified to obey the equations

$$\dot{\sigma}_{iin}^\dagger(t) = -(i\omega_0 + \Gamma_i) \sigma_{iin}^\dagger(t) + \sum_{j=1}^3 \Gamma_{ij} \sigma_{jj, n+1}^\dagger(t). \quad (17)$$

The solutions to Eqs. (17) are readily found by the method of CR⁷ to be

$$\begin{aligned} \rho_{ii}^\dagger(t) &= \sum_n \sigma_{iin}^\dagger(t) \\ &= \exp(-i\omega_0 t) \left[a_1 \Pi_{1i} + a_2 \Pi_{2i} \exp\left(\frac{-\gamma t(8 + \alpha^2 + S)}{\beta^2}\right) \right. \\ &\quad \left. + a_3 \Pi_{3i} \exp\left(\frac{-\gamma t(8 + \alpha^2 - S)}{\beta^2}\right) \right], \end{aligned} \quad (18)$$

where the constants $\{a_i\}$ can be obtained from

initial conditions, and the vectors $\{\Pi_i\}$ are defined by Eqs. (15). We do not, however, require the values of $\{a_i\}$ in order to calculate the fluorescence spectrum, and therefore do not solve for them.

The lateral components of the fluorescence spectrum are related to the matrix elements $\sigma_{ij}^+(t) \equiv \sigma_{ij}^+(t)$, $i \neq j$. To study their evolution using the simple method of CR,⁷ we must distinguish between four ranges of α : $\alpha = 0$; $\gamma/\epsilon \lesssim |\alpha| \lesssim \frac{2}{3}\epsilon/\gamma$; $0 < |\alpha| \lesssim \gamma/\epsilon$; and $|\alpha| \gtrsim \frac{2}{3}\epsilon/\gamma$.

A. $\alpha = 0$

In the absence of the dipole-dipole interaction, it is easily verified that $\Gamma_{23} = \Gamma_{32} = 0$, while the matrix elements $\sigma_{21n}^+(\sigma_{12n}^+)$ and $\sigma_{13n}^+(\sigma_{31n}^+)$ have the same Bohr frequency of $\omega_0 - 2\epsilon(\omega_0 + 2\epsilon)$, and therefore have coupled equations of evolution. These are readily shown to have the form

$$\begin{aligned} \dot{\sigma}_{21n}^+(\sigma_{12n}^+)(t) &= -[i(\omega_0 \mp 2\epsilon) + \frac{1}{2}(\Gamma_1 + \Gamma_2)]\sigma_{21n}^+(\sigma_{12n}^+)(t) \\ &\quad + (\gamma/2|\vec{d}_{ee}|^2)[d_{22}^-(11)d_{11}^+(22)\sigma_{21n+1}^+(\sigma_{12n+1}^+)(t) \\ &\quad \quad + d_{21}^-(13)d_{13}^+(12)\sigma_{13n+1}^+(\sigma_{31n+1}^+)(t)], \\ \dot{\sigma}_{13n}^+(\sigma_{31n}^+)(t) &= -[i(\omega_0 \mp 2\epsilon) + \frac{1}{2}(\Gamma_1 + \Gamma_3)]\sigma_{13n}^+(\sigma_{31n}^+)(t) \\ &\quad + (\gamma/2|\vec{d}_{ee}|^2)[d_{12}^-(31)d_{13}^+(21)\sigma_{21n+1}^+(\sigma_{12n+1}^+)(t) \\ &\quad \quad + d_{11}^-(33)d_{33}^+(11)\sigma_{13n+1}^+(\sigma_{31n+1}^+)(t)], \end{aligned} \quad (19)$$

with the solutions

$$\begin{aligned} \begin{pmatrix} \rho_{21}^+(\sigma_{12}^+)(t) \\ \rho_{13}^+(\sigma_{31}^+)(t) \end{pmatrix} &= \begin{pmatrix} \sum_n \sigma_{21n}^+(\sigma_{12n}^+)(t) \\ \sum_n \sigma_{13n}^+(\sigma_{31n}^+)(t) \end{pmatrix} \\ &= \exp[-i(\omega_0 \mp 2\epsilon)t] \\ &\quad \times \begin{bmatrix} a_1 \begin{pmatrix} 1 \\ 1 \end{pmatrix} \exp(-\frac{3}{2}\gamma t) \\ + a_2 \begin{pmatrix} 1 \\ -1 \end{pmatrix} \exp(-\frac{7}{2}\gamma t) \end{bmatrix}, \end{aligned} \quad (20)$$

where again the constants $\{a_i\}$ can be obtained from initial conditions, but are not required.

B. $\gamma/\epsilon \lesssim |\alpha| \lesssim \frac{2}{3}\epsilon/\gamma$

For values of α within this range, it is easily verified that the spectral lines at ω_{ij} ($i \neq j$) are all nonoverlapping. The equations of evolution of the corresponding density matrix elements are therefore uncoupled, can be written in the form

$$\begin{aligned} \dot{\sigma}_{ij}^+(t) &= -[i\omega_{ij} + \frac{1}{2}(\Gamma_i + \Gamma_j)]\sigma_{ij}^+(t) \\ &\quad + (\gamma/2|\vec{d}_{ee}|^2)d_{ii}^-d_{jj}^+\sigma_{ij}^+(t) \end{aligned} \quad (21)$$

and have the solutions

$$\begin{aligned} \rho_{ij}^+(t) &= \sum_n \sigma_{ij}^+(t) \\ &= \rho_{ij}^+(0) \exp\left[-i\omega_{ij} - \frac{1}{2}\left(\Gamma_i + \Gamma_j - \frac{\gamma d_{ii}^- d_{jj}^+}{|\vec{d}_{ee}|^2}\right)t\right]. \end{aligned} \quad (22)$$

C. $0 < |\alpha| \lesssim \gamma/\epsilon$

For small but nonzero values of the dipole-dipole energy, a simple solution to the master equation of the type used in this article is not possible due to partial overlap of the spectral lines (cf. CR⁷, Appendix). The results obtained in Sec. IV for the spectrum of the collectively radiating two-atom system will therefore be invalid for $0 < |\alpha| \lesssim \gamma/\epsilon$. The error, however, is found to lie only in the widths of the lateral components of the spectrum, and within this region of α an approximate solution will be assumed in the calculations of Sec. V of the effects of collective atomic scattering on the fluorescence spectra of atomic beams having different number densities $\langle N \rangle$.

D. $|\alpha| \gtrsim \frac{2}{3}\epsilon/\gamma$

For very large values of the dipole-dipole interaction, partial overlap of the spectral lines again occurs, and makes impossible a simple solution of the master equation. However, it can be shown that the spectrum obtained in Sec. IV B for $\gamma/\epsilon \lesssim |\alpha| \lesssim \frac{2}{3}\epsilon/\gamma$ is still correct in the region $|\alpha| \gtrsim \frac{2}{3}\epsilon/\gamma$ up to terms of order α^{-2} . For that reason, and because only a very small fraction of the atoms are separated by distances small enough to produce such large values of $|\alpha|$ at the atomic densities considered (see Sec. V), we will use the spectrum obtained in Sec. IV B for all values of $|\alpha| \gtrsim \gamma/\epsilon$.

IV. FLUORESCENCE SPECTRUM AS A FUNCTION OF α

The fluorescence spectrum is given by the real part of the Fourier transform of the correlation function of the dipole-moment operator, $\langle \vec{D}^+(t)\vec{D}^-(t') \rangle$, $t > t'$. From the fluctuation-regression theorem,¹¹ it is well known that for $t > t'$ the two-time average $\langle \vec{D}_{ij}^+(t)\vec{D}^-(t') \rangle$ satisfies the same equation of motion as the one-time average $\langle \vec{D}_{ij}^+(t) \rangle$. Furthermore, it is easily verified⁷ that the quantities $\langle \vec{D}_{ij}^+(t) \rangle / d_{ij}^+$ satisfy the same equations of motion as do the density matrix elements $[\rho_{ij}^+(t)]^*$. The equations of motion are thus solved

for the correlation functions $\langle \vec{D}_{ij}^+(t) \vec{D}^-(t') \rangle$, and the real parts of their Fourier transforms added to yield the fluorescence spectrum.

A. $\alpha = 0$

In the steady state, the correlation functions $\langle \vec{D}_{ij}^+(t') \vec{D}^-(t') \rangle$ are given by the expressions⁷

$$\langle \vec{D}_{ij}^+(t') \vec{D}^-(t') \rangle = (\gamma/2 |d_{ge}|^2) a_{ij}^+ d_{ji}^- \rho_{ii}^0(t'). \quad (23)$$

Using Eqs. (14), (15), (18), (20), and (23), we obtain the following set of steady-state values for the correlation functions $\langle \vec{D}_{ij}^+(t) \vec{D}^-(t') \rangle$:

$$\begin{aligned} \langle \vec{D}_{21}^+(t) \vec{D}^-(t') \rangle &= \langle \vec{D}_{13}^+(t) \vec{D}^-(t') \rangle \\ &= \frac{1}{3} \gamma \exp[i(\omega_0 - 2\epsilon) - \frac{3}{2} \gamma](t - t'); \\ \langle \vec{D}_{12}^+(t) \vec{D}^-(t') \rangle &= \langle \vec{D}_{31}^+(t) \vec{D}^-(t') \rangle \\ &= \frac{1}{3} \gamma \exp[i(\omega_0 + 2\epsilon) - \frac{3}{2} \gamma](t - t'); \\ \langle \vec{D}_{23}^+(t) \vec{D}^-(t') \rangle &= \langle \vec{D}_{32}^+(t) \vec{D}^-(t') \rangle \\ &= \langle \vec{D}_{11}^+(t) \vec{D}^-(t') \rangle = 0; \\ \langle \vec{D}_{22}^+(t) \vec{D}^-(t') \rangle &= \langle \vec{D}_{33}^+(t) \vec{D}^-(t') \rangle \\ &= \frac{2}{3} \gamma \exp(i\omega_0 - \gamma)(t - t'). \end{aligned} \quad (24)$$

The spectrum in the absence of dipole-dipole interaction is then found to be identical with the familiar triplet of the single-atom system,¹²

$$\begin{aligned} I(\omega, 0) &= \frac{\gamma^2}{3\pi} \left(\frac{3}{(\omega - \omega_0 + 2\epsilon)^2 + (3\gamma/2)^2} \right. \\ &+ \frac{3}{(\omega - \omega_0 - 2\epsilon)^2 + (3\gamma/2)^2} \\ &\left. + \frac{4}{(\omega - \omega_0)^2 + \gamma^2} \right), \end{aligned} \quad (25)$$

in agreement with the high-field results for two-atom systems obtained numerically by Agarwal *et al.*¹

B. $\gamma/\epsilon \leq |\alpha| \leq 2/3 \epsilon/\gamma$

From Eqs. (14), (15), (18), (22), and (23), we obtain the following set of steady-state values for the correlation functions:

$$\begin{aligned} \langle \vec{D}_{11}^+(t) \vec{D}^-(t') \rangle &= 0; \\ \langle \vec{D}_{22}^+(t) \vec{D}^-(t') \rangle &= \pm \frac{8\gamma}{3\alpha\beta S(4 + \alpha^2)} \exp[i\omega_0(t - t')] \\ &\times \{ (S - 4)[4 + S \pm (\alpha/2\beta)(8 + \alpha^2 + S)] \\ &\times \exp[-(\gamma/\beta^2)(8 + \alpha^2 + S)(t - t')] \\ &+ (S + 4)[4 - S \pm (\alpha/2\beta)(8 + \alpha^2 - S)] \\ &\times \exp[-(\gamma/\beta^2)(8 + \alpha^2 - S)(t - t')] \}; \\ \langle \vec{D}_{21}^+(t) \vec{D}^-(t') \rangle &= \frac{1}{3} \gamma (1 + \alpha/2\beta) \\ &\times \exp[i(\omega_0 \mp \frac{1}{2} \alpha \epsilon \mp \beta \epsilon) \\ &- \frac{1}{2} \gamma (5 + \alpha/2\beta)](t - t'); \\ \langle \vec{D}_{13}^+(t) \vec{D}^-(t') \rangle &= \frac{1}{3} \gamma (1 - \alpha/2\beta) \\ &\times \exp[i(\omega_0 \pm \frac{1}{2} \alpha \epsilon \mp \beta \epsilon) \\ &- \frac{1}{2} \gamma (5 - \alpha/2\beta)](t - t'); \\ \langle \vec{D}_{23}^+(t) \vec{D}^-(t') \rangle &= \frac{1}{3} \gamma (\alpha^2/2\beta^2) \\ &\times \exp[i(\omega_0 \mp 2\beta \epsilon) \\ &- \gamma(3 - 8/\beta^2)](t - t'). \end{aligned} \quad (26)$$

The two-atom spectrum, including the effects of dipole-dipole interaction, is given by the expression

$$\begin{aligned} I(\omega, \alpha) &= \frac{\gamma^2}{3\pi} \left[\left(1 + \alpha/2\beta \right)^{\frac{1}{2}} \left(5 + \alpha/2\beta \right) \left(\frac{1}{(\omega - \omega_0 + \frac{1}{2} \alpha \epsilon + \beta \epsilon)^2 + \frac{1}{4} \gamma^2 (5 + \alpha/2\beta)^2} + \frac{1}{(\omega - \omega_0 - \frac{1}{2} \alpha \epsilon - \beta \epsilon)^2 + \frac{1}{4} \gamma^2 (5 + \alpha/2\beta)^2} \right) \right. \\ &+ \left(1 - \frac{\alpha}{2\beta} \right)^{\frac{1}{2}} \left(5 - \frac{\alpha}{2\beta} \right) \left(\frac{1}{(\omega - \omega_0 + \frac{1}{2} \alpha \epsilon - \beta \epsilon)^2 + \frac{1}{4} \gamma^2 (5 - \alpha/2\beta)^2} + \frac{1}{(\omega - \omega_0 - \frac{1}{2} \alpha \epsilon + \beta \epsilon)^2 + \frac{1}{4} \gamma^2 (5 - \alpha/2\beta)^2} \right) \\ &+ \left(\frac{\alpha^2}{2\beta^2} \right) \left(3 - \frac{8}{\beta^2} \right) \left(\frac{1}{(\omega - \omega_0 + 2\beta \epsilon)^2 + \gamma^2 (3 - 8/\beta^2)^2} + \frac{1}{(\omega - \omega_0 - 2\beta \epsilon)^2 + \gamma^2 (3 - 8/\beta^2)^2} \right) \\ &\left. + \left(\frac{8}{\beta^2 S} \right) \left((S - 4 + \frac{1}{4} \alpha^2) \frac{(8 + \alpha^2 + S)/\beta^2}{(\omega - \omega_0)^2 + (\gamma/\beta^2)^2 (8 + \alpha^2 + S)^2} + (S + 4 - \frac{1}{4} \alpha^2) \frac{(8 + \alpha^2 - S)/\beta^2}{(\omega - \omega_0)^2 + (\gamma/\beta^2)^2 (8 + \alpha^2 - S)^2} \right) \right]. \end{aligned} \quad (27)$$

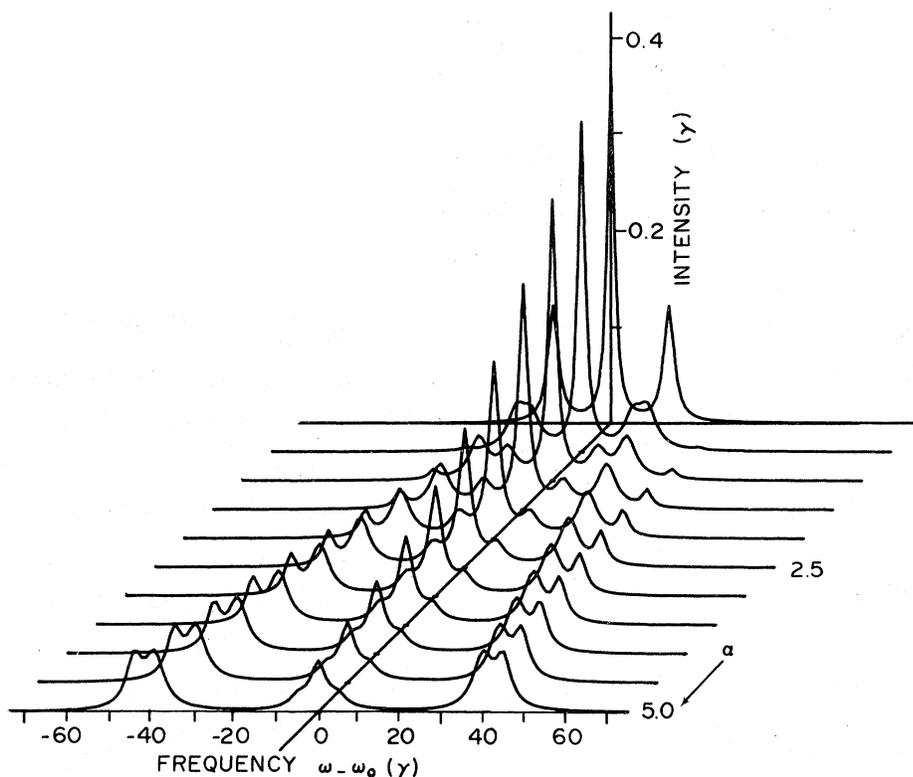


FIG. 2. Fluorescence spectra of two-atom systems with dipole-dipole interaction energy $\alpha\hbar\epsilon$, plotted for $\epsilon = 7\gamma$. The spectrum is symmetric with respect to α .

This spectrum has, in general, eight components, two centered at ω_0 , and three sets of equally displaced satellites, at frequencies $\omega_0 \pm (\frac{1}{2}\alpha + \beta)\epsilon$, $\omega_0 \pm (\frac{1}{2}\alpha - \beta)\epsilon$, and $\omega_0 \pm 2\beta\epsilon$. Their widths and integrated intensities are easily read from Eq. (27). The spectrum is plotted in Fig. 2, and for $\alpha \geq 1$ is clearly very different from the single-atom spectrum. The integrated fluorescence intensity is, however, easily verified to be independent of α , so that the results obtained by Agarwal *et al.*¹ for the intensity-correlation function of the collective two-atom system are not altered by the inclusion of the dipole-dipole interaction.

V. FLUORESCENCE SPECTRUM OF AN ATOMIC BEAM OF NUMBER DENSITY $\langle N \rangle$

In the preceding sections, we demonstrated that the fluorescence spectrum of a two-atom system with small atomic separation is very different from that of a one-atom system. In this section we use these results to obtain the spectrum of an idealized atomic beam of number density $\langle N \rangle$, consisting of randomly distributed atoms moving with uniform velocity. This calculation provides a qualitative illustration of how the dipole-dipole interaction would be expected to affect the spec-

trum of a real atomic beam. To obtain quantitatively the spectrum of a real beam, in which the atoms have a significant relative motion, requires the use of more sophisticated methods of pressure-broadening theory (our treatment is equivalent to the quasistatic approximation).

The constant α which characterizes the strength of the dipole-dipole interaction is a function of the separation \bar{R} between the atoms; from Eq. (4),

$$\alpha(R, \theta) = \frac{3}{2}(1 - 3\cos^2\theta)(c/\omega_0 R)^3(\gamma/\epsilon). \quad (28)$$

Equation (27) which gives the fluorescence spectrum of a pair of atoms as a function of α is valid only for $\gamma/\epsilon \lesssim |\alpha| \lesssim \frac{2}{3}\epsilon/\gamma$. To estimate the spectrum produced by the atoms in the beam with their varying separations, we first calculate the separation R_0 for which $|\alpha(R_0, 0)| = \gamma/\epsilon$. For the Na transition used in the experiments performed to date, we obtain for R_0

$$R_0 = 3^{1/3}c/\omega_0 = 1352 \text{ \AA}. \quad (29)$$

This separation is used to divide the pairs of atoms into two groups: For all pairs separated by $R \lesssim R_0$, $|\alpha| \gtrsim \gamma/\epsilon$, and the spectrum is given by Eq. (27)—exactly for $|\alpha| \lesssim \frac{2}{3}\epsilon/\gamma$, and with errors of order α^{-2} for $|\alpha| \gtrsim \frac{2}{3}\epsilon/\gamma$, as discussed in Sec. III D. For all pairs separated by $R > R_0$, we approximate the spectrum by Eq. (25). Taking

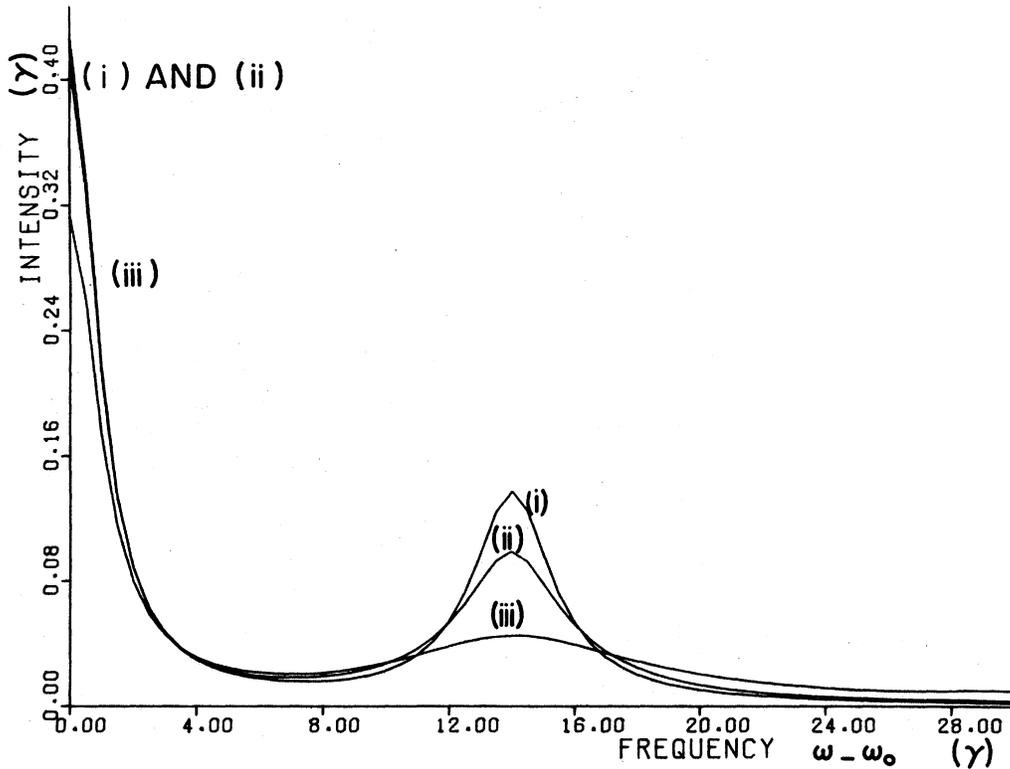


FIG. 3. Fluorescence spectra of idealized atomic beams of number density $\langle N \rangle$, with $\epsilon = 7\gamma$. The spectra are symmetric about $\omega - \omega_0 = 0$. Curves (i), (ii), and (iii) correspond to $\langle N \rangle = 10^{13} \text{ cm}^{-3}$, $\langle N \rangle = 10^{14} \text{ cm}^{-3}$, and $\langle N \rangle = 10^{15} \text{ cm}^{-3}$, in that order.

the limit of $\alpha \rightarrow 0$ in Eq. (27) gives the expression

$$\lim_{\alpha \rightarrow 0} I(\omega, \alpha) = \frac{\gamma^2}{3\pi} \left(\frac{5}{(\omega - \omega_0 + 2\epsilon)^2 + (5\gamma/2)^2} + \frac{5}{(\omega - \omega_0 - 2\epsilon)^2 + (5\gamma/2)^2} + \frac{4}{(\omega - \omega_0)^2 + \gamma^2} \right), \quad (30)$$

and the correct expression for the spectrum of pairs corresponding to values of α in the range $0 < |\alpha| \leq \gamma/\epsilon$ should be somewhere between Eqs. (25) and (30). Thus the error introduced by our use of Eq. (25) involves primarily an underestimate of the widths and overestimate of the peak heights of the lateral components of the spectrum at $\omega_0 \pm 2\epsilon$.

We assume an idealized beam in which the atoms are randomly distributed with average number density $\langle N \rangle$, and all move with the same velocity, so that the separation between a pair remains constant during the radiation process. A simple calculation of the probability $P(R)$ of finding a pair of nearest-neighbor atoms separated by a distance R gives the result

$$P(R) = 4\pi \langle N \rangle R^2 \exp(-\frac{4}{3}\pi \langle N \rangle R^3). \quad (31)$$

We then write for the spectrum of the atomic beam

$$I(\omega) = (4\pi)^{-1} \int_{4\pi} d\Omega \int_0^{R_0} I(\omega, \alpha) P(R) dR + I(\omega, 0) \left(1 - \int_0^{R_0} P(R) dR \right). \quad (32)$$

Equation (32) is plotted in Fig. 3 for a number of values of $\langle N \rangle$. For $\langle N \rangle \leq 10^{13} \text{ cm}^{-3}$, the spectrum is essentially indistinguishable from the single-atom spectrum.¹² As the density increases, however, the average separation of the atoms decreases, and the average dipole-dipole interaction increases in magnitude. The effects on the spectrum are to diminish the intensity of the central component, and to increase both the widths and the intensities of the lateral peaks. These lateral peaks are in fact each a superposi-

tion of a number of satellite lines, and with increasing atomic density become so broad as to be difficult to distinguish at all. Thus the effects of dipole-dipole interaction on the resonance fluores-

cence spectra in intense radiation fields cannot be neglected for more dense atomic beams.

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²H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. A 18, 201 (1978).

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⁴Neglecting dipole-dipole interaction, Agarwal *et al.*¹ find the two-atom and single-atom spectra to be identical at high field strengths.

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⁸The absorption spectrum of this system in a probe field is easily calculated as well, but is of practical interest only when using a detuned driving field [cf. Ref. 7; F. W. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow,

Phys. Rev. Lett. 38, 1077 (1977); Helen S. Freedhoff, J. Phys. B 11, 811 (1978)]. As we are concentrating here on collective atomic effects, we restrict our calculations to resonant driving fields and the fluorescence spectrum.

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¹⁰Corrections to these conditions involve terms of order γ/ϵ , which we neglect.

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