Resonance fluorescence from a $j = 1/2$ to $j = 1/2$ transition

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The calculation is given of the spectral distribution of resonance fluorescence from an atom that is driven near a $j = 1/2$ to $j = 1/2$ transition by a monochromatic, x-polarized electric field and that is subject to radiation damping only. In accordance with Mollow's results for a two-level system, the power spectrum of the scattered radiation corresponding to the x component of the dipole moment contains a coherent part for all driving-field intensities. The coherent part is absent in the fluorescence radiation corresponding to the y and z components of the dipole moment. For weak driving fields all power scattered by these latter components is present in a narrow Lorentzian. Consistent with energy conservation, the width of the Lorentzian corresponds to the probability of excitation of the atom out of its ground level due to the presence of the driving field. For strong driving fields the power spectra corresponding to the x component and to the y or z component mainly differ in the heights and widths of their central incoherent band and their Rabi-precession-frequency–shifted side bands.

Recently, there has been considerable theoretical[1–3] and experimental[4–6] interest in the power spectrum of electric dipole resonance fluorescence induced by a monochromatic electric field as a function of the field amplitude. Mollow[1] treated the fluorescence from a two-level atom driven by a classical electric field which oscillates near the transition frequency. In his work, equilibrium with the driving field is established by radiation damping only. For a weak classical field the power spectrum of the scattered field is given by a δ function centered at the driving frequency. This is consistent with the quantum-mechanical notion of energy conservation for the ingoing and outgoing quanta. It is also in agreement with the notion that resonance fluorescence in a weak field is nothing but scattering which has its source in a damped harmonic dipole oscillator coherently driven by the incident field. For higher field amplitudes Mollow predicts two contributions to the power spectrum: (a) a δ function representing coherent scattering with an intensity increasing less than linearly with the incoming intensity I and decreasing to zero for $I \to \infty$, and (b) an incoherent contribution appearing only at higher intensities with a spectrum that broadens with increasing intensity I and finally features a three-peak structure in which the Rabi frequency can be recognized.

This simple picture is no longer applicable in the case of an atom in isotropic surroundings, because then at least either the upper or the lower level must be degenerate for dipole transitions to be allowed. In general, the three components of the electric dipole vector cannot, in such a situation, be independently treated. For instance, if an atom with a $j = \frac{1}{2} \to j = \frac{1}{2}$ transition is driven by an x-polarized classical electric field, we expect, because of the resulting stationary occupation of the upper levels, fluorescence corresponding to radiation from the y and z components of the dipoles as well as from the x component. For symmetry reasons, only the fluorescence from the latter component can have the δ-function part in the spectrum describing the coherent scattering part, also at higher intensities.

Therefore it is of interest to study the fluorescence arising from the y or z components of the dipole: coherent scattering cannot contribute, but, on the other hand, for weak fields the power spectrum must be narrow because of the energy conservation argument. In this paper we shall give a short account of the actual calculation for a $j = \frac{1}{2} \to j = \frac{1}{2}$ transition along the lines of Mollow and discuss the results.

We consider an atom at rest at the position $\vec{r} = \vec{0}$ and coupled to the electromagnetic field in the electric dipole approximation, i.e., by the interaction Hamiltonian

$$H = -\vec{\mu} \cdot \vec{E}(\vec{r} = \vec{0}, t). \tag{1}$$

The electric field $\vec{E}(\vec{r}, t)$ is the sum of a classical, monochromatic, x-polarized field,

$$\vec{E}_c(\vec{r}, t) = \vec{e}_x E_0(\vec{r}) \cos\omega t, \tag{2}$$

and a quantum-mechanical field with Schrödinger operator $\vec{E}(\vec{r})$ acting on the states of the radiation field in empty space. $\vec{e}_x$ denotes the unit vector in the x direction. The circular frequency splitting between the $j = \frac{1}{2}$ excited level and the $j = \frac{1}{2}$ ground level is ω_0. The two ground states with $m_z = -\frac{1}{2}$ and $m_z = +\frac{1}{2}$ will be denoted by $|1\rangle$ and $|2\rangle$, respectively, and the corresponding excited states by $|3\rangle$ and $|4\rangle$.

In the scattering region, where the incident field $\vec{E}_c$ vanishes, and at a large enough distance r from the atom to be in the radiation zone, the radiation

reemitted with a (complex) polarization vector $\vec{e}_d$ ($\perp \vec{r}$) satisfies

$$\langle [\vec{e}_d^{\,*} \cdot \vec{E}^{(-)}(\vec{r},t)][\vec{e}_d \cdot \vec{E}^{(+)}(\vec{r},t')]\rangle$$

$$= \frac{\omega_0^4}{c^4 r^2} \left\langle \left[\vec{e}_d^{\,*} \cdot \vec{\mu}^{(-)}\left(t - \frac{r}{c}\right)\right]\left[\vec{e}_d \cdot \vec{\mu}^{(+)}\left(t' - \frac{r}{c}\right)\right]\right\rangle. \quad (3)$$

Here $\vec{E}^{(-)}(t)$ and $\vec{E}^{(+)}(t)$ are the usual negative- and positive-frequency parts of the Heisenberg field operator and $\vec{\mu}^{(-)}(t)$ and $\vec{\mu}^{(+)}(t)$ are the negative- and positive-frequency parts of the Heisenberg atomic electric dipole operator, and $\langle \cdots \rangle$ indicates an average over the steady-state ensemble under the influence of Eq. (2). In the steady state the correlation functions

$$g_{ij}(t,t') = \langle \mu_i^{(-)}(t)\, \mu_j^{(+)}(t')\rangle,$$

where $i,j = x,y,z$, depend only on $t' - t$ and satisfy $g_{ij}(t,0) = g_{ji}^*(0,t)$.

The power $S_j(\nu)\,d\nu$ reemitted in all directions by the j component of the atomic dipole in the circular frequency interval $d\nu$ is given by the spectral density

$$S_j(\nu) = \tfrac{3}{4}(\omega_0^4/c^3)\,\mathrm{Re}[\tilde{g}_{jj}(\nu)], \quad (4)$$

where

$$\tilde{g}_{jj}(\nu) = \frac{1}{2\pi}\int_0^\infty g_{jj}(t,0)e^{-i\nu t}\,dt. \quad (5)$$

To calculate $\tilde{g}_{jj}(\nu)$ we use the quantum regression theorem of Lax,[7] which formalizes the correspondence between the equations of motion of a two-time correlation function such as $g_{ij}(t,t')$ and the equations of motion of the reduced density matrix of the atom in the radiation field and driven by the classical field. The quantum radiation field is treated as a zero-temperature heat bath coupled to the atom in the Markoff approximation.[8,9] The resonant approximation is used, i.e.,

$$\vec{\mu} \cdot \vec{E}_c = \tfrac{1}{2}E_0 \vec{e}_x \cdot (\vec{\mu}^{\downarrow} e^{i\omega t} + \vec{\mu}^{\uparrow} e^{-i\omega t}),$$

where the lowering operator $\vec{\mu}^{\downarrow}$ and the raising operator $\vec{\mu}^{\uparrow}$ are the Schrödinger operators corresponding to the Heisenberg operators $\vec{\mu}^{(+)}(t)$ and $\vec{\mu}^{(-)}(t)$, respectively. The only nonvanishing matrix elements of the components of $\vec{\mu}^{\downarrow}$ are

$$\mu = \langle 1 \,|\, \mu_x^{\downarrow} \,|\, 4\rangle = \langle 2 \,|\, \mu_x^{\downarrow} \,|\, 3\rangle = -i\langle 1 \,|\, \mu_y^{\downarrow} \,|\, 4\rangle$$

$$= i\langle 2 \,|\, \mu_y^{\downarrow} \,|\, 3\rangle = \langle 2 \,|\, \mu_z^{\downarrow} \,|\, 4\rangle = -\langle 1 \,|\, \mu_z^{\downarrow} \,|\, 3\rangle.$$

The matrix of $\vec{\mu}^{\uparrow}$ is the Hermitian conjugate of $\vec{\mu}^{\downarrow}$.

The equations of motion of the reduced density matrix elements are

$$\begin{aligned}
\frac{d}{dt}\hat{\rho}_{14} &= (i\Delta - 3\gamma)\hat{\rho}_{14} + iv(\rho_{44} - \rho_{11}),\\
\frac{d}{dt}\hat{\rho}_{23} &= (i\Delta - 3\gamma)\hat{\rho}_{23} + iv(\rho_{33} - \rho_{22}),\\
\frac{d}{dt}\rho_{11} &= 4\gamma\rho_{44} + 2\gamma\rho_{33} + iv(\hat{\rho}_{41} - \hat{\rho}_{14}),\\
\frac{d}{dt}\rho_{22} &= 4\gamma\rho_{33} + 2\gamma\rho_{44} + iv(\hat{\rho}_{32} - \hat{\rho}_{23}),\\
\frac{d}{dt}\rho_{33} &= -6\gamma\rho_{33} + iv(\hat{\rho}_{23} - \hat{\rho}_{32}),\\
\frac{d}{dt}\rho_{44} &= -6\gamma\rho_{44} + iv(\hat{\rho}_{14} - \hat{\rho}_{41}),
\end{aligned} \quad (6)$$

and $\rho_{kl} = \rho_{lk}^*$, where

$$\hat{\rho}_{14} = \rho_{14}e^{-i\omega t} \quad \text{and} \quad \hat{\rho}_{23} = \rho_{23}e^{-i\omega t}.$$

The further equations for ρ_{24}, ρ_{13}, ρ_{12}, and ρ_{34} (Ref. 3) are not needed for the calculation of g_{xx} and g_{yy} but have been used for g_{zz} to check that $g_{yy} = g_{zz}$, as it should. Furthermore, $\Delta = \omega_0 - \omega$, $v = \mu E_0(0)/(2\hbar)$ and $2\gamma = \frac{4}{3}\mu^2\omega_0^3 c^{-3}\hbar^{-1}$ is one-third of the spontaneous-decay rate of the upper level.

With the aid of the rate equations the stationary values $\langle\rho_{kl}\rangle$ of ρ_{kl} can be determined. The only nonvanishing $\langle\rho_{kl}\rangle$ are

$$\begin{aligned}
&\langle\rho_{41}\rangle = \langle\rho_{32}\rangle = v^{-1}\langle\rho_{33}\rangle(\Delta + 3i\gamma)e^{-i\omega t},\\
&\langle\rho_{14}\rangle = \langle\rho_{23}\rangle = \langle\rho_{41}\rangle^*,\\
&\langle\rho_{33}\rangle = \langle\rho_{44}\rangle = \tfrac{1}{2}v^2(\Delta^2 + 9\gamma^2 + 2v^2)^{-1},\\
&\langle\rho_{11}\rangle = \langle\rho_{22}\rangle = \tfrac{1}{2} - \langle\rho_{33}\rangle.
\end{aligned}$$

As a consequence of Eq. (6) the matrix elements of all $\rho_{kl}(t)$ can be linearly expressed in their values at an earlier time t':

$$\rho_{kl}(t) = \sum_{m,n} G_{kl,\,mn}(t,t')\rho_{mn}(t') \quad (t > t'). \quad (7)$$

Because of the structure of Eq. (6) the operator G satisfies symmetry relations such as $G_{14,\,14} = G_{23,\,23}$ or $G_{14,\,23} = G_{23,\,14}$, etc. Application of the regression theorem results in

$$g_{ij}(t,t') = \sum_{k,\,l,\,m,\,p,\,q} \langle\rho_{kl}\rangle G_{pq,\,ml}(t,t')(\mu_i^{\uparrow})_{qp}(\mu_j^{\downarrow})_{mk}. \quad (8)$$

Insertion of the values of the matrix elements of $\vec{\mu}$ leads to

$$\begin{aligned}
g_{xx,\,yy}(t,t') = {}& 2\langle\rho_{44}\rangle(G_{14,\,14} \pm G_{23,\,14})\mu^2\\
&+ 2\langle\rho_{41}\rangle(G_{14,\,11} \pm G_{23,\,11})\mu^2,
\end{aligned} \quad (9)$$

where the symmetry relations of G and the properties of $\langle\rho\rangle$ have been used.

The Fourier transform of $G(t,t')$ occurring when the spectral density expression (4) is worked out is most easily obtained by going over to the Fourier transform of the equations of motion [Eqs. (6)] of the reduced density matrix elements. One finally obtains

$$S_x(\omega+\lambda)=\frac{8}{3}\frac{\omega_0^4\mu^2}{c^3}\langle\rho_{44}\rangle\,\mathrm{Re}\left(\frac{1}{\pi i\lambda}\frac{[(\lambda-3i\gamma)^2-\Delta^2](\lambda-6i\gamma)-2v^2\lambda}{[(\lambda-3i\gamma)^2-\Delta^2](\lambda-6i\gamma)-4v^2(\lambda-3i\gamma)}\right), \tag{10}$$

$$S_y(\omega+\lambda)=\frac{8}{3}\frac{\omega_0^4\mu^2}{c^3}\langle\rho_{44}\rangle\,\mathrm{Re}\left(\frac{1}{\pi i}\frac{[(\lambda-3i\gamma)^2-\Delta^2](\lambda-6i\gamma)-2v^2(\lambda-2i\gamma)}{[(\lambda-3i\gamma)^2-\Delta^2]\lambda(\lambda-6i\gamma)-4v^2(\lambda-3i\gamma)(\lambda-2i\gamma)}\right). \tag{11}$$

In accordance with Mollow[1] the power spectrum $S_x(\nu)$ for all driving-field intensities contains a coherent part given by

$$S_x^{\mathrm{coh}}(\nu)=\frac{8}{3}\frac{\omega_0^4\mu^2}{c^3}\langle\rho_{44}\rangle\,\frac{\Delta^2+9\gamma^2}{\Delta^2+9\gamma^2+2v^2}\,\delta(\nu-\omega). \tag{12}$$

It formally originates from the pole at $\lambda=0$ in Eq. (10). For small fields ($v\to 0$), $S_x^{\mathrm{coh}}(\nu)$ equals the total $S_x(\nu)$.

As was to be expected, the integrated power $\int S_y(\nu)\,d\nu$ equals $\int S_x(\nu)\,d\nu$ for all values of v, as is easily seen from the identical behavior of Eqs. (10) and (11) for $\lambda\to\infty$. However, S_y does not have a pole at $\lambda=0$ and therefore does not contain a coherent part. Instead, for small v, Eq. (11) has a pole at $\lambda=4iv^2\gamma(\Delta^2+9\gamma^2)^{-1}$. For small v *all* scattered power S_y is present in a narrow Lorentzian given by

$$S_y(\nu)=\frac{8}{3}\frac{\omega_0^4\mu^2}{c^3}\langle\rho_{44}\rangle\,\frac{4\gamma v^2(\Delta^2+9\gamma^2)}{(\Delta^2+9\gamma^2)^2(\nu-\omega)^2+16v^4\gamma^2}. \tag{13}$$

The result (13) is consistent with energy conservation in the sense that the width of $S_y(\nu)$ corresponds to the probability of excitation of the atom out of the ground level due to the presence of the x-polarized driving field. Note that in the original *one-photon* treatment of resonance fluorescence of a simple two-level system a similar width occurs,[10,11] a result that is at variance with Mollow's conclusions in the case of monochromatic excitation.[1,12] In our present paper for a $j=\frac{1}{2}\to j=\frac{1}{2}$ atom we have demonstrated that the absorption width of the ground level does appear in the resonance fluorescence due to the y (and z) component of the dipole moment if the atom is driven by a monochromatic x-polarized field.

It is of interest to consider Eqs. (10) and (11) for large values of v or Δ ($\Delta^2+v^2\gg\gamma^2$). For $S_j(\nu)$ we have, in addition to the coherent part given by Eq. (12), the incoherent part

$$S_j^{\mathrm{incoh}}(\omega+\lambda)=\frac{8}{3}\frac{\omega_0^4\mu^2}{c^3}\langle\rho_{44}\rangle\,\mathrm{Re}\left(\frac{1}{\pi i}\sum_{k=0}^{3}\frac{A_k^j}{\lambda-\lambda_k^j}\right), \tag{14}$$

with

$$A_0^x=0,\quad A_1^x=8v^4(\Omega^2+\Delta^2)^{-1}\Omega^{-2},\quad A_2^x=A_3^x=v^2\Omega^{-2},$$

$$\lambda_1^x=3i\gamma(\Omega^2+\Delta^2)\Omega^{-2},\quad \lambda_{2,3}^x=\pm\Omega+3i\gamma(2v^2+\Omega^2)\Omega^{-2},$$

as in Mollow's paper, and

$$A_{0,1}^y=\tfrac{1}{2}(\Delta^2+2v^2)\Omega^{-2}$$
$$\pm\tfrac{1}{2}(4v^4-3\Delta^2\Omega^2)\Omega^{-2}(4v^4+9\Delta^2\Omega^2)^{-1/2},$$

$$A_{2,3}^y=v^2\Omega^{-2},$$

$$\lambda_{0,1}^y=\pm i\gamma\Omega^{-2}(4v^4+9\Delta^2\Omega^2)^{1/2}+i\gamma(3\Omega^2-2v^2)\Omega^{-2},$$

$$\lambda_{2,3}^y=\pm\Omega+i\gamma(3\Omega^2+2v^2)\Omega^{-2}.$$

In these expressions the role of the Rabi frequency $\Omega=(\Delta^2+4v^2)^{1/2}$ in the incoherent part of the radiation can be recognized in the values of λ_2 and λ_3, which are responsible for incoherent scattering at the side bands with $\nu\approx\omega\pm\Omega$.

Finally, we give the results for $v^2\gg\gamma^2$ and $\Delta=0$. For S_x we find, in agreement with Mollow, that the sidebands each carry one-fourth of the total scattered intensity $\int S_x(\nu)\,d\nu$ and the central incoherent band one-half. Because of the different widths, the peak-height ratio at $\nu\approx\omega+\Omega$, ω, and $\omega-\Omega$ is 1:3:1. For S_y we find the same intensity distributions and a peak ratio 3:7:3. Incidentally, for a 90° scattering experiment in which both S_x and S_y are observed the peak ratio is 8:21:8.

It is clear that these results hold only for the special case of the $j=\frac{1}{2}\to j=\frac{1}{2}$ transition studied in this paper. Generally speaking, the spectral distribution of the resonance fluorescence from the various components of the dipole moment will very much depend on the particular transition involved and on the state of polarization of the monochromatic driving fields.

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