Two-photon resonant scattering in strong fields

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An analysis is made of a three-level system in a strong monochromatic field. The field is in two-photon resonance with one of the transitions in the three-level system. Spontaneous radiation is emitted as a result of the other two transitions. The probabilities of finding a particle at the levels under steady-state conditions are calculated. Moreover, the spectral distribution of the emitted spontaneous photons is determined for both transitions.

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Much attention is currently given to the behavior of atomic systems in strong resonant fields. The case of one field modeled by a two-level system with radiation damping has been investigated quite thoroughly.[1,2] In the case of two fields depending monochromatically on time and resonating with two energy differences in atomic spectrum is usually modeled by a three-level system. Emission of spontaneous radiation from such a system has been studied recently. In the general case with arbitrary field intensities the solution of this problem is very tedious and possible only by numerical methods.[3]

We shall consider a three-level system shown in Fig. 1, which is perturbed by just one external monochromatic field. In the case of one-photon resonance of the field frequency with the atomic level difference ω_{31} we are dealing with the spontaneous Raman scattering, which can be investigated quite easily in a strong field.[4] The simplicity of the solution is due to the fact that the law of conservation of parity forbids the $2 \to 1$ spontaneous transition and there is no problem of a steady-state distribution of the level populations.

A system comprising external field photons and spontaneous photons is closed only if we assume that the states 1 and 3 are of the same parity and the difference between their energies ω_{31} is close to 2ω, where ω is the frequency of the external field $\mathscr{E}\cos\omega t$. Thus, the external field is in two-photon resonance with the $1 \rightleftharpoons 3$ transition. The state 3 is deexcited spontaneously to the state 2 of radiative width equal to γ_b in the absence of a strong external field. The state 2 is then deexcited to the state 1 of width γ_a (Fig. 1). It is assumed that detuning from the two-photon resonance

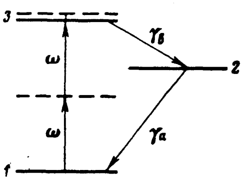

FIG. 1. Three-level system. The $1 \rightleftharpoons 3$ transitions are excited by two photons of an external strong field; the transitions $3 \to 2$ and $2 \to 1$ produce spontaneous radiation.

is sufficiently small that there is no stimulated one-photon emission.

We shall give the results of the solution of this problem in the steady-state case $t \gg 1/\gamma_a,\ 1/\gamma_b$, which is the one of greatest interest from the practical point of view. The aim is to find the steady-state population of the states 1, 2, and 3 of the three-level system mentioned above and the spectral distributions of the photons emitted spontaneously as a result of the $3 \to 2$ and $2 \to 1$ transitions. We shall assume the field $\mathscr{E}$ to be sufficiently strong to cause resonant mixing of the states 1 and 3 and we shall postulate that the two-photon absorption line becomes split. We shall also assume that the energy E_2 of the level 2 is not close to the energy $E_1 + \omega$. Otherwise, we are dealing with two resonant fields of the same frequency ω and the same intensity $\mathscr{E}$, which produce two one-photon resonances: $1 \rightleftharpoons 2$ and $2 \rightleftharpoons 3$. This case is discussed in detail by Whitley and Stroud.[3]

We shall now describe briefly the solution method. The system shown in Fig. 1 is described by Markov equations for the density matrix

$$i\partial\rho/\partial t = [\hat{H}, \rho] + i\hat{\gamma}\rho.$$

where $\hat{\gamma}$ is the spontaneous relaxation matrix and the Hamiltonian $\hat{H} = \hat{H}_0 + V$, in which $\hat{H}_0$ is the Hamiltonian of the unperturbed system and the potential V describes its dipole perturbations by a monochromatic external field. In the resonance approximation[5] the matrix elements V are the well-known two-photon matrix elements. For example,

$$V_{13} = V(e^{2i\omega t} + e^{-2i\omega t}), \quad V = d_{12}d_{23}\mathscr{E}^2/4(\omega_{21} - \omega).$$

Here, d_{ik} are the matrix elements of the dipole moment operator. In the resonance approximation there are two exponential functions of time but only one of them is important and it is the one which gives rise to small oscillations of the density matrix elements.

We can see that, in contrast to the one-photon case, allowance for the nonresonant shift of the levels in an alternating field is important in the two-photon resonance because this shift is of the same order of magnitude in respect of $\mathscr{E}$ (it is a quadratic function of the

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field intensity) as the resonant splitting of the levels into quasilevels. Nonresonant shifts are well known[6] and we shall assume them to be included in the energies of the levels E_i. Moreover, we shall assume that two or more photons are required to go over from the state 3 to the continuous spectrum because ionization predominates over spontaneous decay in the one-photon case.

A system of nine equations for the density matrix elements can now be written in the form

$$
\begin{aligned}
i\dot{\rho}_{11}&=V_{13}\rho_{31}-V_{31}\rho_{13}+2i\gamma_a\rho_{22}, \quad \rho_{21}=\rho_{12}^*,\\
\dot{\rho}_{22}&=2\gamma_b\rho_{33}-2\gamma_a\rho_{22}, \quad \rho_{32}=\rho_{23}^*,\\
i\dot{\rho}_{33}&=V_{31}\rho_{13}-V_{13}\rho_{31}-2i\gamma_b\rho_{33}, \quad \rho_{31}=\rho_{13}^*,\\
i\dot{\rho}_{12}&=\omega_{12}\rho_{12}-i\gamma_a\rho_{12}+V_{13}\rho_{32},\\
i\dot{\rho}_{23}&=\omega_{23}\rho_{23}-i(\gamma_a+\gamma_b)\rho_{32}-V_{13}\rho_{21},\\
i\dot{\rho}_{13}&=\omega_{13}\rho_{13}-i\gamma_b\rho_{13}+V_{13}(\rho_{33}-\rho_{11}).
\end{aligned}
\tag{1}
$$

The condition $\rho_{11}+\rho_{22}+\rho_{33}=1$ is satisfied here. We shall make the substitution of variables

$$\rho_{12}=\sigma_{12}e^{i(\omega_{12}-\delta/2)t}, \quad \rho_{23}=\sigma_{23}e^{i(\omega_{32}-\delta/2)t}, \quad \rho_{13}=\sigma_{13}e^{2i\omega t},$$

but for the other elements of the density matrix we shall retain $\rho_{ik}=\sigma_{ik}$. Here, $\delta=\omega_{31}-2\omega$ is the detuning from the two-photon resonance. The system (1) considered in the resonance approximation now reduces to a system of nine differential equations with constant coefficients in front of the elements σ_{ik}.

Under steady-state conditions, we have $\sigma=0$ and the system of equations for σ_{ik} reduces to the algebraic form. It can be solved quite simply. We shall only give the steady-state population of the level 3:

$$\rho_{33}=\frac{|V|^2\gamma_a}{\gamma_a(\delta^2+\gamma_b^2)+|V|^2(2\gamma_a+\gamma_b)}. \tag{2}$$

In a very strong field we have $\rho_{33}\to\gamma_a/(2\gamma_a+\gamma_b)$. As expected, for $\gamma_b\ll\gamma_a$, we approach $\rho_{33}\to1/2$, whereas for $\gamma_a\ll\gamma_b$, we obtain $\rho_{33}\to0$.

The total probability of the $3\to2$ spontaneous transition per unit time has the self-evident form: $w_{32}^{\text{tot}}=2\gamma_b\rho_{33}$. Since the conditions are steady, the total probability $w_{21}^{\text{tot}}=2\gamma_a\rho_{22}$ of the spontaneous transition $2\to1$ per unit time is equal to w_{32}^{tot}. The spectral composition of the spontaneous radiation can be determined if we know the time dependence of the elements σ_{ik}. If we assume $\sigma_{ik}\propto e^{i\lambda t}$, we obtain a ninth-order equation for the characteristic roots λ. The problem is simplified by the circumstance that the equations for σ_{12} and σ_{23} are independent of the equations for the other elements σ_{ik}. Since these are the matrix elements of interest to us in the determination of the spectral composition of the spontaneous radiation due to the $3\to2$ and $2\to1$ transitions (and this also applies to the conjugate matrix elements σ_{21} and σ_{32}), the quantity λ for these matrix elements assumes only four values which are equal in pairs. These values are readily found:

$$\lambda_\pm=i(\gamma_a+{}^1/_2\gamma_b)\pm(V^2+{}^1/_4\delta^2-{}^1/_4\gamma_b^2-{}^1/_2i\delta\gamma_b)^{1/2}. \tag{3}$$

The above roots give for each transition ($3\to2$ and $2\to1$) two Lorentzian spectral distributions for the spontaneous radiation, corresponding to the emission of light from the quasilevels of the state 3 and the absorption of light by the quasilevels of the state 1.

We shall first consider the transition $3\to2$. It follows from Eq. (3) that the emission spectrum consists of two peaks separated by

$$2\,\mathrm{Re}(V^2+{}^1/_4\delta^2-{}^1/_4\gamma_b^2-{}^1/_2i\delta\gamma_b)^{1/2}, \tag{4}$$

which can be regarded as the two-photon Rabi frequency.[6] It also follows from Eq. (3) that the widths of the peaks are, respectively,

$$\mathrm{Im}\,\lambda_\pm. \tag{5}$$

The peak amplitudes can be found by the technique of two-time correlation functions.[7] In view of the fact that the general results are cumbersome, we shall give only the probabilitites of emission of a spontaneous photon of frequency ν_b as a result of the $3\to2$ transition per unit time $dw_{32}(\nu_b)$ and we shall do this only for very strong fields $V\gg\gamma_a$, γ_b, δ. Then, on the one hand, we find that the expressions (4) and (5) simplify considerably and, on the other, the amplitudes of both peaks become identical and can easily be determined using the area under the peaks found above and equal to w_{32}^{tot}. The identity of the peak amplitude is a consequence of the equal populations of both quasilevels of the state 3 in a very strong field. We thus obtain

$$dw_{32}(\nu_b)=\left[\frac{\gamma_a\gamma_b}{(\nu_b-\omega_{32}-|V|)^2+(\gamma_a+{}^1/_2\gamma_b)^2}+\frac{\gamma_a\gamma_b}{(\nu_b-\omega_{32}+|V|)^2+(\gamma_a+{}^1/_2\gamma_b)^2}\right]\frac{d\nu_b}{2\pi} \tag{6}$$

In this case the two-photon Rabi frequency of a very strong field is $2|V|$. We can easily show that $\int dw_{32}(\nu_b)d\nu_b=w_{32}^{\text{tot}}$. It should be noted that the classical radiation generated by the average dipole moment is negligible in a strong field and all the radiation of Eq. (6) is due to quantum-mechanical fluctuations of the dipole moment.[7]

The expression for the probability $dw_{21}(\nu_a)$, where ν_a is the frequency of the spontaneous photons emitted as a result of the $2\to1$ transition, is of the form similar to Eq. (6) if we make the substitutions $y_b\to\nu_a$ and $\omega_{32}\to\omega_{21}$.

The line profile described by Eq. (6) is shown in Fig. 2 for the typical case of $\gamma_a=\gamma_b$. The peak splitting is simply the well-known Autler–Townes effect. The separation between the two peaks in Fig. 2 decreases on reduction of the perturbation V and retention of zero detuning $\delta=0$. It is clear from Eq. (4) that the separation between the peaks, expressed in terms of units of γ_a adopted in Fig. 2, is $[(2|V|/\gamma_a)^2-1]^{1/2}$. In a weak field, namely when $|V|<\gamma_a/2$, both quasilevels merge into one and we have one resonance corresponding to the results of the perturbation theory. The line profile shown in Fig. 2 is centered on the point $\nu_b-\omega_{32}=0$. We recall that because of the dynamic Stark effect the whole spectrum is shifted by an amount equal to the energy

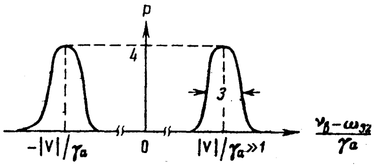

FIG. 2. Spectral distributions of the probability of the emission of light per unit time as a result of the $3 \to 2$ transition in the presence of a very strong field, calculated on the assumption that $\gamma_a = \gamma_b$. The ordinate gives $P = 9\,dw_{32}(\nu_b)(d\nu_b/2\pi)^{-1}$.

difference $\delta E_{32} = \delta E_3 - \delta E_2$, which is of the same order of magnitude as the separation between the resonances.

We shall conclude by pointing out the numerical criteria of strong fields. In the one-photon resonance case at optical frequencies a field can be regarded as strong if $\mathscr{E} > 10^2$ V/cm[4] and it is easy to estimate that in the two-photon resonance case considered here the critical field is considerably higher: $\mathscr{E} > 10^6$ V/cm.

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Transition radiation and transition scattering produced in a vacuum in the presence of a strong electromagnetic field

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In the presence of a strong electromagnetic field (in particular, a constant magnetic field), the vacuum behaves, as is well known, like a medium with permittivity and permeability that depend on the strong-field intensities. Transition radiation and transition scattering can therefore take place in vacuum. The article considers the transition radiation produced when a charge crosses the boundary between a strong magnetic field and a field-free region. The problems solved are those of transition scattering of sufficiently long strong electromagnetic waves by an immobile charge with frequency doubling, and of scattering without a change of frequency in the presence of a strong magnetic field. The same problems are considered also for a moving charge (in all considered cases the scattering takes place also for a charge with mass $M \to \infty$).

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Transition radiation is a rather common phenomenon which occurs when a charge or some other source (having no natural frequency) moves with constant velocity in or near an inhomogeneous medium. If the properties of the medium (the refractive index etc.) vary periodically in space and (or) in time, then the transition radiation acquires distinct features and can be called resonant transition radiation or transition scattering. The use of the last term is quite natural when one deals with a charge that is immobile relative to the medium and scatters a permittivity wave.[1] An effect analogous to transition scattering takes place in vacuum when a gravitational wave is incident on an immobile electric charge or dipole (electric or magnetic).[2]

We consider in this article transition radiation and transition scattering produced likewise in vacuum, but in the presence of a strong electromagnetic field. The gist of the matter that in a strong field electrodynamics becomes, as is well known, nonlinear even in vacuum, since the field gives rise to a vacuum polarization that is analogous to some extent to polarization of a medium. Transition radiation should therefore take place in an inhomogeneous strong field, and when a sufficiently strong wave is incident on a charge, transition scattering should take place. Of course, in a consistent quantum-electrodynamic calculation the transition effects are taken into account in the corresponding problems, but this calls for cumbersome computations. An exam-

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