SECOND-HARMONIC LIGHT SCATTERING NEAR TWO-PHOTON RESONANCE*

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The light scattered by a quantum system, two levels of which are distant by an energy equal to that of two photons of the incident beam, is shown to contain three lines: a central line at 2ω, and two satellites at $2\omega \pm \delta$, with δ and intensity-dependent level splitting term. The transition probabilities for the three lines are calculated in terms of the time evolution operator.

1. Introduction

A recent paper [1] deals with the possibilities of second-harmonic generation by a two-level quantum system at (one-photon) resonance with an external electromagnetic field, and predicts that the light generated by such a system should contain two satellites, $2\omega \pm 2\gamma$ (in addition to 2ω) due to splitting by the resonance-frequency field of each level into two sublevels [2] distant by 2γ, with 2γ dependent on the light intensity I (ac Stark effect). A similar phenomenon of energy level splitting occurs as well for two-photon resonance [3] but here the splitting $\delta \neq 2\gamma$ is a function of I^2 rather than of I. Interaction between a quantum system and an intense, periodically variable field can be dealt with quite generally in terms of quasi-energetic states [4].

We shall consider incoherent light scattering by a quantum system with two levels, fulfilling the condition of two-photon resonance, $2\omega \approx \omega_{u\ell}$, where $\omega_{u\ell}$ is the transition frequency, fig. 1. In our calculations, we omit Stark and Lamb displacements of the levels and their finite width.

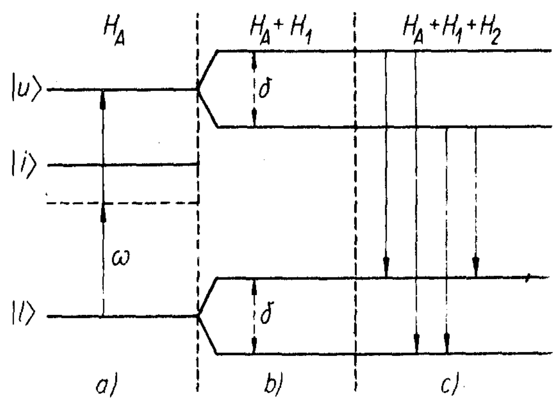

Fig. 1. a). Energy levels in the absence of a field; b). Level splitting in a two-photon resonance field; c). Possible transitions due to H_2-interaction.

2. Time evolution of the two-level system

The hamiltonian of our two-level system is

$$H = H_R + H_A + H_1 + H_2, \tag{1}$$

$$H_R = \sum_k \hbar\omega_k a_k^+ a_k, \tag{2}$$

$$H_A = \hbar\omega_{u\ell} S^z, \tag{3}$$

$$H_1 = \hbar(\eta^{(2)}\alpha^2 \exp(-i2\omega t)S^+ + \text{h.c.}), \tag{4}$$

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$$H_2 = \sum_k{}' \hbar(\eta_k^{(1)} a_k S^+ + \eta_k^{(1)*} a_k^+ S^-). \quad (5)$$

H_R describes the free radiation field, H_A the atomic system, H_1 its two-photon resonance [3] interaction with the incident beam, assumed to be in the coherent state $|\alpha\rangle$ [5], and H_2 its (weak) interaction with the field leading to spontaneous emission (the prime signifies that the incident beam is excluded from the sum); a_k and a_k^+ are photon annihilation and creation operators, whereas S^+, S^- and S^z are spin operators fulfilling the commutation rules:

$$[S^+, S^-] = 2S^z, \qquad [S^z, S^\pm] = \pm S^\pm \quad (6)$$

Furthermore, $\eta^{(2)}$ and $\eta_k^{(1)}$ are coupling constants.

$$\eta^{(2)} = -(\pi\omega_{u\ell}/V)\exp(i2\boldsymbol{k}\cdot\boldsymbol{r}) \times \frac{1}{\hbar}\sum_i{}' \frac{(\boldsymbol{\mu}_{ui}\cdot\boldsymbol{\varepsilon})(\boldsymbol{\mu}_{i\ell}\cdot\boldsymbol{\varepsilon})}{\omega_{i\ell}-\omega} \quad (7)$$

$$\eta_k^{(1)} = i\left(\frac{2\pi\omega_k}{\hbar V}\right)^{1/2} \exp(i\boldsymbol{k}\cdot\boldsymbol{r})\, \boldsymbol{\mu}_{u\ell}\cdot\boldsymbol{\varepsilon}_k, \quad (8)$$

where $\boldsymbol{\mu}_{u\ell}$, $\boldsymbol{\mu}_{i\ell}$ and $\boldsymbol{\mu}_{ui}$ are transition electric dipole matrix elements, $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}_k$ polarisation vectors of the incident and scattered beam, $\omega_{u\ell}$ the transition frequency between the upper $|u\rangle$ and lower $|\ell\rangle$ states, and $\boldsymbol{r}$ the position of the centre of mass. The prime in (7) means that $i \neq u,\ell$. In (4) and (5), we use the rotating-wave approximation, the legitimacy of which has been discussed recently [6,7]. We assume the two-level system to lack a centre of symmetry, i.e. to admit of one- and two-photon electric dipole type transitions between the two states under consideration.

Eqs. (1)–(5) permit to describe the time-evolution of the system. This can be achieved in two steps [1,4]: (*i*) Neglecting H_2, we obtain strict solutions for the matrix elements of the evolution operator, given by the equation

$$i\hbar \frac{dU}{dt} = H_1^{int} U, \quad (9)$$

where H_1^{int} is the hamiltonian in the interaction picture, and (*ii*) applying the perturbation method, taking H_2 into account, we solve the equation:

$$i\hbar \frac{dV}{dt} = H_2^{int} V, \quad (10)$$

with

$$H_2^{int} = U^+ \exp(i\omega_{u\ell} t S^z) H_2 \exp(-i\omega_{u\ell} t S^z) U. \quad (11)$$

Solving (9) and (11) with the initial condition $U = V = 1$ at $t = 0$, we obtain the state of the system at any moment of time t:

$$|\psi(t)\rangle = \exp[-(i/\hbar)H_A t]\, UV|\psi(0)\rangle. \quad (12)$$

We assume the initial state of the system in the form of a linear superposition of the states $|u\rangle$ and $|\ell\rangle$:

$$|\psi(0)\rangle = b_u|u\rangle + b_\ell|\ell\rangle. \quad (13)$$

3. Transition probabilities

The probability per second that a photon will appear in the kth mode irrespective of the state of the system is:

$$w = \frac{d}{dt}[|\langle u; 1_k|\psi(t)\rangle|^2 + |\langle \ell; 1_k|\psi(t)\rangle|^2], \quad (14)$$

where $|u; 1_k\rangle$ signifies that the atom is in the state $|u\rangle$ and one photon occurs in the kth mode. $|\psi(t)\rangle$ is given by (12), with (9), (10) and (13).

On insertion of (12) and integration over all possible values of the scattered wave vector, eq. (14) yields the following expression for the total probability per second of scattering, consisting of three parts with distinct frequencies:

$$w(2\omega) = \frac{16\pi^2(2\omega)^3 I^2}{3\hbar^3 c^5 \delta^2} |\mu_{u\ell}|^2 \left|\frac{1}{\hbar}\sum_i{}' \frac{(\boldsymbol{\mu}_{ui}\cdot\boldsymbol{\varepsilon})(\boldsymbol{\mu}_{i\ell}\cdot\boldsymbol{\varepsilon})}{\omega_{i\ell}-\omega}\right|^2 \quad (15)$$

$$w(2\omega\pm\delta) = \frac{(2\omega\pm\delta)^3(\delta\mp\Delta)^2}{6\hbar c^3 \delta^3} |\mu_{u\ell}|^2 \times [(\delta\pm\Delta)|b_\ell|^2 + (\delta\mp\Delta)|b_u|^2], \quad (16)$$

where $\Delta = 2\omega - \omega_{u\ell}$, the intensity $I = c\hbar\omega|\alpha|^2/V$ is that of the incident beam, and

$$\delta = \sqrt{\Delta^2 + 4|\eta^{(2)}\alpha^2|^2} \quad (17)$$

is the intensity-dependent splitting of the levels [3]. In the present, two-photon resonance case δ is a function of I^2, and not of I as in the case of one photon resonance [2]. If the system was initially in the state $|u\rangle$, i.e. $|b_u| = 1$, and in the absence of coupling with the field ($\eta^{(2)} = 0$), eq. (16) goes over into the well known expression of Einstein for spontaneous emission at the transition frequency $\omega_{u\ell}$. For low intensities $w(2\omega+\delta)\approx 0$, whereas $w(2\omega-\delta)$ can be non-zero, so that the satellites differ in height. For high intensities,

$$2|\eta^{(2)}||\alpha|^2 \gg \Delta, \quad \delta \approx 2|\eta^{(2)}||\alpha|^2,$$

we obtain a symmetric spectrum irrespective of the initial occupation of the levels:

$$w(2\omega\pm\delta) = \tfrac{1}{2} w(2\omega) = \frac{(2\omega)^3\, |\mu_{u\ell}|^2}{6\hbar c^3}. \qquad (18)$$

On going over to $2\omega \to \omega$ and $\delta \to 2\gamma$, eq. (18) becomes identical with the well-known expression for Rayleigh scattering near one-photon resonance [8]. In this interpretation, both processes are analogous.

The transition probability (15) for the central 2ω-line does not depend on the initial occupation of the levels and can be interpreted as second-harmonic light scattering [9]. It should be noted, however, that the mechanism leading to scattered photons of the frequencies 2ω and $2\omega\pm\delta$ is, in our case, quite different from that of the second harmonic generation considered in ref. [1].

4. Conclusions

Above, we have shown that second-harmonic light scattering near two-photon resonance, as considered here, and Rayleigh scattering near one-photon resonance [8], have much in common. Both processes lead to three lines: a central line of frequency 2ω and two satellities $2\omega\pm\delta$ (case 1), or respectively ω and $\omega\pm 2\gamma$ (case 2). In case 1, the splitting δ depends on I^2 rather than on I, as in case 2 for 2γ. Since light beams can attain intensities of $\sim 10^{20}$ erg/cm^{2}s, our δ can be of the order 10^{13} s^{-1}, and if $\Delta \sim 10^9$ s^{-1} then $2|\eta^{(2)}| \times |\alpha|^2 \gg \Delta$, and the transition probability is given by (18). Thus, for the satellites, this probability amounts to $\frac{1}{8}$ of Einstein's spontaneous emission coefficient ($\sim 10^8$ s^{-1}). So large an effect should be easily accessible to observation in the same materials where absorption of two photons is found.

The probabilities calculated by us are directly related with the total active cross-sections as follows:

$$\sigma(\omega_p) = (\hbar\omega/I)\, w(\omega_p).$$

Our considerations are based on a two-level approximation, although the coupling constant $\eta^{(2)}$, eq. (7), necessarily implies states other than $|u\rangle$ and $|\ell\rangle$ whose occupation numbers, however, vary but slightly in comparison with those of the states at resonance. Moreover, $\eta^{(2)}$ can differ from zero even in the strictly two-level case if the term $\sim \boldsymbol{A}\cdot\boldsymbol{A}$ in the interaction hamiltonian yielding the direct double-photon transition is taken into account [10]. On re-defining $\eta_k^{(1)}$ [eq. (8)] to include electric quadrupole and magnetic dipole transitions [10], the assumption of no centre of symmetry is no longer necessary. The system can as well be an atom in the ground state [11], leading nevertheless to double-photon scattering.

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