

Two-photon Resonance Fluorescence from a Three Level System

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Two-photon resonance fluorescence from a three level system is studied in the presence of a second electromagnetic field which couples the two-photon excited state resonantly to a third level. Because of this coupling the two-photon resonance spectrum consists of nine peaks, if the second field which couples the excited level to the third level is absent, we get the usual four peak spectrum.

O espectro da fluorescência de ressonância de dois fótons de um sistema de três níveis é estudado na presença de um segundo campo eletromagnético que acopla ressonantemente o estado excitado de dois-fótons a um terceiro nível. Devido a esse acoplamento o espectro ressonante de dois fótons consiste de nove picos. Se o segundo campo que acopla o nível excitado ao terceiro nível for desligado, obtém-se o usual espectro de quatro picos.

The problem of resonance fluorescence from a two level atom excited resonantly by a strong electromagnetic field has been a subject of much theoretical¹⁻⁸ and experimental⁹⁻¹¹ interest. The fluorescent spectrum from such a system consists of a central peak at the excitation frequency and two symmetrically placed side bands. The problem of a two-level atom interacting with a strong radiation field whose frequency is half that of the transition frequency of the system has been studied recently¹². It is shown that for such a system fluorescent spectrum consists of four peaks, centred at the laser, twice the laser and two other at high and low frequencies which are defined later in the paper.

In this paper we investigate what happens to the two-photon resonance spectrum when the two-photon excited level is resonantly coupled to a third level by a single photon. It will be shown that the spectrum consists of nine peaks.

To investigate the spectrum, we consider a three level atom whose energy level diagram is shown in the figure 1. The transition frequency between the first two levels is denoted by ω_1 and the transition frequency between the level $|2\rangle$ and $|3\rangle$ is denoted by ω_2 . The atom is pumped with two strong laser beams of frequencies ω_a and ω_b such that $2\omega_a = \omega_1$ and $\omega_b = \omega_2$. The atom is simultaneously coupled to the other modes which are initially empty. The Hamiltonian for such a system can be written as

$$\begin{aligned}
 H = & \hbar\omega_1 (a_2^\dagger a_2 + a_3^\dagger a_3) + \hbar\omega_b (a_3^\dagger a_3 + \beta_b^\dagger \beta_b) + \hbar\omega_a \beta_a^\dagger \beta_a \\
 & + A_1 (a_1^\dagger a_2 \beta_a^\dagger - a_2^\dagger a_1 \beta_a) + A_2 (a_2^\dagger a_3 \beta_b^\dagger - a_3^\dagger a_2 \beta_b) \\
 & + \sum_k B_{1k} (a_1^\dagger a_2 \beta_k^\dagger - a_2^\dagger a_1 \beta_k) + \sum_k B_{2k} (a_2^\dagger a_3 \beta_k^\dagger - a_3^\dagger a_2 \beta_k) + \sum_k \hbar\omega_k \beta_k^\dagger \beta_k
 \end{aligned} \tag{1}$$

where a_i^\dagger and a_i are the Fermion creation and destruction operators describing the electronic states of the atom $i=1,2,3$; and these operators obey the Fermi-Dirac statistics. β_j^\dagger and β_j are the photon creation and destruction operators for the laser and the fluorescence field $j = a, b$ and k . A_1, A_2, B_{1k} and B_{2k} are the usual imaginary coupling constants for the radiation and the atomic system. In eq.(1) we have adopted the rotating wave approximation (neglecting e.g., terms like $a_1^\dagger a_2 \beta_k - a_2^\dagger a_1 \beta_k^\dagger$).

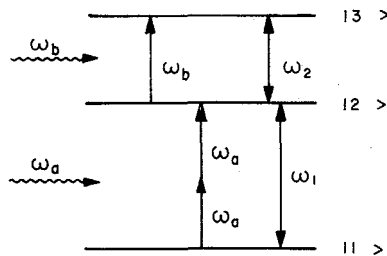


Fig.1 - Three-level system with two laser field of frequencies ω_a and ω_b respectively.

We have also assumed that level $|1\rangle$ does not couple with level $|3\rangle$ directly. From now on A will be taken to be unity. While working with the Hamiltonian we take into account the fact that $n_1 + n_2 + n_3 = 1$, where $n_i = a_i^\dagger a_i$, are the electron number operators. The above relation emphasizes the fact that the atom can only be in either of the three states and all other states of the system are quite far away from three states of interest.

We use retarded double-time Green functions to study the fluorescence spectra. Let $G_{12}(\omega)$ and $G_{23}(\omega)$ represent the Fourier transform of the Green's functions $\langle\langle a_{12}^\dagger; a_{21}^\dagger \rangle\rangle$ and $\langle\langle a_{23}^\dagger; a_{32}^\dagger \rangle\rangle$ respectively, that is

$$G_{12}(\omega) = \langle\langle a_{12}^\dagger; a_{21}^\dagger \rangle\rangle(\omega) \quad \text{and} \quad G_{23}(\omega) = \langle\langle a_{23}^\dagger; a_{32}^\dagger \rangle\rangle(\omega)$$

The expectation value here is taken in a product state of a monochromatic coherent state for each of the initially occupied field modes and an arbitrary initial state for the three level atom.

The excitation spectra due to the electronic transitions $1 \leftrightarrow 2$ and $2 \leftrightarrow 3$ are obtained from the imaginary parts of the Fourier transforms of the Green's functions G_{12} and G_{23} respectively, thus our problem reduces to the determination of the Green's functions G_{12} and G_{23} from the Hamiltonian given in eq.(1). In this paper we are interested only in looking at the effects of coupling of the two-photon excited level to the third level by a single photon. For this we need only $G_{12}(\omega)$, but if one is interested in the complete spectra from the system then $G_{23}(\omega)$ is also required. The properties of the Green's function can be found in the literature^{13,14}. The equation of motion for the Green's function is given by

$$\omega \langle\langle A(t); B(t') \rangle\rangle(\omega) = \frac{1}{2\pi} \langle [A(t); B(t)] \rangle + \langle\langle [A(t), H]; B(t') \rangle\rangle(\omega) \quad (2)$$

The operators $A(t)$ and $B(t')$ are in the Heisenberg representation, the subscript ω and the time argument will be suppressed for convenience in this paper.

Using the Hamiltonian given in eq.(1), from eq.(2), we get the following equation of motion for $G_{12}(\omega)$

$$\begin{aligned}
(\omega - \omega_1) G_{12}(\omega) &= \frac{\bar{n}_1 - \bar{n}_2}{2\pi} + A_1 \langle\langle (n_2 - n_1) \beta_a; a_2^+ a_1 \rangle\rangle + A_2 \langle\langle a_1^+ a_3 \beta_b^+; a_2^+ a_1 \rangle\rangle \\
&+ \sum_k B_{1k} \langle\langle (n_2 - n_1) \beta_k; a_2^+ a_1 \rangle\rangle + \sum_k B_{2k} \langle\langle a_1^+ a_3 \beta_k^+; a_2^+ a_1 \rangle\rangle . \quad (3)
\end{aligned}$$

Since the fluorescence is very weak as compared to the laser field, so we can make the following approximation

$$\langle\langle (n_2 - n_1) \beta_k; a_2^+ a_1 \rangle\rangle \approx (\bar{n}_2 - \bar{n}_1) \langle\langle \beta_k; a_2^+ a_1 \rangle\rangle .$$

Now writing the equation of motion for $\langle\langle \beta_k; a_2^+ a_1 \rangle\rangle$ and substituting its value back into eq.(3) we have the following equation,

$$g_{12} G_{12}(\omega) = \frac{(\bar{n}_1 - \bar{n}_2)}{2\pi} + A_1 \langle\langle (n_2 - n_1) \beta_a; a_2^+ a_1 \rangle\rangle + A_2 \langle\langle a_1^+ a_3 \beta_b^+; a_2^+ a_1 \rangle\rangle \quad (4)$$

where

$$\begin{aligned}
g_1 &= (\omega - \omega_1 - \frac{\gamma_1}{2}) , \\
\frac{\gamma_1}{2} &= \frac{B_{1k}^2 (1 - 2\bar{n}_2 - \bar{n}_3)}{\omega - \omega_k + i\Gamma}
\end{aligned}$$

and γ_2 which appears in later equations is given by

$$\frac{\gamma_2}{2} = \sum_k \frac{B_{2k}^2 (1 - 2\bar{n}_3 - \bar{n}_1)}{\omega - \omega_k + i\epsilon}$$

Imaginary parts of γ_1 and γ_2 are the spontaneous emission probabilities of level $|2\rangle$ and $|3\rangle$ respectively.

In the same approximation, we derive the following equations of motion

$$\begin{aligned}
g_a \langle\langle (n_2 - n_1) \beta_a; a_2^+ a_1 \rangle\rangle &= -2A_1 \langle\langle a_1^+ a_2 \beta_a^+; a_2^+ a_1 \rangle\rangle + \langle\langle a_2^+ a_1 \beta_a \beta_a; a_2^+ a_1 \rangle\rangle \\
&+ A_2 \langle\langle a_2^+ a_3 \beta_b^+; a_2^+ a_1 \rangle\rangle , \quad (5)
\end{aligned}$$

$$g_{2a} \langle\langle a_2^+ a_1 \beta_a \beta_a; a_2^+ a_1 \rangle\rangle = 2A_1 \bar{n}_a \langle\langle (n_2 - n_1) \beta_a; a_2^+ a_1 \rangle\rangle , \quad (6)$$

$$g_1 \langle \langle a_{12}^+ a_{21}^+ \beta_b^+ \beta_a^+ \rangle \rangle = \frac{(\bar{n}_1 - \bar{n}_2) \bar{n}_a}{2\pi} + 2A_1 \bar{n}_a \langle \langle (n_2 - n_1) \beta_a^+; a_{21}^+ \rangle \rangle + A_2 \bar{n}_a \langle \langle a_{13}^+ \beta_b^+; a_{21}^+ \rangle \rangle, \quad (7)$$

$$P \langle \langle a_{23}^+ \beta_b^+ \beta_a^+; a_{21}^+ \rangle \rangle = -A_1 \bar{n}_a (\omega - \omega_a - \gamma_2) \langle \langle a_{13}^+ \beta_b^+; a_{21}^+ \rangle \rangle, \quad (8)$$

$$Q \langle \langle a_{13}^+ \beta_b^+; a_{21}^+ \rangle \rangle = -\frac{A_2 (\bar{n}_1 - \bar{n}_2) \bar{n}_a}{2\pi} + A_2 Q \langle \langle a_{23}^+ \beta_b^+ \beta_a^+; a_{21}^+ \rangle \rangle, \quad (9)$$

where

$$g_a = (\omega - \omega_a - \gamma_1), \quad g_{2a} = \omega - (2\omega_a - \omega_1) - \frac{\gamma_1}{2}, \quad (9)$$

$$P = (\omega - \omega_a - \frac{\gamma_1 + \gamma_2}{2}) (\omega - \omega_a - \gamma_2) + 4A_2^2 \bar{n}_b + A_1^2 \bar{n}_a \quad (10)$$

and

$$Q = (\omega - \omega_a - \frac{\gamma_1}{2}) (\omega - \omega_1 - \frac{\gamma_2}{2}) + 4A_2^2 \bar{n}_b. \quad (11)$$

In deriving the above equations we have used the following decoupling scheme

$$\langle \langle (n_2 - n_3) \beta_b^+ \beta_b^+ \beta_a^+; a_{21}^+ \rangle \rangle \approx \bar{n}_b \langle \langle (n_2 - n_3) \beta_a^+; a_{21}^+ \rangle \rangle \quad (12)$$

and

$$\langle \langle a_{13}^+ \beta_b^+ \beta_a^+ \beta_b^+; a_{21}^+ \rangle \rangle \approx \bar{n}_a \langle \langle a_{13}^+ \beta_b^+; a_{21}^+ \rangle \rangle \quad (13)$$

where $\bar{n}_a = \langle \beta_a^+ \beta_a^+ \rangle$. The decoupling scheme of eqs. (12) and (13) is equivalent to the Hartree-Fock self consistent field approximation¹⁴ and this is expected to be valid in the limit of high photon densities, when

$$\bar{n}_a \text{ or } \bar{n}_b \gg 1 \quad \text{and} \quad \bar{n}_k = \langle \beta_k^+ \beta_k^+ \rangle \ll 1.$$

Using eqs. (5) to (9) after some rearrangement of terms, we put equation (4) in the following form

$$G_{12}(\omega) = \frac{\bar{n}_1 - \bar{n}_2}{2\pi} \left[\frac{1}{g_1} \left(1 - \frac{2A_1^2 \bar{n}_1}{L} \right) \left(1 - \frac{A_2^2 \bar{n}_2 P}{Q} \right) + \frac{A_1^2 A_2^2 \bar{n}_1 \bar{n}_2}{RLQ} \right] \quad (14)$$

where

$$L = \frac{g_\alpha g_{2\alpha} g_1 + 4A_1^2 \bar{n}_1 (g_1 + g_{2\alpha})}{g_{2\alpha}}$$

and

$$R = P + A_1^2 \bar{n}_1 \frac{\omega - \omega_1 - \gamma/2}{\omega - \omega_1 - \gamma/2}$$

If the second laser field is absent, that is when $\bar{n}_2 = 0$, eq.(12) reduces to

$$G_{12}(\omega) = \frac{\bar{n}_1 - \bar{n}_2}{2\pi} \left[\frac{1}{g_1} \left(1 - \frac{2A_1^2 \bar{n}_1}{L} \right) \right] \quad (15)$$

This is exactly the same expression for the Green function as in reference 12 for the case of two photon resonance fluorescence. Eq.(15) predicts for the fluorescent spectrum four Lorentzian peaks at the laser frequency, twice the laser frequency and two other peaks at ω_\pm and it is given by

$$\omega_\pm = \omega_\alpha \pm (\omega_\alpha^2 + 2\eta_\alpha^2)^{1/2}$$

where $\eta_\alpha = -4A_1^2 \bar{n}_1$, η_α^2 has the dimensions of the frequency squared, its value depends upon the strength of the laser field and the oscillator strength of the transition involved.

Green function in eq.(14) incorporates the effect of the coupling of the excited level to a third level on the two-photon resonance spectrum.

If one is concerned with the details of the spectrum, one has to get the imaginary part of the Green function given in eq.(14). As here we are concerned only with the number of peaks produced from the system, only the knowledge of propagators is enough to predict the number of peaks. The propagators g_1^{-1} and L^{-1} predict four peaks as mentio-

ned above for the case of two-photon resonance fluorescence. These peaks are due to the interaction of laser field of frequency ω_a with the atom. The propagators Q^{-1} describes two peaks at

$$\Omega_{\pm} = \omega_a \pm \eta_b \quad \text{where} \quad \eta_b = -4A_1^2 \bar{n}_b$$

These two peaks are a consequence of the interference of the second laser field into the lower system corresponding to the transition frequency ω_1 . We obtain three more peaks from the propagator R^{-1} .

The location of the peaks comes from the solution of the following cubic equation

$$x^3 - x^2 \left[\omega_a + \frac{\gamma_1 + \gamma_2}{2} \right] + x \left[\frac{\omega_a (\gamma_1 + \gamma_2)}{2} - \left(\eta_b^2 + \frac{\eta_a^2}{4} \right) \right] + \omega_a^2 \left[\eta_b^2 - \frac{\gamma_1 \gamma_2}{4} \right] = 0 \quad (16)$$

where

$$x = \omega - \omega_a - \frac{\gamma_2}{2}$$

The roots of eq. (16) cannot be put into a simple form; but the three roots lie within the limit of ω_{\pm} .

So in all we have nine peaks from such a system. If we set $\omega_1 = \omega_a$ that is we have a single photon resonance between the first two levels of the atom, the Green function in eq.(14) degenerates to the double optical resonance case and it gives seven peaks for the lower part of the spectrum as predicted in recent studies¹⁵⁻¹⁶. To my knowledge, no experimental results related to this are available, but I am sure that this theoretical study will give an incentive to experimentists to look for these frequencies in the fluorescent spectrum.

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