Detection of three-photon relaxation of an atom near a phase conjugator through absorption measurements

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Spontaneous excitation of an atom near a phase conjugator should be amenable to experimental verification through a measurement of the atomic probe absorption profile. It is shown that the absorption decreases quickly with increasing reflectivity, due to a population of the excited state. The responsible physical process is a three-photon event.

A two-state atom with resonance frequency ω_0 is located near the surface of a four-wave-mixing phase conjugator (PC). When the atom is in its excited state $|e\rangle$, then it can decay spontaneously to the ground state $|g\rangle$ under emission of a photon with frequency ω_0, as represented by diagram (a) in Fig. 1. The space near a four-wave-mixing PC always contains photons, which can be understood as follows.[1] The vacuum modes of the electromagnetic field interact with the two pump beams (frequency $\bar{\omega}$) through the third-order susceptibility of the medium. This generates the phase-conjugate replica of the vacuum field, which consists of photons traveling away from the crystal. These photons, with a frequency distribution around $\bar{\omega}$, can be absorbed by the atom when it is in its ground state. A subsequent spontaneous emission of a photon, followed by an absorption of a second photon with frequency $\bar{\omega}$, then leads to the energy-conserving diagram (b) in Fig. 1. After this three-photon process, the atom is in the excited state. This process is sometimes called spontaneous excitation,[2] although it is basically a combination of two stimulated absorptions and a spontaneous decay. It follows from the diagram that the fluorescent photon must have a frequency $2\bar{\omega} - \omega_0$.[3,4]

The fluorescence spectrum consists of two lines that are located symmetrically around the pump frequency $\bar{\omega}$, and detection of the line at $2\bar{\omega} - \omega_0$ would confirm the occurrence of the three-photon process. Alternatively, the temporal photon statistics carries information on this process.[5] A major experimental complication is that the fluorescent photons are emitted in the same directions as the photons that are emitted spontaneously by the PC. This could make the fluorescence signal indistinguishable from the background, and thereby undetectable. In this Brief Report we propose an alternative technique for the observation of three-photon events, which does not rely on fluorescence measurements. When a low-intensity laser beam is directed along the surface of the PC, then the atom will absorb photons from this beam. With I_p, ϵ, and ω designating the intensity, polarization, and frequency of the probe beam, respectively, the absorbed energy per unit of time is given by[6]

$$I(\omega) = \frac{I_p \omega}{\epsilon_0 \hbar c} \mathrm{Re} \int_0^\infty d\tau\, e^{i\omega\tau} \langle [\boldsymbol{\mu}(\tau)\cdot\boldsymbol{\epsilon}^*, \boldsymbol{\mu}(0)\cdot\boldsymbol{\epsilon}] \rangle , \quad (1)$$

with $\boldsymbol{\mu}$ the atomic dipole moment. This spectral absorption profile $I(\omega)$ will be affected by three-photon processes, but its measurement does not require the detection of any fluorescence.

The evaluation of $I(\omega)$ is straightforward. First, we transform the atom's correlation function in the integrand to the Schrödinger picture, which gives

$$\langle [\boldsymbol{\mu}(\tau)\cdot\boldsymbol{\epsilon}^*, \boldsymbol{\mu}(0)\cdot\boldsymbol{\epsilon}] \rangle = \mathrm{Tr}(\boldsymbol{\mu}\cdot\boldsymbol{\epsilon}^*) e^{-i(L_a - i\Gamma)\tau}[\boldsymbol{\mu}\cdot\boldsymbol{\epsilon}, \bar{\rho}] . \quad (2)$$

Here, the Liouvillians L_a and Γ account for the free evolution and the relaxation, respectively, and they are given by

$$L_a \sigma = \omega_0 [P_e, \sigma] , \quad (3)$$
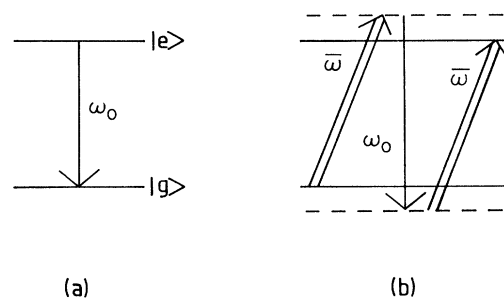

(a)   (b)

FIG. 1. Diagram (a) corresponds to ordinary spontaneous decay from the excited state to the ground state. During the decay, a photon with frequency ω_0 is emitted. In the process of diagram (b), the atom is initially in the ground state. First, it absorbs a photon with frequency $\bar{\omega}$ (double arrow), then it emits a fluorescent photon, and subsequently it absorbs a second photon with frequency $\bar{\omega}$. This process leaves the atom in the excited state, and from energy conservation it follows that the fluorescent photon must have frequency $2\bar{\omega} - \omega_0$.

$$\Gamma\sigma = \tfrac{1}{2}A_e(P_e\sigma + \sigma P_e - 2d^\dagger\sigma d)$$

$$+ \tfrac{1}{2}A_g(P_g\sigma + \sigma P_g - 2d\sigma d^\dagger) , \tag{4}$$

in terms of the projectors $P_e = |e\rangle\langle e|$, $P_g = |g\rangle\langle g|$, and the raising operator $d = |e\rangle\langle g|$. The two rate constants are $A_e = A(1 + |P|^2/2)$ and $A_g = A|P|^2/2$, where A is the Einstein coefficient for spontaneous decay in empty space and P is the Fresnel coefficient for reflection of a plane wave. The steady-state density operator $\bar{\rho}$ is the solution of

$$(L_a - i\Gamma)\bar{\rho} = 0 , \tag{5}$$

and is found to be

$$\bar{\rho} = \bar{n}_e P_e + \bar{n}_g P_g , \tag{6}$$

with

$$\bar{n}_e = \frac{|P|^2}{2(1+|P|^2)}, \quad \bar{n}_g = 1 - \bar{n}_e . \tag{7}$$

Then we calculate the exponential in Eq. (2), and substitute the result into Eq. (1). This yields

$$I(\omega) = \hbar\omega_0 I_p B(\bar{n}_g - \bar{n}_e)\frac{1}{\pi}\mathrm{Re}\frac{1}{\tfrac{1}{2}(A_e + A_g) - i(\omega - \omega_0)} , \tag{8}$$

where

$$B = \frac{\pi}{\epsilon_0\hbar^2 c}|\langle e|\boldsymbol{\mu}\cdot\boldsymbol{\epsilon}|g\rangle|^2 \tag{9}$$

is the Einstein coefficient for stimulated transitions.

The absorption profile $I(\omega)$ appears to be a Lorentzian at $\omega = \omega_0$ and with a halfwidth at half maximum equal to

$\tfrac{1}{2}(A_e + A_g) = \tfrac{1}{2}A(1 + |P|^2)$. This shows that the width increases linearly with the intensity-reflection coefficient $|P|^2$. The strength of the line is proportional to the population difference

$$\bar{n}_g - \bar{n}_e = \frac{1}{1+|P|^2} , \tag{10}$$

which decreases with increasing reflectivity. For an atom in empty space this would be $\bar{n}_g - \bar{n}_e = 1$, because the atom would be in the ground state. The term proportional to $\bar{n}_g$ correspond to stimulated absorptions, whereas the term proportional to $\bar{n}_e$ comes from stimulated emissions and counts therefore as negative. The peak value of the profile is

$$I(\omega_0) = \frac{2\hbar\omega_0 I_p B}{\pi A}\frac{1}{(1+|P|^2)^2} , \tag{11}$$

which decreases very rapidly with increasing $|P|^2$. This is due to the fact that the line broadens and loses strength for increasing values of the reflectivity. For very large values of $|P|^2$ the line disappears completely as a consequence of $\bar{n}_g \simeq \bar{n}_e$. Stimulated absorptions and emissions of probe-beam photons then occur at the same rate, which gives effectively $I(\omega) \simeq 0$. It should be experimentally feasible to measure the atomic absorption during the switching on of the PC (turning up the pump power). Any diminishing of the absorption with increasing reflectivity would then indicate a population of the excited state, which is necessarily brought about by a three-photon process.

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