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Thermal stimulation of cooperative two-photon decay in a microcavity

N. A. Enaki, M. A. Macovei

Institute of Applied Physics, Academy of Sciences of Moldova,
Academiei str. 5, Chisinau MD-2028, Moldova.

Abstract

The cooperative two-photon spontaneous decay of an excited atomic system in a microcavity is investigated. We demonstrate that the presence of a small number of thermalized photons in the microcavity mode stimulate the cooperative generation rate of the coherent entangled photon pairs.

It is well known that the spontaneous decay of a single atom in a microcavity differs substantially from decay in free space [1]. Such behavior of an atom inside a microcavity can be explained by the fact that near the transition frequency the rate of spontaneous decay is proportional to the EMF mode density.

The study of two-photon light generation has attracted much attention [2-4]. The first report of an experimental observation of two-photon coherent light generation in which excited *Li* atoms were used was made by Nikolaus *et al* [3]. Experiments involving Rydberg atoms have demonstrated the real possibility of building a two-photon micromaser [4]. Since in a microcavity the rate of two-photon decay in a three-level cascade system increases substantially, it is of interest to analyze in such a setting the cooperative emission of radiation by Rydberg atoms in microcavities.

In the present paper we study the cooperative decay of an ensemble of such atoms with a cascade pattern of the levels, where at a finite temperature the intermediate level is arbitrarily offset from resonance with a microcavity mode. In this case the intermediate level is essentially vacant, and the fact that it lies between the excited and ground states leads to a sizable increase in the two-photon cooperative transition amplitude. Using the statistical method of eliminating operators of the EMF and the virtual intermediate state of electrons in the atoms [2] one can obtain the following equation for an operator of the atomic subsystem, $O(t)$, in the process of two-photon spontaneous decay inside the microcavity.

$$\begin{aligned} \frac{d}{dt} \langle O(t) \rangle &= i\omega_{21} \sum_{j=1}^N \langle [R_{zj}, O(t)] \rangle \\ &+ \frac{1}{\hbar^4} \sum_{k_1 k_2} \sum_{j,l=1}^N \left[\frac{(\vec{g}_{k_1} \cdot \vec{d}_{31})(\vec{g}_{k_2} \cdot \vec{d}_{32})}{\omega_{23} - \omega_{k_2}} - \frac{(\vec{g}_{k_1} \cdot \vec{d}_{32})(\vec{g}_{k_2} \cdot \vec{d}_{31})}{\omega_{31} - \omega_{k_2}} \right]^2 \\ &\times \left[[1 + n(k_1) + n(k_2)] \left(e^{-i[(\vec{k}_1 + \vec{k}_2) \cdot (\vec{r}_j - \vec{r}_l)]} \frac{\langle R_l^\dagger [R_j^-, O(t)] \rangle}{i(\omega_{k_1} + \omega_{k_2} - \omega_{21} + 2i\Gamma)} + e^{i[(\vec{k}_1 + \vec{k}_2) \cdot (\vec{r}_j - \vec{r}_l)]} \right) \right] \end{aligned}$$

$$\begin{aligned} & \times \frac{\langle [R_j^\dagger, O(t)] R_l^- \rangle}{i(\omega_{k_1} + \omega_{k_2} - \omega_{21} - 2i\Gamma)} \Big) + n(k_1)n(k_2) \Big(e^{-i[(\vec{k}_1 + \vec{k}_2) \cdot (\vec{r}_j - \vec{r}_l)]} \frac{\langle [R_l^\dagger, [R_j^-, O(t)]] \rangle}{i(\omega_{k_1} + \omega_{k_2} - \omega_{21} + 2i\Gamma)} \\ & + e^{i[(\vec{k}_1 + \vec{k}_2) \cdot (\vec{r}_j - \vec{r}_l)]} \frac{\langle [[R_j^\dagger, O(t)], R_l^-] \rangle}{i(\omega_{k_1} + \omega_{k_2} - \omega_{21} - 2i\Gamma)} \Big) \Big], \end{aligned} \quad (1)$$

where the operators R_j^\pm and R_{zj} satisfy the usual commutation relations for spin operators. Here ω_{21} is the atomic transition frequency between the dipole-forbidden $|2\rangle$ and $|1\rangle$ states; $d_{3\beta}$ is the dipole moment of the transition between levels $|3\rangle$ and $|\beta\rangle$ ($\beta=1,2$); $\vec{g}_k = \sqrt{2\pi\hbar\omega_k/V}\vec{e}_\lambda$, with \vec{e}_λ the polarization vector of a photon ($\lambda = 1, 2$) of frequency ω_k and V the cavity volume. In deriving equation (1), we have considered the offset $\Delta = |\omega_k - \omega_{23}|$ ($\Delta = |\omega_k - \omega_{31}|$) much greater than the damping factor $\Gamma(k)$ of the microcavity in order to two-photon process can occur. Next we limit ourselves to one microcavity mode. When the number of atoms is large, the process of cooperative two-photon decay becomes appreciable stronger. If we ignore fluctuations in the number of particles when the number of atoms is large ($N \gg 1$), one can easily obtain for the atomic inversion operator

$$\frac{d}{dt}\langle R_z(t) \rangle = -\frac{1}{\tau^{(b)}}\langle R_z \rangle - \frac{1}{\tau_0^{(b)}}(j(j+1) - \langle R_z \rangle^2 + \langle R_z \rangle), \quad (2)$$

where $1/\tau_0^{(b)} = 4g^4 d_0^4 (1+2n)/(\Gamma\Delta^2\hbar^4)$, $1/\tau^{(b)} = 8g^4 d_0^4 n^2/(\Gamma\Delta^2\hbar^4)$ and $j = N/2$. The solution of this equation is

$$\langle R_z(t) \rangle = \frac{1+q}{2} - \frac{c}{2} \tanh\left[\frac{1}{2\tau_r}(t-t_0)\right],$$

where $\tau_r = \tau_0^{(b)}/c$ is the time of cooperative spontaneous decay of the ensemble of atoms, $t_0 = \tau_r \ln \frac{N-[1+q-c]}{[1+q+c]-N}$ is the time lag of the pulse of collective emission of a pair of photons in the microcavity, $q = \tau_0^{(b)}/\tau^{(b)}$, and $c = \sqrt{(1+q)^2 + 4j(j+1)}$. The Eq. (2) describing two-photon cooperative spontaneous decay implies that a thermalized field affects not only the Einstein coefficient $1/\tau^{(b)}$ corresponding to stimulated decay but also the rate of two-photon spontaneous decay, $1/\tau_0^{(b)}$. Clearly, a thermalized field facilitates the process of cooperative two-photon decay. This constitutes one of main difference between two-photon dipole-forbidden emission and one-photon cooperative spontaneous emission. Evidently, two-photon cooperative spontaneous emission prevails over stimulated thermalized transition only if $N(1+2n) > n^2$. These estimates suggest that when $n < 1$, the term $1/\tau^{(b)}$, which corresponds to induced decay, is negligible in comparison to the term $1/\tau_0^{(b)}$, which corresponds to spontaneous decay.

In the absence of radiators inside the microcavity, one can calculate the fluctuations of the electromagnetic field operators:

$$\delta_0^2 = \langle a^{\dagger 2} a^2 \rangle - \langle a^\dagger a \rangle^2 = n^2.$$

It would be very interesting to find the fluctuations in the number of photons of the electromagnetic field that are generated by the excited radiators in the process of two-photon emission in the microcavity. In doing this one introduce a function that accounts

for fluctuations of the electromagnetic field in relation to thermalized fluctuations:

$$\delta_r^2 = \delta^2 - \delta_0^2, \quad (3)$$

where $\delta^2 = \langle a^{\dagger 2}(t)a^2(t) \rangle - \langle a^\dagger(t)a(t) \rangle^2$. Since experiments often monitor the dynamics of the population difference of the atomic subsystem in the microcavity, we express the electromagnetic field fluctuations δ_r^2 in terms of the kinetics of atomic population inversion.

Let $G^{(1)}(t) = \langle a^\dagger(t)a(t) \rangle$ then

$$\frac{d}{dt}G^{(1)}(t) = \frac{d}{dt}\langle a^\dagger(t)a(t) \rangle = \left\langle \frac{da^\dagger(t)}{dt}a(t) + a^\dagger(t)\frac{da(t)}{dt} \right\rangle. \quad (4)$$

Eliminating the heat-bath operators, in the Born-Marcov approximation $\frac{d}{dt}G^{(1)}(t) \ll \Gamma G^{(1)}(t)$, one can represent the function $G^{(1)}(t)$ via the atomic inversion operator:

$$G^{(1)}(t) = n - \frac{1}{\Gamma} \frac{d}{dt} \langle R_z(t) \rangle. \quad (5)$$

For a large number of excited atoms in the Born-Marcov approximation $\frac{d}{dt}G^{(2)}(t) \ll \Gamma G^{(2)}(t)$ we find for the second-order correlation function $G^{(2)}(t) = \langle a^{\dagger 2}(t)a^2(t) \rangle$:

$$G^{(2)}(t) = 2n^2 - [3n + 1/2] \frac{1}{\Gamma} \frac{d}{dt} \langle R_z(t) \rangle + \left[\frac{1}{\Gamma} \frac{d}{dt} \langle R_z(t) \rangle \right]^2. \quad (6)$$

One can observe that as the two-photon absorption probability $w \sim \langle a^{\dagger 2}a^2 \rangle = G^{(2)}(t)$, at low-temperatures it is proportional to the two-photon flux Φ :

$$w \sim d/dt \langle R_z(t) \rangle \sim \Phi.$$

We note that the probability w also depends on the square of the two-photon flux, but this dependence is ignored in our approximation. Note that for one-photon superradiance, the function $G^{(2)}(t)$ is proportional to the square of the one-photon flux, or Φ^2 . This occurs because $a^\dagger \sim R^+$ for one-photon emission, while for two-photon emission we have $a^{\dagger 2} \sim R^{+}$. Hence

$$w \sim \langle R^{+2}R^{-2} \rangle \sim \langle [d/dt R_z]^2 \rangle$$

for one-photon superradiance, and

$$w \sim \langle R^+R^- \rangle \sim d/dt \langle R_z \rangle$$

for two-photon superradiance.

Now we can easily derive a formula for the relative fluctuations of the electromagnetic field inside the microcavity:

$$\delta_r^2 = -(n + 1/2) \frac{1}{\Gamma} \frac{d}{dt} \langle R_z(t) \rangle. \quad (7)$$

This implies that in each decay event, photons are generated in pairs and the emission intensity becomes proportional to N^2 , while the second-order correlation function for the photons remains much greater than the square of the first-order correlation function. In this case, at low temperatures and for large numbers of atoms, we can speak of photon superbunching, i.e. $\delta_r^2/\delta_0^2 \gg 1$.

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