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Two-Photon Population Inversion by Squeezed Light in a Fabry-Perot Microcavity.

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Abstract. – A three-level atom interacting with a squeezed-vacuum field is driven into a coherent superposition of the ground state and upper excited state, which can lead to a two-photon population inversion. We describe a possible technique for testing this prediction, using a microcavity Fabry-Perot interferometer. This could provide a useful technique for pumping a two-photon laser.

The availability of tunable squeezed radiation sources leads to the possibility of many novel effects and techniques in quantum optics and atomic spectroscopy. New phenomena predicted in the interaction of squeezed fields with atomic systems include the inhibition of atomic phase decays [1], subnatural linewidth spectroscopy [2], linear rather than quadratic dependence on the intensity of the two-photon absorption rate [3], and pairwise excitation of atomic states [4]. To date, these predictions have not been experimentally tested. The reason for this is that some of the effects are rather subtle. For example, there is no change due to squeezed correlations in the atomic population when a two-level medium interacts with squeezed radiation. There are changes in the polarization, but this can only be indirectly detected through changes in the fluorescent spectrum, and this is difficult to measure even if microcavity techniques are used.

More recently, the dynamical properties of a three-level atom in a squeezed vacuum have been calculated [5, 6]. The results have shown that in a three-level atom in the cascade configuration coupled only to squeezed modes, there are spectacular qualitative changes in the steady-state level populations relative to ordinary laser spectroscopy, including two-photon population inversions. These are much more readily observable than fluorescent spectra. The effects, however, are sensitive to the number of squeezed modes coupled to the atom. Present sources of squeezed light can couple to only a small fraction of the modes enveloping the atom. The population inversion, proportional to the solid angle subtended, would be so small as to be practically unobservable in an atom located in a free-space squeezed vacuum.

In this letter we present for the first time a readily testable prediction of squeezed-light spectroscopy. Instead of a free-space squeezed vacuum [5, 6], we propose using an atomic beam transiting slowly through a plane-parallel Fabry-Perot microcavity irradiated with a

Gaussian squeezed beam. This is precisely the output produced in recently developed tunable single-mode parametric oscillators [7]. Since the microcavity enhances the coupling to modes orthogonal to the mirrors, we require only the Gaussian beam to be in a squeezed state. By exciting two-photon transitions in the atomic beam, unique signatures of squeezed-light spectroscopy will be impressed on the atomic populations, including two-photon inversions. This is not possible in two-level spectroscopy.

We consider a three-level atom with the state $|1\rangle$, $|2\rangle$ and $|3\rangle$ in the ladder configuration ($E_3 > E_2 > E_1$), located inside a microscopic Fabry-Perot cavity, and coupled to a three-dimensional electromagnetic field. The transition frequencies of $|2\rangle \to |1\rangle$ and $|3\rangle \to |2\rangle$ are ω_1 and ω_2, respectively, with $\omega_1 - \omega_2 = \Delta$. The corresponding natural linewidths are γ_1 and γ_2. The cavity consists of two plane mirrors which lie parallel to the (x, y)-plane (fig. 1). The first is a perfectly reflecting mirror located at $z = 0$, and the second is a partially transmitting lossless mirror with a real reflectivity R, located at $z = -L$. The cavity is driven by a squeezed Gaussian beam which is propagating in the z-direction normal to the mirrors, as in fig. 1. We assume that this multimode squeezed-vacuum field input has a wide bandwidth compared to γ_1, γ_2 and Δ. The field can be expressed in terms of the asymptotic free-field plane-wave input operators $a_{\text{in}}(\boldsymbol{k}, s)$ as

$$A(k) = \sum_s \int \mathrm{d}\Omega_k k U_s^*(\boldsymbol{k})\, a_{\text{in}}(\boldsymbol{k}, s), \tag{1}$$

with

$$\sum_s \int \mathrm{d}\Omega_k |U_s(\boldsymbol{k})|^2 = 1, \tag{2}$$

where $U_s(\boldsymbol{k})$ are amplitudes of the input squeezed field, and s is the polarization index ($s = 1, 2$).

For sufficiently wide-band fields, we can assume a delta-correlation in frequency, which we label using the corresponding wave number, $k = \omega/c$. Thus

$$\begin{cases} \langle A^\dagger(k) A(k')\rangle = N(k)\,\delta(k - k'), \\ \langle A(k) A(k')\rangle = M(k)\,\delta(2k_0 - k - k'), \end{cases} \tag{3}$$

where $k_0 = \omega_0/c$, and ω_0 is the carrier frequency of the squeezed field. The parameters N and $M = |M| \exp[i\varphi_s]$, which appear in eq. (3), are measures of the squeezing such that $|M|^2 \leqslant$ $\leqslant N(N+1)$. Here the equality holds for a minimum-uncertainty squeezed state, and φ_s is the phase of the squeezed field. From eqs. (1)-(3) we find the correlation relations for the

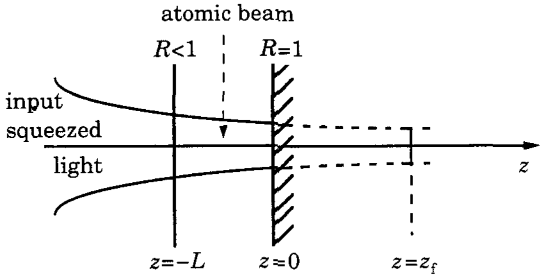

Fig. 1. – Geometry of the microscopic Fabry-Perot cavity with the input squeezed light focused at a point z_f.

free-field plane-wave operators as

$$\begin{cases} \langle a_{\text{in}}^{\dagger}(\boldsymbol{k}, s)\, a_{\text{in}}(\boldsymbol{k}', s')\rangle = N(k)\, U_s^*(\boldsymbol{k})\, U_{s'}(\boldsymbol{k}')\, \delta(k - k')/k^2 , \\ \langle a_{\text{in}}(\boldsymbol{k}, s)\, a_{\text{in}}(\boldsymbol{k}', s')\rangle = M(k)\, U_s(\boldsymbol{k})\, U_{s'}(\boldsymbol{k}')\, \delta(2k_0 - k - k')/kk' . \end{cases} \tag{4}$$

By a consideration of boundary conditions at the mirrors [8], we derive the following expression for the electric field inside the cavity, in the Heisenberg picture:

$$\boldsymbol{E}^{(+)}(\boldsymbol{r}, t) = i \sum_s \int \mathrm{d}^3\boldsymbol{k} \left(\frac{2ck}{\varepsilon_0 \hbar (2\pi)^3} \right)^{1/2} Y(\theta_k, L)\, \hat{e}_{\boldsymbol{k}s}^{\parallel} \exp[i\boldsymbol{k}^{\parallel}\cdot\boldsymbol{r}] \sin(k_z r)\, a_{\text{in}}(\boldsymbol{k}, s), \tag{5}$$

where $Y(\theta_k, L)$ is the cavity transfer function

$$Y(\theta_k, L) = \frac{i(1 - R^2)^{1/2}}{1 - R \exp[2ikL \cos\theta_k]} , \tag{6}$$

with θ_k the angle between the $\boldsymbol{k}$ vector and the z-axis. In eq. (5), $\hat{e}_{\boldsymbol{k}s}^{\parallel}$ and $\boldsymbol{k}^{\parallel}$ are projections onto the (x, y)-plane of the polarization vectors $\hat{e}_{\boldsymbol{k}s}$ and the wave vector $\boldsymbol{k}$, respectively.

The effect of $Y(\theta_k, L)$ on the electric field inside the cavity is most clearly exhibited in the form $|Y(\theta_k, L)|^2$, which is identified as the Airy function of the cavity [8]. If R is close to 1 and $L = \lambda/2$, the function $|Y(\theta_k, L)|^2$ will exhibit a sharp peak centred at $\cos\theta_k = 1$. This means that the field inside the cavity is affected only by those modes which are contained in a small solid angle around the z-axis.

The questions we are interested in concern the final state of the atom and any two-photon population inversions. To answer these questions we apply the master equation for a three-level atom in a wide-band squeezed field [5]. We note that the spectral filtering of microcavities is typically too broadband to reduce the input bandwidth, and hence the Markovian approximation can still be used here. It is not difficult to show that the stationary density matrix has the following form:

$$\rho_{ss} = \begin{pmatrix} \rho_{11} & 0 & \rho_{13} \\ 0 & \rho_{22} & 0 \\ \rho_{31} & 0 & \rho_{33} \end{pmatrix}, \tag{7}$$

where ρ_{ij} are the steady-state values of the density matrix elements and are given by

$$\begin{cases} \rho_{22} = (N_1 + N_2 + \Gamma_2)[N_1(N_2 + \Gamma_2) - |\eta_{12}|^2 |M|^2]/D , \\ \rho_{33} = [N_1 N_2 (N_1 + N_2 + \Gamma_2) - (N_1 + N_2 - \Gamma_1)|\eta_{12}|^2 |M|^2]/D , \\ \rho_{11} = 1 - \rho_{22} - \rho_{33} , \\ \rho_{13} = \rho_{31}^* = \eta_{12}^* M(\Gamma_1 N_2 - \Gamma_2 N_1 + \Gamma_1 \Gamma_2)/D , \end{cases} \tag{8}$$

with

$$D = (N_1 + N_2 + \Gamma_2)(3N_1N_2 + 2N_1\Gamma_2 + \Gamma_1 N_2 + \Gamma_1\Gamma_2 - 3|\eta_{12}|^2 |M|^2) , \tag{9}$$

and

$$N_i = \beta_i N \qquad (i = 1, 2) .$$

Here the damping rates Γ_i corresponding to a normal vacuum are given by

$$\Gamma_i = \frac{3}{2}\,\gamma_i\,\frac{(1+R)}{(1-R)}\int_0^1 du(1+u^2)\,\frac{\sin^2(kr_z u)}{1+F\sin^2(kLu)}\,, \tag{10}$$

where $u = \cos\theta_k$, $r = (r_x, 0, r_z)$ is a coordinate of the atom, and $F = 4R/(1-R)^2$. In the limit $R \to 1$, with $r_z = L/2$ and $L = \lambda/2$, Γ_i approaches $1.5\gamma_i$, which is in agreement with the result found by Milonni and Knight [9] for a two-level atom inside a cavity, as tested in experiments by Hulet *et al.* [10]. This result shows that the spontaneous-emission rate is three times greater than the free-space rate, when the atom is midway between perfect plane parallel mirrors separated by half a wavelength.

The parameters γ_{12} and β_i $(i = 1, 2)$ are new constants introduced by the squeezed vacuum. These depend on the matching of the internal-cavity modes given by the cavity transfer functions $Y(\theta_k, L)$ to the input squeezed modes given by the functions $U_s(\boldsymbol{k})$. We define the functions $U_s(\boldsymbol{k})$ in the case of a Gaussian polarisation matched beam as [11]

$$U_s(\boldsymbol{k}) = \mathcal{N}^{-1/2}(\boldsymbol{\mu}_i^* \cdot \hat{e}_{ks}^{\parallel *})\exp\left[-\frac{1}{4}k^2 W_0^2 \sin^2\theta_k - ikz_{\mathrm{f}}\cos\theta_k\right], \tag{11}$$

where $U_s(\boldsymbol{k})$ is the Fourier transform of a Gaussian input beam, focused at $z = z_{\mathrm{f}}$, with waist-size W_0, and $\mathcal{N}$ is a normalisation constant.

The following expressions for the damping coefficients γ_{12}, β_i are obtained in this case:

$$\gamma_{12} = \frac{3}{2}(\gamma_1\gamma_2)^{1/2}\,\frac{(1+R)}{(1-R)^3}\,\frac{1}{\mathcal{N}}\,B^2(r_x)\,, \qquad \beta_i = \frac{3}{2}\,\gamma_i\,\frac{(1+R)}{(1-R)^3}\,\frac{1}{\mathcal{N}}\,|B(r_x)|^2\,, \tag{12}$$

where

$$B(r_x) = \int_0^1 du(1+u^2)\,\frac{\sin(kr_z u)}{1+F\sin^2(kLu)}\,J_0(kr_x(1-u^2)^{1/2})\cdot$$

$$\cdot[1 - R\cos(2kLu) + iR\sin(2kLu)]\exp\left[-\frac{1}{4}k^2W_0^2(1-u^2) - ikz_{\mathrm{f}}u\right]. \tag{13}$$

Here J_0 is the zeroth-order Bessel function, and the normalisation constant is

$$\mathcal{N} = \int_0^1 du(1+u^2)\exp\left[-\frac{1}{2}k^2W_0^2(1-u^2)\right]. \tag{14}$$

Using the above equations for the damping coefficients we can calculate the steady-state populations of the atomic levels. It is evident from eq. (7) that in the squeezed vacuum the density matrix is not diagonal due to the presence of coherences ρ_{13} and ρ_{31}. In this case the states $|1\rangle$, $|2\rangle$, and $|3\rangle$ are no longer eigenstates of the system. The density matrix (7) can

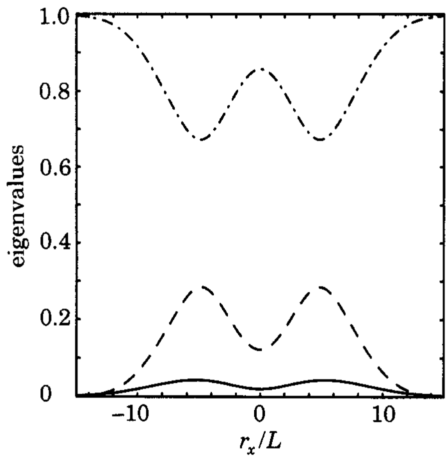

Fig. 2.

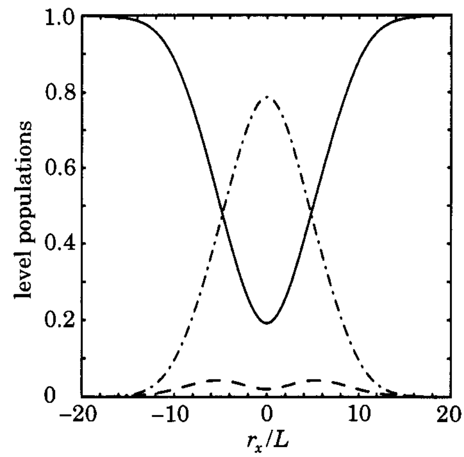

Fig. 3.

Fig. 2. – Eigenvalues P_2 (solid line), P_- (dashed line), and P_+ (dash-dotted line) as a function of r_x/L for $R = 0.99$, $\gamma_2 = 0.01\gamma_1$, $z_f = 25\lambda$, $r_z = L/2 = \lambda/4$, and $W_0 = 3.0\lambda$.

Fig. 3. – Same as in fig. 2 but for the steady-state population of the atomic states: $|1\rangle$ (solid line), $|2\rangle$ (dashed line), and $|3\rangle$ (dash-dotted line).

be rediagonalised by including ρ_{13} and ρ_{31} to give the new «dressed» states $|\psi_\pm\rangle$, $|\psi_2\rangle$ where

$$\left\{\begin{array}{l} |\psi_+\rangle = [(P_+ - \rho_{33})|1\rangle + \rho_{31}|3\rangle]/[(P_+ - \rho_{33})^2 + |\rho_{13}|^2]^{1/2}, \\ |\psi_-\rangle = [\rho_{13}|1\rangle + (P_- - \rho_{11})|3\rangle]/[(P_- - \rho_{11})^2 + |\rho_{13}|^2]^{1/2}, \\ |\psi_2\rangle = |2\rangle. \end{array}\right. \tag{15}$$

In the new basis, the diagonal probabilities $P_\pm$, P_2 are

$$\left\{\begin{array}{l} P_\pm = \dfrac{1}{2}(\rho_{11} + \rho_{33}) \pm \dfrac{1}{2}[(\rho_{11} - \rho_{33})^2 + 4|\rho_{13}|^2]^{1/2}, \\ P_2 = \rho_{22}. \end{array}\right. \tag{16}$$

Thus, the squeezed vacuum causes the system to decay into new states $|\psi_\pm\rangle$, which are linear combinations of the states $|3\rangle$ and $|1\rangle$. The population of $|3\rangle$ is optimised when the ratio γ_1/γ_2 is increased. Figure 2 shows the eigenvalues P_+, P_- and P_2 as a function of r_x/L with $R = 0.99$, $z_f = 25\lambda$, $r_z = L/2 = \lambda/4$, $\gamma_2 = 0.01\gamma_1$, $N = 0.2$, and $W_0 = 3.5\lambda$. The values of the parameters W_0 and z_f maximise the amplitude and phase matching between the cavity and input squeezed modes for this values of the reflectivity R. The figure demonstrates that the atom is driven into coherent superpositions of the ground and the most excited states, and there is a small probability that the atom is in the state $|2\rangle$. We illustrate in fig. 3 the populations of the atomic states $|1\rangle$, $|2\rangle$, and $|3\rangle$. It is seen that there is a population inversion between the states $|3\rangle$ and $|2\rangle$. For $r_x/L \approx 0$, there is also a two-photon population inversion between the states $|3\rangle$ and $|1\rangle$. An atom located inside an optical cavity and driven by a squeezed field can reach a two-photon population inversion of 78% of the maximum for $R = 0.99$ and $\gamma_2/\gamma_1 = 0.01$.

In conclusion, this letter treats a wide-band squeezed Gaussian input to a three-level atom located inside an optical microcavity. The results violate predictions on the population distribution given by rate equation treatments of the atomic states. This effect is caused by two-photon coherences introduced by the squeezed vacuum, which create coherent superpositions of the atomic states. These superpositions can lead not only to a one-photon population inversion, but also to a two-photon population inversion, when the second transition has a lower decay rate than the first. This could provide a useful technique for pumping a two-photon laser. More significantly, it indicates fundamental difference between conventional and squeezed-radiation spectroscopy, which become easily observable in multiphoton transitions excited in microcavity geometries.

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