Theory of collective Raman scattering from a Bose-Einstein condensate

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Recent experiments have demonstrated superradiant Raman scattering from a Bose-Einstein condensate driven by a single off-resonant laser beam. We present a quantum theory describing this phenomenon, showing Raman amplification of matter wave due to collective atomic recoil from 3-level atoms in a Λ-configuration. When atoms are initially in a single lower internal state, a closed two-level system is realized between atoms with different internal states, and entangled atom-photon pairs can be generated. When atoms are initially prepared in both the lower internal states, a fraction of atoms recoiling in the backward direction can be generated.

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Important progress in the study of the coherent interaction between atoms and photons have been recently obtained using Bose-Einstein condensates (BEC) of low-density alkali atoms [1]. In the case where the atoms interact only with far off-resonant optical fields, the dominant atom-photon interaction is two-photon Rayleigh scattering. In this situation, collective atomic recoil lasing (CARL) [2, 3, 4, 5] causes exponential enhancement of the number of scattered photons and atoms. Experimentally, CARL from a BEC has been observed so far in the Superradiant regime [6, 7, 8, 9], in which photons are scattered into the end-fire modes along the major axis of an elongated condensate. In these experiments, the atoms after the collective scattering remain in the original internal, gaining a recoil momentum $\hbar(\vec{k}_2-\vec{k}_1)$, where $\vec{k}_2$ and $\vec{k}_1$ are the wave vectors of the pump and scattered photons, respectively. The scattered atoms may experience further collective scattering, leading to the observed superradiant cascade [6].

In two recent experiments [10, 11] it has been observed superradiant Raman scattering, in which the atoms remain, after the process, in a different hyperfine state not resonant with the pump laser beam. As a consequence, no further scattering of pump photons occurs. In this Brief Report, we present a theory of the collective atomic recoil lasing from a 3-level atomic BEC which describes the observed phenomena. In particular, the theory demonstrates that maximum atom-photon entanglement can be generated in this system.

We consider a cloud of BEC atoms which have three internal states labeled by $|b\rangle$, $|c\rangle$, and $|e\rangle$ with energies $E_b<E_c<E_e$, respectively. The two lower states $|b\rangle$ and $|c\rangle$ can be hyperfine states in each of which the atoms can live for a long time. They are coupled to the upper state $|e\rangle$ via, respectively, a classical pump field and a quantized probe field of frequencies ω_2 and ω_1 in the Λ-configuration. The interaction scheme is shown in Fig. 1.

The second quantized Hamiltonian to describe the system at zero temperature is given by

$$\hat{H}=\hat{H}_{atom}+\hat{H}_{atom-field}, \tag{1}$$

where $\hat{H}_{atom}$ gives the free evolution of the the atomic fields and $\hat{H}_{atom-field}$ describes the dipole interactions between the atomic fields and the pump and probe fields. We assume the condensate to be sufficiently diluite in order to neglect the atom-atom interaction. The free atomic Hamiltonian is given by

$$\hat{H}_{atom}=\sum_{\alpha=b,c,e}\int d^3x\hat{\psi}^\dagger_\alpha(\vec{x},t)\left[-\frac{\hbar^2}{2m}\nabla^2\right]\hat{\psi}_\alpha(\vec{x},t), \tag{2}$$

where $\hat{\psi}_\alpha(\vec{x},t)$ and $\hat{\psi}^\dagger_\alpha(\vec{x},t)$ are the boson annihilation and creation operators in the interaction picture for the $|\alpha\rangle$-state atoms at position $\vec{x}$, respectively. They satisfy the standard boson commutation relation $[\hat{\psi}_\alpha(\vec{x},t),\hat{\psi}^\dagger_\beta(\vec{x}',t)]=\delta_{\alpha\beta}\delta(\vec{x}-\vec{x}')$ and $[\hat{\psi}_\alpha(\vec{x},t),\hat{\psi}_\beta(\vec{x}',t)]=[\hat{\psi}^\dagger_\alpha(\vec{x},t),\hat{\psi}^\dagger_\beta(\vec{x}',t)]=0$.

The atom-laser interaction in the dipole approximation is described by the following Hamiltonian

$$\hat{H}_{atom-field}=-\hbar\int d^3x\left[\frac{1}{2}\Omega\hat{\psi}^\dagger_e(\vec{x},t)\hat{\psi}_b(\vec{x},t)e^{i(\vec{k}_2\cdot\vec{x}-\Delta_2 t)}\right.$$
$$\left.+g_1\hat{a}_1(t)\hat{\psi}^\dagger_e(\vec{x},t)\hat{\psi}_c(\vec{x},t)e^{i(\vec{k}_1\cdot\vec{x}-\Delta_1 t)}+H.c.\right], \tag{3}$$

where $\omega_{b,c}=(E_e-E_{b,c})/\hbar$ are the resonant frequencies for the two atomic transitions, $\Delta_2=\omega_2-\omega_b$, $\Delta_1=\omega_1-\omega_c$, $g_1=\mu_{ce}\mathcal{E}_1/\hbar$ and $\Omega=\mu_{be}E_2/\hbar$ with $\mu_{\alpha\beta}$ denoting a transition dipole-matrix element between states $|\alpha\rangle$ and $|\beta\rangle$, $\mathcal{E}_1=\sqrt{\hbar\omega_1/2\epsilon_0 V}$ being the electric field per photon for the quantized probe field of frequency ω_1 in a mode volume V, and E_2 being the amplitude of the electric field for the classical pump laser beam of frequency ω_2. Finally, $\hat{a}^\dagger_1(t)$ and

$\hat{a}_1(t)$ are photon creation and annihilation operators for the probe field, satisfying the boson commutation relation $[\hat{a}_1(t), \hat{a}_1^\dagger(t)] = 1$.

We consider the case where the pump laser is detuned far enough away from the atomic resonance that the excited state population remains negligible, a condition which requires that $\Delta_2 \gg \gamma_e$, where γ_e is the natural width of the atomic transition between the excited state $|e\rangle$ and the hyperfine ground state $|b\rangle$. In this regime the atomic polarization adiabatically follows the ground state population, allowing the formal elimination of the excited state atomic field operator. Writing the Heisenberg equation for $\hat{\psi}_e \exp[i(\vec{k}_2\cdot\vec{x} - \Delta_2 t)]$ and dropping the kinetic term, we obtain

$$\hat{\psi}_e(\vec{x},t) \approx -\frac{1}{\Delta_2}\left\{\frac{1}{2}\Omega\hat{\psi}_b(\vec{x},t) + g_1\hat{a}_1(t)\hat{\psi}_c(\vec{x},t)e^{-i\theta+i\delta t}\right\}e^{i(\vec{k}_2\cdot\vec{x}-\Delta_2 t)} \tag{4}$$

where $\theta = (\vec{k}_2 - \vec{k}_1)\cdot\vec{x}$ and $\delta = \Delta_2 - \Delta_1 = \omega_2 - \omega_1 - \Delta_{cb}$, with $\Delta_{cb} = (E_c - E_b)/\hbar$. Substituting Eq.(4) into Eq.(3) and neglecting the small light shifts proportional to $|\Omega|^2$ and $g_1^2\hat{a}_1^\dagger\hat{a}_1$, we arrive at the following effective Hamiltonian:

$$\hat{H} = \sum_{\alpha=b,c}\int d^3x\hat{\psi}_\alpha^\dagger(\vec{x},t)\left[-\frac{\hbar^2}{2m}\nabla^2\right]\hat{\psi}_\alpha(\vec{x},t) + i\hbar g\int d^3x\left[\hat{a}^\dagger\hat{\psi}_b(\vec{x},t)\hat{\psi}_c^\dagger(\vec{x},t)e^{i\theta} - \text{H.c.}\right] - \hbar\delta\hat{a}^\dagger\hat{a}, \tag{5}$$

where $g = g_1\Omega/2\Delta_2$ and $\hat{a} = i\hat{a}_1 e^{i\delta t}$.

Neglecting shape effects due to the finite size of the condensate, we can perform the expansion on momentum eigenstates [5]:

$$\hat{\psi}_b = \mathcal{C}\sum_{n=-\infty}^{+\infty}\hat{b}_n e^{in\theta} \qquad \hat{\psi}_c = \mathcal{C}\sum_{n=-\infty}^{+\infty}\hat{c}_n e^{in\theta} \tag{6}$$

where $[\hat{c}_n, \hat{c}_m^\dagger] = \delta_{n,m}$, $[\hat{b}_n, \hat{b}_m^\dagger] = \delta_{n,m}$, $[\hat{b}_n, \hat{c}_m] = [\hat{b}_n, \hat{c}_m^\dagger] = 0$ and $\mathcal{C}$ is a normalization constant. Substituting Eqs.(6) into Eq.(5), the Hamiltonian becomes:

$$\hat{H} = \sum_{n=-\infty}^{+\infty}\{\hbar\omega_r n^2(\hat{b}_n^\dagger\hat{b}_n + \hat{c}_n^\dagger\hat{c}_n) + i\hbar g(\hat{a}^\dagger\hat{c}_n^\dagger\hat{b}_{n-1} - \text{H.c.})\} - \hbar\delta\hat{a}^\dagger\hat{a}. \tag{7}$$

and the Heisenberg equations for $\hat{b}_n$, $\hat{c}_n$ and $\hat{a}$ are

$$\frac{d\hat{b}_n}{dt} = -i\omega_r n^2\hat{b}_n - g\hat{a}\hat{c}_{n+1} \tag{8}$$

$$\frac{d\hat{c}_n}{dt} = -i\omega_r n^2\hat{c}_n + g\hat{a}^\dagger\hat{b}_{n-1} \tag{9}$$

$$\frac{d\hat{a}}{dt} = i\delta\hat{a} + g\sum_n\hat{b}_n\hat{c}_{n+1}^\dagger, \tag{10}$$

where $\omega_r = \hbar q^2/2m$ is the recoil frequency and $\hbar\vec{q} = \hbar(\vec{k}_2 - \vec{k}_1)$ is the photon recoil momentum. In Eqs.(6), $\hat{b}_n$ and $\hat{c}_n$ are annihilation operators for the modes $|b,n\rangle$ and $|c,n\rangle$, corresponding to atoms in the internal state $|b\rangle$ and $|c\rangle$, respectively, and with momentum $\vec{p} = n\hbar\vec{q}$. Notice that Eqs.(8)-(10) conserve the total number of atoms N, i.e. $\sum_n\{\hat{b}_n^\dagger\hat{b}_n + \hat{c}_n^\dagger\hat{c}_n\} = \hat{N}$, and the total momentum $\hat{Q} = \hat{a}^\dagger\hat{a} + \sum_n n\{\hat{b}_n^\dagger\hat{b}_n + \hat{c}_n^\dagger\hat{c}_n\}$. Furthermore, the number of atoms in the subsystem $\mathcal{C}_n = \{|b,n\rangle, |c,n+1\rangle\}$ is also conserved, i.e. $\hat{b}_n^\dagger\hat{b}_n + \hat{c}_{n+1}^\dagger\hat{c}_{n+1} = \hat{N}_n$ is a constant for every n. This means that each subsystem $\mathcal{C}_n = \{|b,n\rangle, |c,n+1\rangle\}$ is closed. However, atoms belonging to different $\mathcal{C}_n$ are coupled by the common radiation field $\hat{a}$.

The system of Eqs.(8)-(10) describes the two-photon Raman scattering, in which an atom is transferred from the state $|b,n\rangle$ to the state $|c,n+1\rangle$ when it scatters a photon from the pump to the probe, i.e. when it "emits" a probe photon, whereas the atom is transferred from the state $|c,n\rangle$ to the state $|b,n-1\rangle$ when it scatters a photon from the probe to the pump, i.e. when it "absorbs" a probe photon. The main difference with respect to the normal CARL regime is that after emission of a probe photon the atom changes the internal state from $|b\rangle$ to $|c\rangle$. In particular, if atoms are initially in the internal state $|b\rangle$, they can only emit probe photons. As a consequence, in the Superradiant regime, in which emission dominates over absorbtion, atoms are transferred from the initial state $|b,0\rangle$ to the final state $|c,1\rangle$, where they can not anymore emit probe photons, experiencing subsequent superradiant scattering. Hence, when atoms are initially in the state $|b,0\rangle$, the condensate behaves as a closed two-level system.

In the linear regime where $N_{c1} \ll N_b$, where N_b and N_{c1} are the number of atoms in the initial state $|b,0\rangle$ and in the recoiling state $|c,1\rangle$, we may assume $\hat{b}_0 \approx \sqrt{N_b}$ and the Hamiltonian (7) reduces to:

$$\hat{H}_{eff} = \hbar\omega_r \hat{c}_1^\dagger \hat{c}_1 + i\hbar g\sqrt{N_b}(\hat{a}^\dagger \hat{c}_1^\dagger - \hat{a}\hat{c}_1) - \hbar\delta \hat{a}^\dagger \hat{a}. \tag{11}$$

This means that we are investigating a system which is analogous to the non-degenerate optical parametric amplifier (OPA) [12, 13] and involves the generation of correlated atom-photon pairs. The evolved state at time t is a pure bipartite state

$$|\psi\rangle = \frac{1}{\sqrt{1+\langle \hat{n}_c\rangle}} \sum_{n=0}^{\infty} \left(\frac{\langle \hat{n}_a\rangle}{1+\langle \hat{n}_c\rangle}\right)^{n/2} e^{in\phi}|n,n\rangle, \tag{12}$$

where $\langle \hat{n}_a\rangle = \langle \hat{a}^\dagger \hat{a}\rangle$ and $\langle \hat{n}_c\rangle = \langle \hat{c}_1^\dagger \hat{c}_1\rangle$. Eq.(12) shows maximal entanglement between atoms and photons (according to the excess von Neumann entropy criterion [14]) and has the same form of the twin-beam state of radiation generated from an OPA and used to realize continuous variable optical teleportation [16]. The idea of using Raman-scattering from an optically driven BEC as a source of atom-photon pairs was originally proposed by Moore and Meystre [15], however without exploiting the amplification CARL process. In the ordinary quantum CARL a detailed theory for the interaction of quantized atomic and optical fields in the linear regime has been developed, with emphasis on the manipulation and control of their quantum statistics and the generation of quantum correlations and entanglement between matter and light waves [3, 4, 17] . From such model it results that, in the linear regime, the quantum CARL Hamiltonian reduces to that for three coupled modes, the first two modes corresponding to atoms having lost or gained a quantum recoil momentum $\hbar\vec{q}$ in the two-photon Bragg scattering between the pump and the probe, and the third mode corresponding to the photons of the probe field. Starting from vacuum, the state at a given time is a fully inseparable three mode state [17]. For certain values of the parameters the state has the same form of Eq. (12), but in general the presence of a third mode reduces the entanglement between the other two modes. In the present work we have shown that the collective atomic recoil lasing from a 3-level atomic BEC can be a more useful source for the production of the atom-photon entanglement and its application [18]

An other potentially interesting situation is when atoms initially occupy both the two ground states, $|b,0\rangle$ and $|c,0\rangle$, so that the resulting dynamics is that of a pair of two-level systems coupled by the radiation field. In fact, if atoms are initially present in $|c,0\rangle$, photons emitted spontaneously by the transition from $|b,0\rangle$ to $|c,1\rangle$ may drive the other transition between $|c,0\rangle$ and $|b,-1\rangle$, although detuned by $2\omega_r$ from resonance. However, if the number of emitted photons is large enough, a fraction of atoms with momentum $-\hbar\vec{q}$ will be produced. In the following we discuss in details this effect using parameters close to those of ref. [10].

Taking into account only the four atomic states $\{|b,0\rangle, |c,0\rangle, |b,1\rangle, |c,-1\rangle\}$ and treating the bosonic operator as c-numbers, we can derive from Eq.(8)-(10), the following system of equations:

$$\frac{dS_{1,2}}{dt} = -i(\delta \mp \omega_r)S_{1,2} + gAW_{1,2} - \gamma_{1,2}S_{1,2} \tag{13}$$

$$\frac{dW_{1,2}}{dt} = -2g(AS_{1,2}^* + \text{c.c}) \tag{14}$$

$$\frac{dA}{dt} = gN_b(S_1 + S_2) - \kappa A \tag{15}$$

where $S_1 = (b_0 c_1^*/N_b)\exp(-i\delta t)$, $S_2 = (b_{-1}c_0^*/N_b)\exp(-i\delta t)$ $W_1 = (|b_0|^2 - |c_1|^2)/N_b$, $W_2 = (|b_{-1}|^2 - |c_0|^2)/N_b$, $A = ae^{-i\delta t}$ and N_b is the number of atoms initially in the state $|b,0\rangle$. To Eqs.(13) we have added a damping term $-\gamma_{1,2}S_{1,2}$ taking into account for the coherence decay observed experimentally. Also, we have added to Eq.(15) a damping term $-\kappa A$ modelling, in a "mean-field" theory [19], radiation loss, where $\kappa = cT/2L$ if the radiation is circulating in a ring cavity (where T is the mirror transmittivity and L is the cavity length). In the free-space case, i.e. without optical cavity, $T = 1$ and L is of the order of the condensate length. In the superradiant regime, for $\kappa \gg g\sqrt{N_b}$ and $t \gg \kappa^{-1}$, we can adiabatically eliminate the radiation field. Assuming $\delta = \omega_r$ and $\kappa \gg \omega_r$, Eq.(15) gives $A \approx (gN_b/\kappa)(S_1 + S_2)$, which, when substituted in Eqs.(13) and (14), yields:

$$\frac{dS_1}{dt} = -\gamma_1 S_1 + (G/2)W_1(S_1 + S_2) \tag{16}$$

$$\frac{dW_1}{dt} = -G\left[2|S_1|^2 + (S_1S_2^* + \text{c.c})\right] \tag{17}$$

$$\frac{dS_2}{dt} = -(\gamma_2 + 2i\omega_r)S_2 + (G/2)W_2(S_1 + S_2) \tag{18}$$

$$\frac{dW_2}{dt} = -G\left[2|S_2|^2 + (S_1S_2^* + \text{c.c})\right], \tag{19}$$

where $G = 2g^2N_b/\kappa$ is the superradiant gain. If the number N_c of atoms initially in the state $|c,0\rangle$ is zero, then $S_2 = 0$ and the solution of Eqs.(16) and (17) yields the well-known hyperbolic tangent shape for the Superradiant decay of the fraction of atoms $P_b = |b_0|^2/N_b$ in the initial state $|b,0\rangle$ [9]:

$$P_b = 1 - \frac{1}{2}(1-\Gamma)\{1 + \tanh[G(1-\Gamma)(t-t_D)/2]\} \tag{20}$$

where $\Gamma = 2\gamma_1/G$ and $t_D = [G(1-\Gamma)]^{-1}\ln[N_b(1-\Gamma)]$ is the delay time. Asymptotically, P_b tends to the stationary value $\Gamma < 1$.

In the experiment of ref. [10], a cigar-shaped ^{87}Rb condensate was illuminated with single laser beam π polarized and detuned by $\Delta_2/(2\pi) = -340$ MHz from the D_2 line transition ($\lambda = 780$ nm), between $|b\rangle = |5^2S_{1/2}, F=1, m_F=1\rangle$ and $|e\rangle = |5^2P_{3/2}, F=1, m_F=1\rangle$. After emission of a photon σ_+ polarized in the end-fire mode of the condensate, the atoms return to the ground state $|c\rangle = |5^2S_{1/2}, F=2, m_F=2\rangle$, recoiling at an angle of 45^0 with momentum $\vec{p} = \hbar\vec{q}$. The emitted photon is shifted by $-(\Delta_{cb}+\omega_r)$, where $\Delta_{cb} = (2\pi)6.8$GHz is the shift between the hyperfine ground states and $\omega_r = \hbar k_2^2/m = (2\pi)7.5$kHz is the recoil frequency. Normal emission with the atom back to the same ground state $|b\rangle$ is avoided aligning the polarization of the laser beam parallel to the main axis of the condensate. The condensate contained $N_b = 10^7$ atoms and had Thomas-Fermi radii of $R_\parallel$ =165 μm and $R_\perp$ =13.3 μm, so that $g_1 = 5\times10^7/s$ and $g \approx 10^5\sqrt{I}/s$, where I is the laser intensity in mW/cm^2. Assuming $\kappa = c/2R_\parallel \approx 10^{12}/s$, the predicted superradiant gain is $G/I \approx 2\times10^5 cm^2/(mWs)$. The measured gain was $G/I \approx 3\times10^4 cm^2/(mWs)$ and the loss rate was $2\gamma_1 = 6.2\times10^4/s$. For $I = 7.6 mW/cm^2$, $\Gamma \approx 0.27$, thus approximately 73% of atoms were transferred from the initial state $|b,0\rangle$ to the final state $|c,1\rangle$, with a momentum $\vec{p} = \hbar\vec{q}$.

Let now consider the effects of having $N_c = \alpha N_b$ atoms in the ground state $|c,0\rangle$, with initial momentum equal to zero. In fig.2(a) we show the results of the numerical integration of Eqs.(16)-(19), with $G = 10\omega_r$, $\gamma_1 = \gamma_2 = 0.3\omega_r$, and different values of $\alpha = 0.1, 0.5, 1$. We observe that it is possible to transfer almost 20% of atoms in the state $|b,-1\rangle$, moving backward with momentum $\vec{p} = -\hbar\vec{q}$. The fraction of backward atoms is rather small due to the off-resonance by $2\omega_r$ of the frequency $\omega_1 = \omega_2 - (\Delta_{cb}+\omega_r)$ of the superradiant field. Increasing the laser intensity it is possible to make the two populations of $|b,0\rangle$ and $|b,-1\rangle$ almost equal, if initially $N_b = N_c$. Fig.2(b) shows the photon flux per atom, $2\kappa|a|^2/N_b = G|S_1+S_2|^2$, for $\alpha = 0.1, 0.5, 1$. The radiation peak reduces increasing α, because the absorbtion from the second transition becomes more important.

In conclusion, we presented a quantum theory describing the experimentally observed superradiant Raman scattering from a Bose-Einstein condensate driven by a single off-resonant laser beam. We showed that collective atomic recoil lasing (CARL) from 3-level atoms in a Λ-configuration, realized using two hyperfine levels of the ground state, produces Raman amplification of matter waves. In particular, when atoms are initially in one of the two lower states, a pure two-level system is realized between atoms with different internal states and different momentum, and entangled atom-photon pairs are generated. In this case, the system behaves as a non-degenerate optical parametric amplifier. When the atoms are initially in both the hyperfine levels of the ground state, photons emitted superradiantly by atoms in the first two-level system can be absorbed by atoms in the second two-level system, generating a condensate recoiling in the backward direction. We observe that in this case it should be possible to measure experimentally any eventual difference between decoherence rates for atoms recoiling in opposite directions. In fact, a recent experiment [9] gave evidence of a phase-diffusion contribution to atomic decoherence depending on the detuning from the two-photon Bragg resonance condition. In the present case, superradiant photons resonantly emitted in one transition do not satisfy the resonance condition for the other transition. Hence, it should be possible to evaluate the phase-diffusion contribution to decoherence measuring the final steady-state fraction of atoms in the two recoiling condensates.

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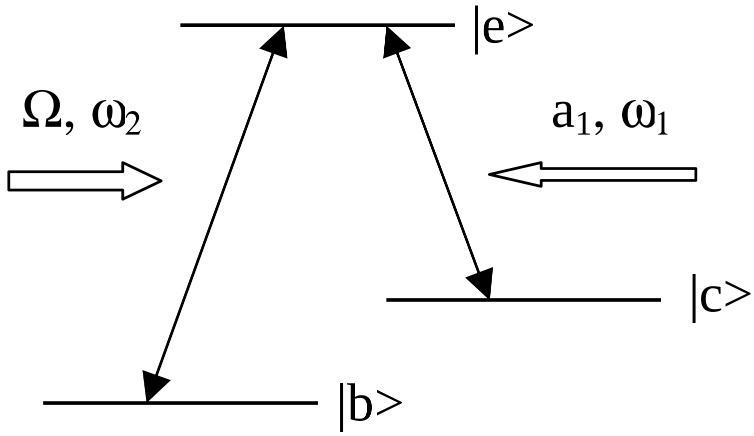

FIG. 1: Three-level Λ-shaped atoms coupled to a quantized probe laser a_1 and a classical coupling laser Ω with frequency ω_1 and ω_2, respectively.

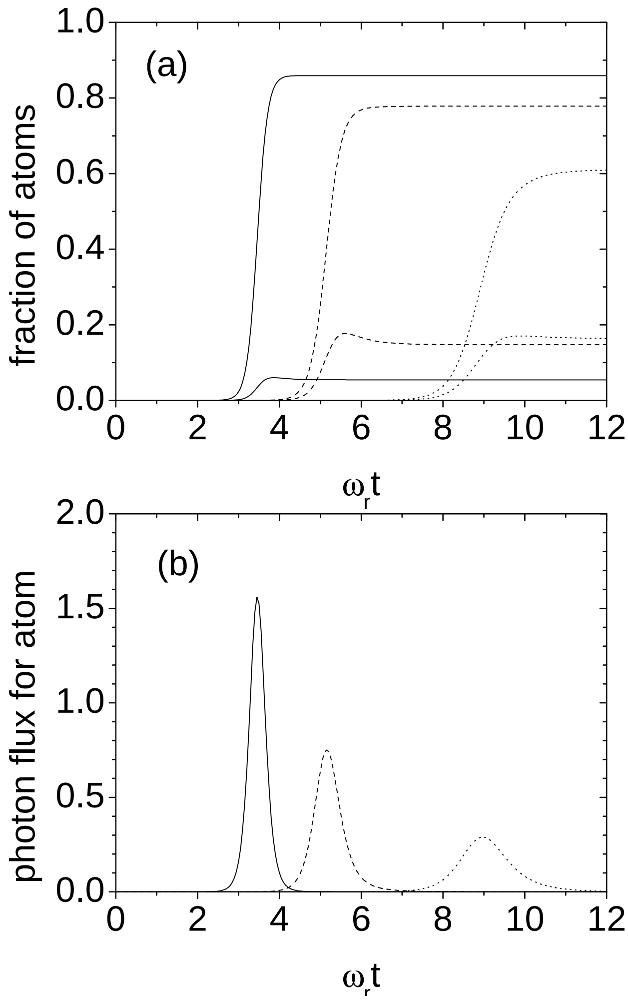

FIG. 2: (a): Fraction of atoms in $|c, 1\rangle$ (upper curves) and in $|b, -1\rangle$ (lower curve) vs. $\omega_r t$, for $G = 10\omega_r$, $\gamma_1 = \gamma_2 = 0.3\omega_r$ and $\alpha = N_c/N_b = 0.1$ (continuous lines), $\alpha = 0.5$ (dashed lines) and $\alpha = 1$ (dotted lines). (b): photon flux for atom, $2\kappa|a|^2/N_b = G|S_1 + S_2|^2$, for $\alpha = 0.1$ (continuous line), $\alpha = 0.5$ (dashed line) and $\alpha = 1$ (dotted line).