Photon scattering from a strongly driven multi-atom system: second-order correlations and squeezing

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Photon scattering from a strongly driven many-particle system is investigated. The second-order correlation function for light emitted from a strongly and near-resonantly driven dilute cloud of atoms is discussed. It is shown that photon scattering from strongly driven multi-atom systems exhibits bunching together with super-Poissonian or sub-Poissonian statistics. Next, squeezing in the resonance fluorescence emitted by a regular structure of atoms is discussed. In a suitable modified environment, squeezing even occurs for a resonant driving field, in contrast to the regular vacuum case.

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How to characterize light is a basic but ubiquitous problem in many branches of physics. In quantum physics, correlation functions are widely used in discussing the foundations of the underlying theory. As one of the most important model system, Young's double-slit experiment, despite its simplicity, exhibits the first-order coherence properties of light and allows to explore fundamental questions such as complementarity and uncertainty relations[1]. The remarkable progress of trapping atoms makes it possible to investigate experimentally the interference of the fluorescence light from two driven atoms which play the role of the slits in Young's experiment[2]. In this experiment, the two slits were replaced by two $^{198}\mathrm{Hg}^{+}$ ions in a linear trap and the interference pattern in the light scattered from the two ions was observed. However, it was shown that, in the strong-field limit, the interference vanished at strong driving[3−10]. This restricts potential applications, e.g., coherent backscattering from disordered structures of atoms[11], the generation of squeezed coherent light by scattering light off of a regular structure[12], the lithography[13,14], or precision measurements and optical information processing.

The interference vanishing at strong driving can be understood from the two-particle collective dressed states. In the strong-field limit, the two-particle collective dressed states are uniformly populated, i.e., the probabilities of the symmetric transitions and of the antisymmetric transitions are the same, so that the fringes with bright center and those with dark center cancel with each other. Some of us presented a scheme to recover first-order interference with almost full visibility in strong fields by tailoring the surrounding electromagnetic bath with a suitable frequency dependence[15], e.g., with the help of cavities. Based on this idea, we have proposed a scheme to generate squeezing in strong driving field[16].

However, it is not possible to extract the quantum properties from the first-order correlation functions. This motivated the study of second-order correlations, initiated by the intensity-correlation experiments conducted by Hanbury-Brown *et al.*[17]. Subsequently, second-order correlation measurements have found applications in many fields of modern physics[18], such as astronomy[19], optics[20,21], high-energy physics[22], condensed matter physics[23−25], and atomic physics[26]. The normalized second-order correlation functions $g^{(2)}(\tau)$ is measured by two detectors positioned at $\vec{R}_1$ and $\vec{R}_2$. $g^{(2)}(\tau=0)$ describes the photon statistics (e.g., sub/super-Poissonian), whereas $g^{(2)}(\tau\neq 0)$ indicates photon bunching or antibunching. Particularly, sub-Poissonian and antibunching exhibit the quantum properties of the radiation field. We study photon scattering from a strongly and near-resonantly driven dilute cloud of atoms[27], and recently we have demonstrated that n-particle atomic correlations can be directly detected via light scattering from an ensemble of laser-driven atoms in which either a dipole-dipole or a Rydberg-Rydberg interaction exits[28].

In this letter, we study a many-particle system radiated by a strong driving field. In the strongly driven case, Mollow spectrum with three distinct peaks shows up[29], for which the spectral properties can be defined individually. To start with, we focus on the case of a strong near-resonant driving of the ensemble and study the generation of correlated light from a disordered many-particle system. It is shown that the second-order correlations for various combinations of photons from different spectral lines exhibit bunching together with super-Poissonian or sub-Poissonian photon statistics, depending on the detector positions. Later, we propose a scheme to generate strong squeezing light from a linear chain of identical atoms by embedded the system in a modified reservoir. Our research shows that with a suitable modification of the surrounding electromagnetic bath, squeezing is recovered at strong driving even in the resonant case.

We describe the system in a suitable dressed state picture. All particles have identical atomic transition frequencies ω_0, and are localized at positions $\vec{r}_j$ with $j\in\{1,2,\cdots,N\}$. We define the inter-particle separation vectors as $\vec{r}_{ij}=\vec{r}_i-\vec{r}_j$. The external laser field has frequency $\omega_L=ck_L=2\pi c/\lambda_L$, wave vector $\vec{k}_L$ and wavelength λ_L. In electric dipole and rotating wave approximations, the system Hamiltonian can be written as $H = H_0 + H_I$[15], and

$$H_0 = \sum_k \hbar(\omega_k - \omega_L)a_k^\dagger a_k + \sum_{j=1}^{N} \hbar\tilde{\Omega}_j R_{zj}, \tag{1a}$$

$$H_I = \mathrm{i}\sum_k \sum_{j=1}^{N} (\vec{g}_k \cdot \vec{d}_j)\{a_k^\dagger S_j^- e^{-i(\vec{k}-\vec{k}_L)\cdot\vec{r}_j} - \text{H.c.}\}, \tag{1b}$$

$$S_j^- = \frac{R_{zj}}{2}\sin 2\theta_j - R_{21}^{(j)}\sin^2\theta_j + R_{12}^{(j)}\cos^2\theta_j, \tag{1c}$$

where H_0 represents the Hamiltonian of the free electromagnetic field (EMF) and free dressed atomic subsystems, respectively, H_I accounts for the interaction of the laser-dressed atoms with the EMF, and a_k and $a_k^\dagger$ are the field annihilation and creation operators obeying the standard commutation relations for bosons, respectively. The atomic operators $R_{\alpha\beta}^{(j)} = |\tilde{\alpha}\rangle_{jj}\langle\tilde{\beta}|$ describe the transitions between the dressed states $|\tilde{\beta}\rangle_j$ and $|\tilde{\alpha}\rangle_j$ in atom j for $\alpha \neq \beta$ and dressed-state populations for $\alpha = \beta$, and satisfy the commutation relations of the su(2) algebra. The dressed states $|\tilde{\alpha}\rangle_j$ entering the operators $R_{\alpha\beta}^{(j)}$ can be represented through the bare states $|\alpha\rangle_j$ via the transformations

$$\begin{aligned} |1\rangle_j &= \sin\theta|\tilde{2}\rangle_j + \cos\theta|\tilde{1}\rangle_j, \\ |2\rangle_j &= \cos\theta|\tilde{2}\rangle_j - \sin\theta|\tilde{1}\rangle_j\,. \end{aligned} \tag{2}$$

We further define $R_{zj} = |\tilde{2}\rangle_{jj}\langle\tilde{2}| - |\tilde{1}\rangle_{jj}\langle\tilde{1}|$ is the difference of the upper and lower dressed state population, and $\tilde{\Omega} = \tilde{\Omega}_j = \sqrt{\Omega^2 + (\Delta/2)^2}$ is the generalized Rabi frequency, with $2\Omega = (\vec{d}\cdot\vec{E}_L)/\hbar$. Here, $\vec{E}_L$ is the electric laser field strength, and $\vec{d} \equiv \vec{d}_j$ is the transition dipole matrix element. The detuning $\Delta = \omega_0 - \omega_L$ is characterized by $\cot 2\theta = \Delta/(2\Omega)$.

The master equation for an arbitrary atomic operator $Q\,(t)$ under Born approximation, Markovian approximation and secular approximation can be obtained by

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}\langle Q\,(t)\rangle = {} & \mathrm{i}\tilde{\Omega}\sum_{j=1}^{N}\langle[R_{zj}, Q\,(t)]\rangle \\ & - \frac{\gamma\,(\omega_L)}{4}\sin^2(2\theta)\sum_{j=1}^{N}\mathcal{L}\,(R_{zj}) \\ & - \gamma\,(\omega_-)\sin^4\theta\sum_{j=1}^{N}\mathcal{L}(R_{12}^{(j)}) \\ & - \gamma\,(\omega_+)\cos^4\theta\sum_{j=1}^{N}\mathcal{L}(R_{21}^{(j)}), \end{aligned} \tag{3a}$$

$$\mathcal{L}(A) = \langle A[A^\dagger, Q(t)]\rangle + \langle[Q(t), A]A^\dagger\rangle, \tag{3b}$$

where ω_L and $\omega_\pm = \omega_L \pm 2\tilde{\Omega}$ are the dressed state transition frequencies with the decay rate $\gamma\,(\omega_L)$ and $\gamma\,(\omega_\pm)$, respectively. We find in the master equation that, in the dressed states, the Mollow spectrum has already showed up. The central band emits at the laser frequency ω_L, and the left (right) band at ω_- (ω_+)[30,31]. In the strong-field limit, different lines of the spectral are well-separated. In particular, in the following we will denote light originating from $R_{zj}\sin(2\theta)/2$ as the central spectral component indicated by C, and $R_{21}^{(j)}\cos^2\theta$ and $R_{12}^{(j)}\sin^2\theta$ as the right (R) and left (L) spectral sideband components. In what follows, we shall use this decomposition to investigate the properties of the scattered light.

We proceed by investigating an atomic sample of arbitrary shape and of characteristic size d, consisting of distinguishable non-overlapping two-level particles (see Fig. 1). The typical inter-particle separation l satisfies the restriction $\lambda_L \ll l \ll d$ with $d/c < \tau_\mathrm{s}$, where τ_s is the spontaneous decay time.

What we are interested in is the normalized second-order correlation functions $g^{(2)}(\tau)$, which is defined as

$$\begin{aligned} g_{mn}^{(2)}(\tau,\vec{R}_1,\vec{R}_2) &= \frac{G_{mn}^{(2)}(\tau,\vec{R}_1,\vec{R}_2)}{I_m(\vec{R}_1)I_n(\vec{R}_2)} \\ &= \frac{\langle a_m^\dagger(\vec{R}_1)a_n^\dagger(\tau,\vec{R}_2)a_n(\tau,\vec{R}_2)a_m(\vec{R}_1)\rangle}{I_m(\vec{R}_1)I_n(\vec{R}_2)}, \end{aligned} \tag{4}$$

i.e., as the unnormalized second order correlation function normalized to the intensities $I_m(\vec{R}_1)$ and $I_n(\vec{R}_2)$. The quantity $g_{mn}^{(2)}$ for $\{m, n\} \in \{C,\ R,\ L\}$ can be interpreted as a measure for the probability of detecting one photon emitted in mode m and another photon emitted in mode n with time-delay τ.

To calculate the correlation function, we assume laser driving on resonance ($\theta = \pi/4$), and a large atomic ensembles ($N \gg 1$) such that the secular approximation is valid. We also assume that all possible pairs of atoms contribute equally to the second-order correlation functions. This assumption is valid as long as the angle between the wave vectors of the incident laser and the scattered photons is small, that is $\{\phi, \phi_0\}$ should be of order of few degrees (see Fig. 1). Finally, we for the moment consider a single interparticle distance vector $\vec{r}_{ji}$ for

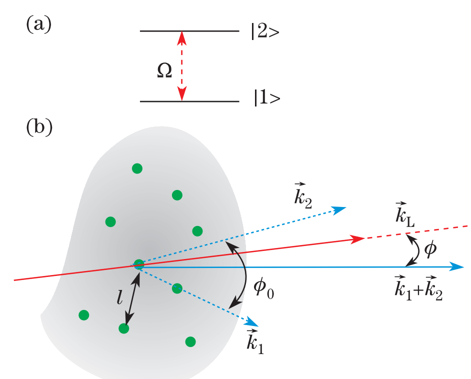

Fig. 1. (Color online) Schematic setup of a dilute atomic ensemble pumped with a coherent field with wave-vector $\vec{k}_L$. (a) Energy levels of each of the ensemble particles, and the interaction with the strong coherent light with coupling strength Ω; (b) ensemble with typical inter-particle distance length scale denoted by l with $l \gg 2\pi/k_L$. We consider the case of photon pair emission in forward direction and denote the angle between the two emitted photons with wave-vectors $\vec{k}_1$ and $\vec{k}_2$ as ϕ_0, and $\vec{k}_1 + \vec{k}_2 \approx 2\vec{k}_L$. The direction of the emission cone defined by $\vec{k}_1$ and $\vec{k}_2$ is characterized by the angle ϕ between $(\vec{k}_1 + \vec{k}_2)/2$ and $\vec{k}_L$.

all pairs only, but this restriction will be relaxed later on. Based on these assumptions, we find that the correlation and cross-correlation functions of photons scattered into the different spectral bands can be represented as

$$g^{(2)}_{CC}(\tau,\vec{R}_1,\vec{R}_2) = 1 + 2\cos(\delta_1)\cos(\delta_2)e^{-2\gamma\tau}\,, \quad \text{(5a)}$$
$$g^{(2)}_{LL}(\tau,\vec{R}_1,\vec{R}_2) = g^{(2)}_{RR}(\tau,\vec{R}_1,\vec{R}_2) = 1 + \cos(\delta_1 - \delta_2)e^{-3\gamma\tau}\,, \quad \text{(5b)}$$
$$g^{(2)}_{LR}(\tau,\vec{R}_1,\vec{R}_2) = g^{(2)}_{RL}(\tau,\vec{R}_1,\vec{R}_2) = 1 + \cos(\delta_1 + \delta_2)e^{-3\gamma\tau}\,, \quad \text{(5c)}$$
$$g^{(2)}_{CX}(\tau,\vec{R}_1,\vec{R}_2) = g^{(2)}_{XC}(\tau,\vec{R}_1,\vec{R}_2) = 1 \text{ for } X \in \{L,R\}, \quad \text{(5d)}$$

where $\delta_s = (\vec{k}_s - \vec{k}_L)\vec{r}_{ji}$ with $\vec{k}_s$ being the wave-vector of the photon s scattered in direction $\vec{R}_s$ ($s \in \{1,2\}$).

To estimate the signal obtained from a cloud of randomly distributed particles, the correlation functions $g^{(2)}_{XY}(\tau,\vec{R}_1,\vec{R}_2)$ have to be averaged over the different interatomic distance vectors $\vec{r}_{ij}$ in the cloud, which potentially could eliminate the correlations. We choose an averaging procedure as simple as possible: (i) an isotropic average over the relative orientation $\mathbf{n}$ of the atoms over the unit sphere, followed by (ii) an average of the interatomic distance $r_{ji} = |\vec{r}_{ji}|$ over an interval of order of the laser wave-length, around their typical distance l[32]:

$$\langle\cdots\rangle_{\rm conf} = (k_L/4\pi)\int_{l-2\pi/k_L}^{l+2\pi/k_L} \mathrm{d}r_{ji} \int \mathrm{d}\Omega_{\mathbf{n}} \cdots\,. \quad (6)$$

The obtained results for the normalized second-order correlation function after the configuration average for the photons scattered in the central spectral band are shown in the upper row of Fig. 2. It can be seen that even after the averaging, the correlation function exhibits a sharp peak around direction of the the incident laser wave-vector $\vec{k}_L$, indicating super-bunching. Figure 2(a) shows the case in which two single-photon detectors are placed symmetrical with respect to the laser wave-vector $\vec{k}_L$ direction given by $\phi = 0$. Figure 2(b) depicts the same correlation function but detected with a single two-photon detector ($\phi_0 = 0$) for different emission directions ϕ. Note that the correlation function for two photons emitted both from the left or from the right spectral side-band does not show a directionality in space.

The bottom row of Fig. 2 shows the corresponding results for the two photon cross correlation with one photon emitted from the left, and one from the right spectral side-band. Again, we find maxima at $\phi_0 = 0$ for two individual detectors placed symmetrically around the incident laser direction. But in contrast to the central band correlation function, this maximum is not peaked, but rather broad. If a two-photon detector is used, a narrow maximum is observed in the forward direction (Fig. 2(d)). Interestingly, in this case, depending on the precise positioning around the forward direction, both Poissonian and sub-Poissonian photon-statistics can be generated. The emitted light intensity is proportional to the square of the number of particles, and thus can potentially be intense.

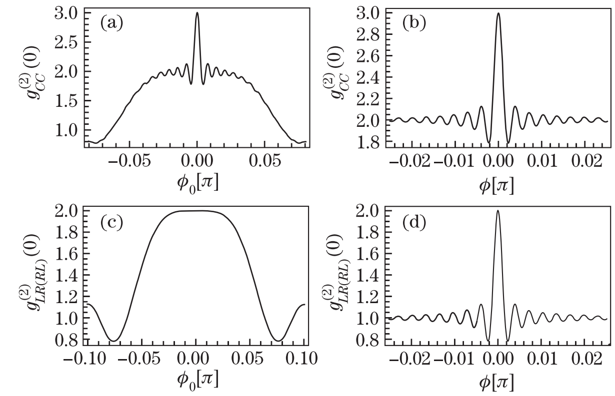

Fig. 2. Normalized and configuration averaged second-order correlation functions. (a, b) Correlation function $g^{(2)}_{CC}(0)$ between two photons emitted from the central spectral band is shown, (c, d) corresponding function $g^{(2)}_{LR}(0) = g^{(2)}_{RL}(0)$ for pairs with one photon emitted from each sideband. In (a,c), the correlation functions are shown as a function of the opening angle ϕ_0 between the two photons (see Fig. 1), which corresponds to the case of two distinct detectors. The two photons are measured at positions symmetric with respect to the incident laser field direction, i.e., $\phi = 0$. In (b,d), photon pairs with $\phi_0 = 0$ emitted in the same direction are considered, and plotted as a function of the emission direction ϕ. For all subfigures, the interparticle length scale is chosen as $l = 50\,\lambda_L$.

We discuss the atomic sources of squeezing from a regular N identical atoms system in a strong field. It has been shown that only few or no squeezing could be achieved with strong driving fields. To see the squeezing, the field is expressed in the term of the Hermitian amplitude operators $x_\sigma = (a^\dagger e^{i\sigma} + a e^{-i\sigma})/2$. In the bare states, we have $a^\dagger = \sum_{n=1}^{N} S_n^+$ and $a = \sum_{n=1}^{N} S_n$, where S_n^+ (S_n) is the upper (lower) operator for the nth atom. The condition for squeezing can be written in terms of the normally ordered variance:

$$\langle:(\Delta x_\sigma)^2:\rangle < 0, \quad (7)$$

in certain space-time intervals. In the multi-atom system, the individual atomic operators can be expressed as $S_n^\pm = S^\pm \exp[\mp i(\omega_L t + \phi - \vec{k}_L\cdot\vec{R}_n - \frac{\omega_L}{c}|\vec{r} - \vec{R}_n|)]$, where $\vec{R}_n$ and $\vec{r}$ are the position of the nth atom and the detector, and ϕ is the phase of the exciting laser wave. We assume that the linear dimensions of the scattering volume are small compared with the distance between the scattering volume and the point of the observation, therefore we may write

$$\vec{k}_L\cdot\vec{R}_n + \frac{\omega_L}{c}|\vec{r} - \vec{R}_n| \approx \vec{k}'\cdot\vec{r} + (\vec{k}_L - \vec{k}')\cdot(\vec{R} + \vec{r}_n), \quad (8)$$

where we have used the abbreviation $\vec{k}' = \frac{\omega_L}{c}\frac{\vec{r}-\vec{R}}{|\vec{r}-\vec{R}|}$.

We focus on the squeezing in modified reservoirs. In the free space, the decay rates from the three bands are equal $\gamma(\omega_+) \approx \gamma(\omega_-) \approx \gamma(\omega_L)$. However, in the modified reservoir this relation no longer holds. Particularly, we assume that the mode density at one of the side bands is either reduced or enhanced, for example via a cavity. Thus, we assume that $\gamma_+ = \gamma(\omega_+)$ is no longer the same as $\gamma_- = \gamma(\omega_-)$, while $\gamma_0 = \gamma(\omega_L)$ remains unchanged. We write operators into the dressed states and obtain

$$\begin{aligned}\langle:(\Delta x_\sigma)^2:\rangle =& \frac{N}{4}\{\left(\kappa_+^2\cos^4\theta+\kappa_-^2\sin^4\theta+2\kappa_0^2\sin^2\theta\cos^2\theta\right)\\&+\left(\kappa_+^2\cos^4\theta-\kappa_-^2\sin^4\theta\right)\langle R_z\rangle\\&+\frac{1}{2}\sin^2 2\theta\left(\kappa_0^2-\kappa_+\kappa_-\right)|F|\cos\left(2\sigma+2\psi\right)\\&-\frac{1}{2}\kappa_0^2\sin^2 2\theta\langle R_z\rangle^2[1+|F|\cos\left(2\sigma+2\psi\right)]\},\end{aligned} \tag{9}$$

where

$$F = \frac{1}{N}\sum_{n=1}^{N}\exp[2\mathrm{i}\left(\vec{k}_L-\vec{k}'\right)\cdot\vec{r}_n] = |F|\,\mathrm{e}^{\mathrm{i}\arg F}, \tag{10a}$$

$$\psi = \vec{k}'\cdot\vec{r}-\omega t+\left(\vec{k}_L-\vec{k}'\right)\cdot\vec{R}+\frac{1}{2}\arg F-\phi, \tag{10b}$$

where θ is defined as $\cot 2\theta = \Delta/(2\Omega)$. In the long-time limit, the atoms assume the steady-state indicated by subindex s, and we have $\langle R_z\rangle \equiv \langle R_{zi}\rangle_s$. The parameters $\kappa_\pm$ are related to the decay rates by $\kappa_+^2/\kappa_-^2 \approx \gamma_+/\gamma_-$.

We consider that the atoms are distributed in a linear chain. The distance vector between two neighboring atoms is $\vec{r}_0$. Equation (10a) can be simplified when detect the fluorescent light in such directions which satisfy the condition $(\vec{k}-\vec{k}')\cdot\vec{r}_0 = n\pi, n = 0, \pm 1, \pm 2, \cdots$. In these directions, we have $|F| = 1$. If we choose the phase ψ appropriately, from Eq. (9), it is clearly seen that the resulting strength of squeezing in N times stronger than in the single-atom case. From Eq. (3a), we derive the equations of motion and obtain the steady-state solutions

$$\langle R_{zi}\rangle_s = \frac{\gamma_-\sin^4\theta-\gamma_+\cos^4\theta}{\gamma_-\sin^4\theta+\gamma_+\cos^4\theta}, \tag{11a}$$

$$\langle R_{21}^{(i)}\rangle_s = \langle R_{12}^{(i)}\rangle_s = 0. \tag{11b}$$

Suppose for instance that in the reservoir we have $\kappa_+ \ll \kappa_-$, i.e., $\gamma_+ \ll \gamma_-$. For a resonant driving field ($\theta = \pi/4$), one has $\langle R_z\rangle \approx 1$, and Eq. (9) becomes

$$\langle:(\Delta x_\sigma)^2:\rangle = \frac{N}{8}\kappa_+\{\kappa_+-\kappa_-|F|\cos[2(\sigma+\psi)]\}, \tag{12}$$

which is negative for $2(\sigma+\psi) = 2n\pi$ ($n \in \{0,1,\cdots\}$) and $|F| = 1$. On the contrary, for $\kappa_+ \gg \kappa_-$, and a resonant driving field ($\theta = \pi/4$), one has $\langle R_z\rangle \approx -1$, and therefore

$$\langle:(\Delta x_\sigma)^2:\rangle = \frac{N}{8}\kappa_-\{\kappa_--\kappa_+|F|\cos[2(\sigma+\psi)]\}, \tag{13}$$

which again is negative for $2(\sigma+\psi)=2n\pi$ ($n \in \{0,1,\cdots\}$) and $|F|=1$. From these results we find that the quantum fluctuations of the scattered light can be squeezed even in the case of a resonant driving field, in contrast to the free space case.

In Fig. 3, we show the uncertainties $\langle:(\Delta x_\sigma)^2:\rangle$ given by Eq. (9) as a function of the detuning $\Delta/2\Omega$ in different modified environments. The vertical solid line at $\Delta/2\Omega=0$ indicates the resonant case, and the shadow areas below the horizontal solid line correspond to the situations that suqeezing happen ($\langle:(\Delta x_\sigma)^2:\rangle<0$). It is shown that the modification of the environmental vacuum reservoir improves the squeezing, as the minimum of the curves becomes more and more negative with increasing modification (see the dash-dotted, dashed, and dotted curves in Fig. 3). From our analytical result, one can see that the squeezing originates from contributions from the sidebands of the Mollow triplet, i.e., the terms proportional to $\kappa_+\kappa_-$.

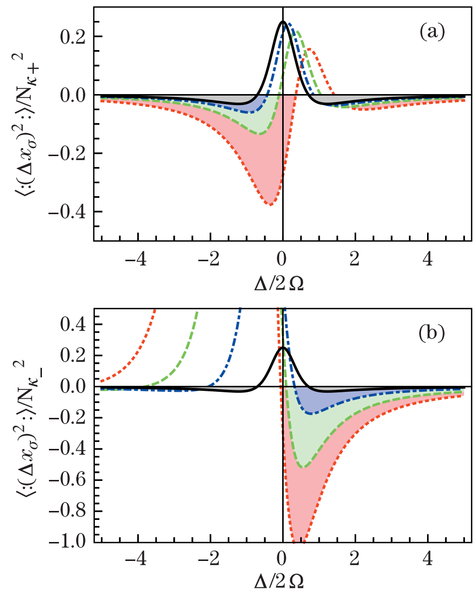

Fig. 3. (Color online) Uncertainty $\langle:(\Delta x_\sigma)^2:\rangle$ plotted against the detuning $\Delta/2\Omega$. (a) $\gamma_+ < \gamma_-$. The dotted (red) line depicts $\gamma_+/\gamma_- = 0.05$, the dashed (green) one shows $\gamma_+/\gamma_- = 0.2$, the dash-dotted (blue) line is for $\gamma_+/\gamma_- = 0.5$, while the solid (black) curve corresponds to the free space $\gamma_+/\gamma_- = 1$. (b) $\gamma_+ > \gamma_-$. The dotted (red) line depicts $\gamma_+/\gamma_- = 150$, the dashed (green) one shows $\gamma_+/\gamma_- = 50$, the dash-dotted (blue) line is for $\gamma_+/\gamma_- = 10$, while the solid (black) curve corresponds to the free space $\gamma_+/\gamma_- = 1$. Here $|F| = 1$, $\kappa_+ \approx \kappa_0$ and $2(\sigma+\psi) = 2n\pi$ with $n \in \{0,1,\cdots\}$.

In conclusion, we study a many-particle system radiated by a strong driving field, for which the spectral properties can be defined individually. To investigate the quantum effects of the radiation field, we study the second-order correlations of light scattered from a disordered system, and we find that the second-order correlations for various combinations of photons from different spectral lines exhibit bunching together with super-Poissonian or sub-Poissonian photon statistics, tunable by the choice of the detector positions. Later, to overcome the limitation of squeezing in weak driving field, we propose a scheme to generate strong squeezing light from a linear chain of identical atoms by the modification of the reservoir. It is shown that squeezing can be recovered at strong driving even in the resonant case.

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