Quantum dynamics and statistical properties of atom-molecule Bose-Einstein condensates

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Based on a two-mode boson model, we study nonclassical properties of the atom-molecule Bose-Einstein condensate. The effects of nonlinear collisions on the dynamics of the molecular formation is studied both in classical and quantum treatments. We find that the conversion from atoms to molecules can be suppressed strongly due to nonlinearity induced localization of the atomic population. In addition, we study statistical properties of the atom-molecule condensed system by calculating the intensity correlation functions numerically. We find that the effect of nonlinearity leads to the appearance of superchaotic molecular pulses, while maintaining the atomic field sub-Poissonian. The joint quantum statistical properties of the atoms and the molecules always show antibunching.

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Recently, possibility of preparing atom-molecule Bose-Einstein condensate (AMBEC) has attracted wide attention [1–4]. Condensed bosonic atoms can be converted to a molecular condensate by using either the photoassociation process [1,5,6] or the so-called Feshbach resonance method [2,3,7,8]. Wieman's group in JILA [7,8] measured the first-order temporal mutual coherence of atoms and molecules by using two-photon Ramsey experiment. Similarly with previous works [9,10], almost coherent Rabi oscillations (Ramsey interference pattern) were demonstrated. Theoretically, their experimental results were reproduced partially by using coupled Gross-Pitaevski (GP) equations [11–14]. According to the mean-field theory (MFT), large-amplitude coherent oscillation between the two-field modes is expected. Vardi *et al.* [15] found that quantum-field solutions modify the Bose-enhanced factor of the oscillation frequency. Moreover, many-body quantum effects lead to the appearance of collapse and revival of the coherent oscillations [5,15].

In this paper, we study nonclassical properties of the AMBEC based on a two-mode bosonic model [5,15]. The role of nonlinearity on quantum dynamics of the AMBEC is studied. We find that nonzero interspecies and intraspecies interactions result in modulational instability, and modify the dynamics of the AMBEC. The conversion of atoms into molecules is shown to be dramatically suppressed due to nonlinearity induced localization of the atomic population. We also study the statistical properties of the AMBEC by calculating the intensity correlation functions numerically. Our results show that due to the nonlinearity, the initial sub-Poissonian molecular field is transformed into superchaotic molecular pulses, whereas the atomic field remains in the sub-Poissonian region. In addition, the joint quantum statistical properties of the atoms and the molecules always exhibit antibunching.

We consider the atom-molecule condensate system coupled via one-color photoassociation or Feshbach resonance. The total system can be described phenomenologically by a two-mode bosonic Hamiltonian [5,15] ($\hbar=1$):

$$\hat{H}=-\frac{\delta}{2}\hat{a}^{\dagger}\hat{a}+\frac{g}{2}(\hat{b}^{\dagger}\hat{a}^{2}+H.c.)+\lambda_{ab}\hat{a}^{\dagger}\hat{a}\hat{b}^{\dagger}\hat{b}+\frac{\lambda_{a}}{2}\hat{a}^{\dagger}\hat{a}^{\dagger}\hat{a}\hat{a}+\frac{\lambda_{b}}{2}\hat{b}^{\dagger}\hat{b}^{\dagger}\hat{b}\hat{b}, \quad (1)$$

where δ denotes the detuning between the molecular and atomic field. The atomic-field and molecular-field operators, $\hat{a}$ and $\hat{b}$ obey the standard bosonic commutation relationship. The atom-atom, molecule-molecule, and atom-molecule elastic interactions are described by the s-wave scattering strengths λ_a, λ_b, and $\lambda_{ab}(\lambda_{ba})$ [2,6], respectively. It should be mentioned that λ_b and λ_{ab} are still unknown [4,6]. Following Refs. [16,17], we take $\lambda_b\sim 2\lambda_a(\sim 10^{-3}$ g) and $\lambda_{ab}\sim -1.5\lambda_a$. Due to the conserved particle number $\hat{N}=\hat{n}_a+2\hat{n}_b$, where $\hat{n}_a=\hat{a}^{\dagger}\hat{a}$ and $\hat{n}_b=\hat{b}^{\dagger}\hat{b}$, the Hilbert space is spanned by the basis [18]: $|\phi_n\rangle=|N-2n\rangle_a|n\rangle_b$ for $n=0,1,\ldots,[N/2]$, where $[z]$ denotes taking the greatest integer of any real number z. The dynamics of the AMBEC for arbitrary initial states can be solved by using the Runge-Kutta numerical scheme.

Before investigating the quantum dynamics of the AMBEC, we first derive the classical equations of motion and discuss their steady states. The GP-type equations for the atomic and molecular fields can be obtained from the Heisenberg equations by replacing $\hat{a}\rightarrow\langle\hat{a}\rangle=\sqrt{\langle N\rangle}\Phi_a$ and $\hat{b}\rightarrow\langle\hat{b}\rangle=\sqrt{\langle N\rangle}\Phi_b$, where the wave functions for the two-field modes $\Phi_j=|\Phi_j|e^{i\phi_j}$ $(j=a, b)$ obey the normalized condition: $|\Phi_a|^2+2|\Phi_b|^2=1$. To get the steady states and the modulational instability conditions [19], we introduce two conjugate variables [20,21]: $x=|\Phi_a|^2$ and $\varphi=2\phi_a-\phi_b$. They obey the canonical relations: $dx/d\tau=-\partial\mathcal{H}/\partial\varphi$, $d\varphi/d\tau=\partial\mathcal{H}/\partial x$, and the canonical Hamiltonian is given by

$$\mathcal{H}=\Delta x-\frac{\Lambda}{2}x^{2}-x\sqrt{1-x}\cos(\varphi), \quad (2)$$

where the scaled time $\tau=\sqrt{2}Gt$. In derivation of the above equations, we have introduced

$$\Delta = (\sqrt{2}G)^{-1}(\delta - \Lambda_{ab} + \Lambda_b/2), \tag{3}$$

$$\Lambda = (G/\sqrt{2})^{-1}(\Lambda_a - \Lambda_{ab} + \Lambda_b/4), \tag{4}$$

where $G=\sqrt{\langle N\rangle}g$ and $\Lambda_{ij}=\langle N\rangle\lambda_{ij}$. The stationary solutions obey $(dx/d\tau)_{x_0}=(d\varphi/d\tau)_{\varphi_0}\equiv 0$. From the relation $x_0\sqrt{1-x_0}\sin(\varphi_0)=0$, we find that there are three kinds of fixed points: (i) pure molecular phase with $x_0=0$, (ii) in-phase type of steady state with $\varphi_0=0$, (iii) out-of-phase type of steady state with $\varphi_0=\pi$ [16]. The value of x_0 determined by a cubic equation

$$\Delta - \Lambda x_0 - \frac{1-3x_0/2}{\sqrt{1-x_0}}\cos(\varphi_0) = 0 \tag{5}$$

can be solved numerically. It was shown that the atomic phase at $|\Delta|\to\infty$ can be converted to the molecular phase or vice versa by varying the detuning Δ adiabatically [5,21]. In the intermediate region, there are two types of atom-molecule coexisting phases: the in-phase steady state for $\varphi_0=0$ in the parameter region $\Delta<1$, and the out-of-phase one for $\varphi_0=\pi$ in $\Delta>-1$ region.

Stability analysis of the steady states can be performed by introducing small perturbations: $x=x_0+\delta x$ and $\varphi=\varphi_0+\delta\varphi$. A coupled linearized equation for the perturbations gives the eigenfrequencies

$$\omega^2 = \omega_0^2 - \Lambda x_0\sqrt{1-x_0}\cos(\varphi_0), \tag{6}$$

where $\omega_0=\sqrt{x_0(1-3x_0/4)\cos^2(\varphi_0)/(1-x_0)}$. For a negligible nonlinear interaction $\Lambda=0$, aroused from either $\Lambda_i=\Lambda_{ij}=0$ or $\Lambda_{ab}=\Lambda_a+\Lambda_b/4$, both of the two steady states ($\varphi_0=0$, π) are stable [21]. However, for nonzero Λ, the above two steady states can be modulationally unstable when the eigenfrequencies become imaginary [22], i.e., $\omega^2<0$. In this case, the perturbations in the AMBEC undergo exponential growth with the growth rate being the imaginary part of ω. We find that, for $\varphi_0=0$, the MI occurs in the positive Λ region with $\Lambda>1$ and $\Delta<1$. For $\varphi_0=\pi$, the AMBEC is unstable in the negative Λ region with $\Lambda<-1$ and $\Delta>-1$. The MI region (and also the dynamics) of the AMBEC is invariant under the transformation $\Lambda\to-\Lambda$, $\varphi\to\pi-\varphi$, and $\Delta\to-\Delta$. Thus, in the following we only consider the case of positive Λ.

Dynamical evolution of the AMBEC can be described by the fraction of atoms converted to molecules [5,19], $f_M=2\langle\hat{b}^\dagger\hat{b}\rangle/\langle\hat{N}\rangle$, which measures the ratio of the atom number in the bound state to the total atom number. In the MFT, $f_M=2|\Phi_b|^2$. For the negligible nonlinear interaction $\Lambda=0$, there are analytical solutions for f_M [23]. Considering the initial condition $x(0)=1$, then $f_M=2f_1 sn^2(\sqrt{f_2}\tau/\sqrt{2},m)$, where $sn(z,m)$ is a Jacobi elliptic function with the elliptic modulus $m=f_1/f_2$, and $f_{1,2}=1/4[\Delta^2+2\mp\sqrt{\Delta^2(\Delta^2+4)}]$. In particular, for the exact "resonant" case with $\Delta=0$, $f_M=\tanh^2(\tau/2)$ since $m=1$. Starting from the zero initial value, f_M grows quickly to the maximum value 1 without any oscillation [the dotted line in Fig. 1(a)], which corresponds to a complete and irreversible molecule formation process. Non-

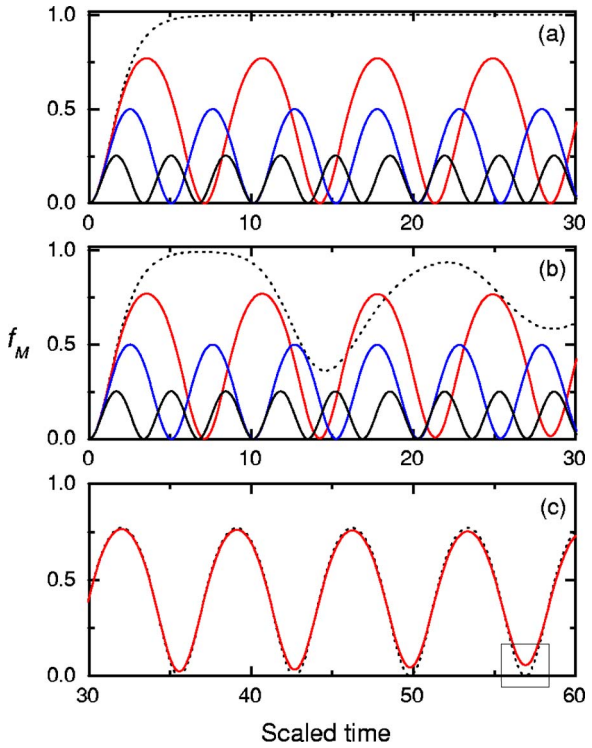

FIG. 1. (Color online) f_M vs the scaled time τ for (a) mean-field solutions, (b) quantum solutions, and (c) comparison of the two predictions for $\Lambda=0.4243$. The parameters are $\Delta=0$, $N=10^4$, and the nonlinearity in (a) and (b) are: $\Lambda=0.0$ (the dotted lines), $\Lambda=0.4243$ (the red lines), $\Lambda=0.9419$ (the blue lines), and $\Lambda=1.6971$ (the black solid lines).

zero Δ, however, results in periodic oscillations of f_M with smaller amplitude. If $\Delta>\Delta_c\equiv 1/\sqrt{2}$, the amplitude of f_M is always less than 1/2.

To investigate the effect of nonzero Λ, we need to know the values of the two-body interaction strengths. However λ_b and λ_{ab} have not been determined even in low-energy region [4,6]. Following Ref. [17], we adopt $\lambda_b=2\lambda_a$ and $\lambda_{ab}=-1.5\lambda_a$. Nonzero Λ can be achieved by tuning λ_a, such as $\lambda_a=1.0\times10^{-3}\,g$ ($\Lambda=0.4243$), $\lambda_a=2.22\times10^{-3}\,g$ ($\Lambda=0.9419$), and $\lambda_a=4.0\times10^{-3}\,g$ ($\Lambda=1.6971$). All the above parameters are in the stable region. The MFT results show that the effect of nonlinearity leads to periodic oscillations between atoms and molecules. As shown in Fig. 1(a), the amplitude of f_M decreases monotonically with the increase of Λ. If $\Lambda>0.94$, the whole curve of f_M is below 1/2, i.e., no more than 50% of atoms are converted to molecules. This nonlinearity induced localization of the atomic population is similar to the macroscopic self-trapping in the two-component BEC [20]. However, unlike the two-mode BEC system with Josephson-like coupling, the oscillation amplitude in AMBEC reduces continuously from unity to zero.

In Fig. 1(b), we evaluate numerically the quantum solutions of f_M for an initial number state $|\psi(0)\rangle=|N\rangle_a|0\rangle_b$. In the absence of nonlinearity $\Lambda=0$ and $\Delta=0$, the quantum solution of f_M [the dotted line in Fig. 1(b)] breaks away from the MFT result around the point $\tau\sim7.03$, where all the particles are in molecular state. This departure cannot be explained in the framework of the MFT. A semiclassical analysis of the

AMBEC in the vicinity of the all-molecules phase shows that there are two contributions to the atomic occupation [15]: (i) spontaneous dissociation into atomic vacuum, which is proportional to $\sqrt{N}\sinh^2(\tau/2)$, (ii) stimulated part proportional to $\langle \hat{n}_a \rangle$, which is negligible near the all-molecule phase. The Bose-enhanced spontaneous dissociation into the atomic vacuum results in collapse and revival of the atom-molecular oscillations [5,15], which is just similar with the phenomenon observed in the two-photon down-conversion process [18].

We further consider quantum solution of f_M for the case of $\Lambda \neq 0$. As shown in Fig. 1(b), we find that the quantum results almost coincide with the corresponding classical solutions shown in Fig. 1(a). Both of them exhibit "periodic" oscillations with decreased amplitude. The agreement of two solutions originates from the fact that the trajectories do not reach the all-molecule phase, and the contribution of quantum fluctuation of atomic mode is *not* significant compared to the coherent stimulated process. In Fig. 1(c), we compare the quantum solution (red solid line) with the MFT solution (dotted line) for $\Lambda=0.4243$. We find that the quantum solution is in fact the collapsed oscillations. From Fig. 1(b), we also find that the collapse time is enhanced significantly by the nonlinearity, i.e., the coherent oscillations are damped more slowly with the increase of Λ.

The observation of the first-order temporal coherence between atoms and molecules opens up the possibility to form molecular condensate. However, there remains many open questions to be answered [7], such as the detailed quantum state of the molecules, and their higher-order coherence functions in the temporal domain. Recently, Meiser *et al.* [24] studied the short-time dynamics of molecule formation and the second-order correlation function by using perturbation theory. They found that the second-order degree of coherence of the molecular field for an initial number state approaches unity in the limit of large N. Motivated by their work, we calculate the equal-time intensity correlation functions beyond the short-time limit

$$g_j^{(2)} = \frac{\langle \hat{n}_j(\hat{n}_j - 1)\rangle}{\langle \hat{n}_j\rangle^2}. \tag{7}$$

It is known from quantum optics that if $0 \leqslant g_j^{(2)} < 1$ then the field is in the so-called sub-Poissonian. The fields in a sub-Poisson distribution always exhibit nonclassical *antibunching* phenomena. $g_j^{(2)}=1$ represents a coherent state with Poisson distribution, while $g_j^{(2)}>1$ is characteristic for a super-Poisson distribution. In particular, for thermal or chaotic field, $g_j^{(2)}=2$.

Starting from the number state, the initial stage of the second-order coherence functions of two field modes obey $g_a^{(2)}=1-1/N$ and $g_b^{(2)}=1-4/N+\mathcal{O}(N^{-2})$ [24]. Both of them are less than 1. For zero nonlinearity $\Lambda=0$, as shown in Fig. 2(a), we find that the two field modes are transformed from sub-Poissonian to super-Poissonian, successively. Similarly with previous discussions, the AMBEC system with zero nonlinearity shares the same model of degenerate parametric amplifier [25]. Quantum fluctuations of the atomic vacuum

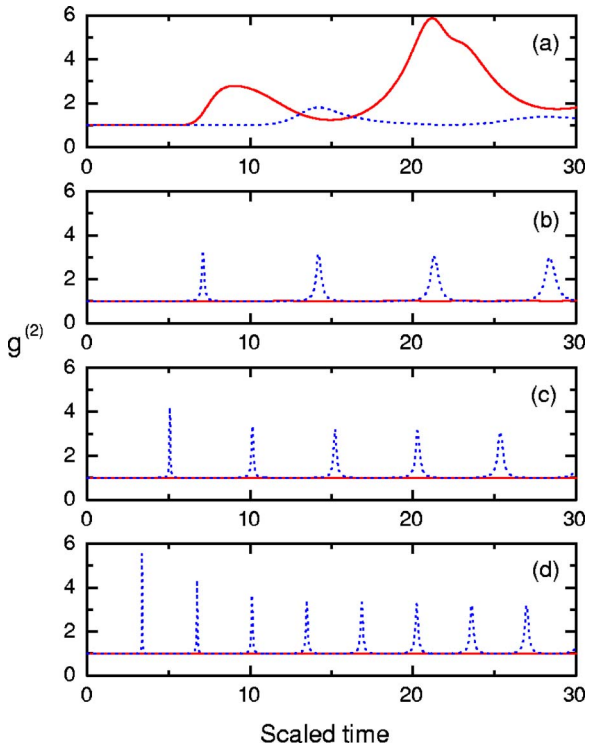

FIG. 2. (Color online) $g_a^{(2)}$ and $g_b^{(2)}$ vs the scaled time τ for various Λ. From top to bottom: (a) $\Lambda=0$, (b) $\Lambda=0.4243$, (c) $\Lambda=0.9419$, and (d) $\Lambda=1.6971$. The red solid lines are for $g_a^{(2)}$ and the blue dotted lines for $g_b^{(2)}$. Other parameters are the same as Fig. 1.

leads to the appearance of super-Poissonian, and the peaks of $g_a^{(2)}$ near the points $\tau \sim 7.03$ and 21.0, where the particles are almost in the molecular mode. The magnitude of $g_a^{(2)}>2$ indicates that the atoms generated from the photodissociation exhibits superchaotic (strong bunching) behavior [26].

The second-order coherence function $g_j^{(2)}$ for nonzero Λ are calculated in Figs. 2(b)–2(d). We find that the effects of nonlinearity result in sharp peaks in $g_b^{(2)}$ with their positions at the minima of f_M. Their magnitudes are larger than 2 but not divergent due to the nonzero minimums of the mean molecular number. Our results imply that the initial sub-Poissonian statistics of $g_b^{(2)}$ is transformed into a super-Poissonian, and thereby a superchaotic molecular pulse can be realized at certain values of time. The second-order coherence functions of the atomic field are also plotted in Fig. 2 (the red lines). We find that super-Poissonian statistics of $g_a^{(2)}$ is fully suppressed for $\Lambda \neq 0$. More specifically, the intensity correlation function of the atomic field oscillates between sub-Poissonian and super-Poissonian with very small amplitude. Further increase of Λ, such as to 1.6971, $g_a^{(2)}<1$, which implies that the atomic field is always in sub-Poissonian. The reduction of the super-Poissonian of atomic field to sub-Poissonian comes also from the suppression of spontaneous atomic emission. The increase of Λ dominates the contribution of the coherent stimulated process.

Finally, we study the joint quantum statistical properties of the atoms and the molecules, defined by

$$g_{ab}^{(2)} = g_{ba}^{(2)} \equiv \frac{\langle \hat{n}_a \hat{n}_b \rangle}{\langle \hat{n}_a \rangle \langle \hat{n}_b \rangle}. \quad (8)$$

We find that $g_{ab}^{(2)}(\tau)$ is always less than 1. Physically, such a result means that the atoms and the molecules do not tend to be created simultaneously, i.e., the antibunching between the two field modes. Similar results have been addressed for optical field in the two-photon down-conversion process [18].

In summary, we have studied the role of nonlinearity on quantum dynamics and statistical properties of the AMBEC. The former problem is related to the atom-molecule conversion rate. We have shown that even in the simplest two-mode model without "rogue photodissociation" [27], the conversion can be strongly settled down due to the nonlinear analog of macroscopic self-trapping effect. We also studied statistical properties of the AMBEC by calculating the intensity correlation functions numerically. Our results show that the nonlinearity Λ in the stable region can modify both $g_a^{(2)}$ and $g_b^{(2)}$ significantly. The initial sub-Poissonian molecular field is transformed into superchaotic molecular pulses, whereas the atomic field remains in sub-Poissonian region. Our results also show that the joint quantum statistical properties of the atoms and the molecules always exhibit antibunching.

It should be mentioned that Prataviera *et al.* [28] have studied higher-order mutual coherence of two chemically different species: the light field and the matter field. Their scheme make use of the combination of the traditional absorption photodetector and a matter-wave detector relied on photoionization of the atoms. Similarly, photoionization of the diatomic molecules can be proposed in the atom-molecule photoassociation experiments to measure the molecules and their statistical properties directly.

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[1] P. D. Drummond, K. V. Kheruntsyan, and H. He, Phys. Rev. Lett. **81**, 3055 (1998).
[2] E. Timmermans *et al.*, Phys. Rev. Lett. **83**, 2691 (1999); Phys. Rep. **315**, 199 (1999).
[3] F. A. van Abeelen and B. J. Verhaar, Phys. Rev. Lett. **83**, 1550 (1999).
[4] R. Wynar *et al.*, Science **287**, 1016 (2000).
[5] J. Javanainen and M. Mackie, Phys. Rev. A **59**, R3186 (1999).
[6] D. J. Heinzen, R. Wynar, P. D. Drummond, and K. V. Kheruntsyan, Phys. Rev. Lett. **84**, 5029 (2000).
[7] E. A. Donley *et al.*, Nature **417**, 529 (2002).
[8] N. R. Claussen, S. J. J. M. F. Kokkelmans, S. T. Thanpson, E. A. Donley, E. Hodby, and C. E. Wieman, Phys. Rev. A **67**, 060701(R) (2003).
[9] D. S. Hall, M. R. Matthews, C. E. Wieman, and E. A. Correll, Phys. Rev. Lett. **81**, 1543 (1998).
[10] F. Minardi, C. Fort, P. Maddaloni, M. Modugno, and M. Inguscio, Phys. Rev. Lett. **87**, 170401 (2001).
[11] M. J. Holland, J. Park, and R. Walser, Phys. Rev. Lett. **86**, 1915 (2001);S. J. J. M. F. Kokkelmans and M. J. Holland, *ibid.* **89**, 180401 (2002).
[12] E. G. M. van Kempen, S. J. J. M. F. Kokkelmans, D. J. Heinzen, and B. J. Verhaar, Phys. Rev. Lett. **88**, 093201 (2002).
[13] T. Köhler, T. Gasenzer, and K. Burnett, Phys. Rev. A **67**, 013601 (2003).
[14] R. A. Duine and H. T. C. Stoof, Phys. Rev. Lett. **91**, 150405 (2003).
[15] A. Vardi V. A. Yurovsky, and J. R. Anglin, Phys. Rev. A **64**, 063611 (2001).
[16] B. J. Cusack T. J. Alexander, E. A. Ostrovskaya, and Y. S. Kivshar, Phys. Rev. A **65**, 013609 (2001).
[17] M. K. Olsen and L. I. Plimak, Phys. Rev. A **68**, 031603(R) (2003).
[18] G. Drobný and I. Jex, Phys. Rev. A **45**, 1816 (1992); **45**, 4897 (1992).
[19] M. Koštrun *et al.*, Phys. Rev. A **62**, 063616 (2000).
[20] A. Smerzi, S. Fantoni, S. Giovanazzi, and S. R. Shenoy, Phys. Rev. Lett. **79**, 4950 (1997).
[21] Y. Wu and R. Côté, Phys. Rev. A **65**, 053603 (2002); Y. Wu, and C. P. Sun, Commun. Theor. Phys. **38**, 675 (2002).
[22] G. P. Agrawal, *Nonlinear Fiber Optics* (Academic Press, San Diego, 2001).
[23] A. Ishkhanyan, G. P. Chernikov, and H. Nakamura, Phys. Rev. A **70**, 053611 (2004).
[24] D. Meiser and P. Meystre, Phys. Rev. Lett. **94**, 093001 (2005).
[25] D. F. Walls and G. J. Milburn, *Quantum Optics* (Springer-Verlag, Berlin, 1994).
[26] U. V. Poulsen and K. Mølmer, Phys. Rev. A **63**, 023604 (2001).
[27] J. Javanainen and M. Mackie, Phys. Rev. Lett. **88**, 090403 (2002).
[28] G. A. Prataviera, E. V. Goldstein, and P. Meystre, Phys. Rev. A **60**, 4846 (1999).