Quantum state transfer from light to molecules via coherent two-color photo-association in an atomic Bose-Einstein condensate

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Abstract. By using a quantized input light, we theoretically revisit the coherent two-color photo-association process in an atomic Bose-Einstein condensate. Under the single-mode approximations, we show two interesting regimes of the light transmission and the molecular generation. The quantum state transfer from light to molecules is exhibited, without or with the depletion of trapped atoms.

Since the remarkable realizations of Bose-Einstein condensates (BEC) in cold dilute atomic gases, many novel properties of the macroscopic quantum degenerate gases have been experimentally exhibited or theoretically predicted [1]. Recently, by creating a quantum degenerate molecular gases via a magnetic Feshbach resonance [2,3] or an optical photo-association (PA) [4,5] in an atomic BEC, the appealing physical properties of the formed atom-molecule mixtures have attracted much interests as a whole new dimension in the study of ultracold atomic physics and even atom optics [2–5]. The Bose-enhanced coherent PA process was also termed as superchemistry since it is completely beyond the classical Arhenius chemical kinetics in such an ultralow temperature of the μK range [6]. Although many novel quantum effects are predicted for the coupled atomic-molecular condensates, such as the molecular damping due to the quantum noise terms [6], the essential features of the current PA experiments can be well described by the single-mode or even mean-field approaches, such as the famous experiment of coherent two-color PA experiment of Winkler et al. [4]. Hence the extremely complicated methods fully taking into account of the dissipated effects are *not* practically needed.

On the other hand, there also have been many interests recently in making and exploring the new applications of a coherent atomic beam or an atom laser [7]. Most recently, Haine et al. introduced a scheme to generate controllable atom-light entanglement by using a Raman atom laser system [8]. The key point of their scheme is to use an input

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squeezed light instead of a classical light [9]. Therefore an interesting question may arise: is it possible to realize the quantum control of molecule-light system or, as the first step, a non-classical molecule laser? The possible novel applications of a molecule laser can be expected in, e.g., the precise matter-wave interferometry technique [10].

The previous literatures on the coherent PA process are all focused on the stable and higher atom-molecule conversions by treating the external optical fields as the classical [4–6,11]. In this paper, by considering a quantized light as the weak PA field, we study the quantum dynamics and statistics of coherent molecular output via two-color PA in an atomic condensate. The basic physical assumption is that one photon is encoded onto two atoms to form a diatomic molecule, and then the quantum state of the photons are converted onto the created stable molecules. As we described above, the essential features of the current PA experiments can be well described by the single-mode or even mean-field approaches, we use the simple single-mode model to treat this photon-atom-molecule system instead of a fully analysis. We find two dynamical regimes, the optical transmission and the molecular formation, and for the later, the quantum state transfer from light to molecules can be observed, which indicates a possible way for the quantum control of the molecule-light system purely by an optical method.

Turning to the situation of Figure 1, we assume for simplicity that large number of Bose-condensed atoms are initially prepared in a magnetic trap when a strong control laser field with slowly varying Rabi frequency $\Omega(t)$ is applied. The PA is described by a slowly varying single-mode
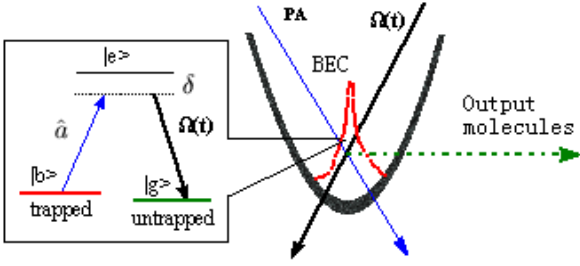
Fig. 1. (Color online) Schematic figure of the coherent two-color photo-association by using a quantized input light. The atoms are initially condensed in the trapped state $|b\rangle$. The free-quasi-bound-bound transitions and the momentum kick [8] are induced by two lasers: an input light $\hat{a}(t)$ with Rabi frequency ϵ and a classical control light with Rabi frequency $\Omega(t)$. δ is the intermediate detuning from the excited molecular state.

operator $\hat{a}(t)$, which induces two atoms to form one quasibound molecule in the exited state e. This state mediates the out-coupling Raman transitions and then the stable molecules are created in the untrapped state g due to the optical momentum kick [8]. Since the particles interactions can be tuned by using a Feshbach resonance [12,13] and goes to zero for a sufficiently *dilute* condensate, we ignore it here for the present purpose. In fact, the impacts of non-linear particles interactions on the dynamics of the output coherent matter waves have been well studied in the literatures [14,15]. In the second quantized notation, boson annihilation operators for the trapped atoms and the molecules in two states are denoted by $\hat{b}$, $\hat{e}$ and $\hat{g}$, respectively. By focusing on the different modes couplings, the effective Hamiltonian of this photon-atom-molecule system is then ($\hbar = 1$)

$$\hat{H}_4 = \delta\hat{e}^\dagger\hat{e} + \frac{1}{2}(\epsilon\hat{e}^\dagger\hat{b}\hat{b}\hat{a} + \epsilon^*\hat{a}^\dagger\hat{b}^\dagger\hat{b}^\dagger\hat{e})$$
$$+[\Omega(t)\hat{g}^\dagger\hat{e} + \Omega^*(t)\hat{e}^\dagger\hat{g}], \quad (1)$$

where δ is the intermediate detuning and $\hat{a}$ denotes the quantized input light. Note that we assume here the two-photon resonance (hence without the $\hat{g}^\dagger\hat{g}$ term) and also neglect another free-energy term $\hat{a}^\dagger\hat{a}$ since it does not have any essential impact on the main physical results of our model [9,24]. This model, comparing with that used by Winkler et al. to describe their recent PA experiment [4], only have the new feature of using an input light of quantum nature, and the incoherent process of the excited-state molecular damping or the rogue molecular dissociations are also ignored here [11]. And the weak PA field condition $\epsilon \ll \Omega$ can safely avoid any heating effects in the PA. Obviously we have: $\hat{b}^\dagger\hat{b} + 2(\hat{e}^\dagger\hat{e} + \hat{g}^\dagger\hat{g}) \equiv N_0$ with N_0 being the total number for a condensate of all atoms or twice the total molecules, indicating the stationary quantity under the time evolution of the system.

The roles of atomic association in the two-color PA has been studied extensively [4–6,11], even in an optical cavity [16]. Here, by using a quantized input light, we firstly consider the quantum dynamics of the system under the Bogoliubov approximation, i.e., by neglecting the atomic

depletions and treat $\hat{b}$ as a *c*-number b_0 with $|b_0|^2 = N_0$. The general depleted case will be treated later. It should be mentioned that this atomic state is assumed to be in a Glauber coherent state instead of a Fock state. Hence the reduced Hamiltonian describes nothing but a simple linear three-state system for which the perfect quantum state transfer is easily expected. In fact, by using the unitary transformation

$$U = \begin{pmatrix} \sin\theta_1\sin\theta_2 & \cos\theta_1 & \sin\theta_1\cos\theta_2 \\ \cos\theta_2 & 0 & -\sin\theta_2 \\ \cos\theta_1\sin\theta_2 & -\sin\theta_1 & \cos\theta_1\cos\theta_2 \end{pmatrix}, \quad (|U| = -1) \tag{2}$$

with the mixing angles defined by $\tan\theta_1(t) = \omega/\Omega(t)$, $\tan 2\theta_2(t) = 2\lambda(t)/\delta$ and $\lambda(t) = \sqrt{\omega^2 + \Omega^2(t)}$ with $\omega = \epsilon N_0/2$, the reduced Hamiltonian can be diagonalized as

$$\mathcal{H}_B \equiv U\hat{H}_B U^\dagger = \omega_+\hat{A}^\dagger\hat{A} + \omega_0\hat{E}^\dagger\hat{E} + \omega_-\hat{G}^\dagger\hat{G},$$
$$\omega_\pm = \frac{1}{2}(\delta \pm \sqrt{4\lambda^2(t) + \delta^2}); \quad \omega_0 = 0, \tag{3}$$

with $(\hat{a}, \hat{e}, \hat{g})^T = U(\hat{A}, \hat{E}, \hat{G})^T$. Thereby, based on an extending stimulated Raman adiabatic passage (STIRAP) method [17,18], the quantum transfer process can be clearly exhibited from the optical state to the molecular state. And this photon-molecule system then can serve as an another scenario of the well-known three-state optics (TSO) techniques which have been intensively studied in recent years for many systems, e.g., between different internal atomic quantum states [18], from the light to macroscopic atomic ensembles or propagating atomic beams [13,19–21] and vice versa. The new feature introduced here is that, in some sense, the photons are stored in the stable diatomic molecules, which may indicate an optical control of the molecule-light system or even their entanglement [8].

The essential mechanism of the TSO is the two-channel quantum interference effect which leads to an effectively unpopulated intermediate state for certain conditions [18]. This can be explicitly shown by calculating the three states populations. Here, by specifying the value of Ω to make two mixing angles as time-independent, we find for the intermediate state

$$N_e(t) \equiv \langle\hat{e}^\dagger(t)\hat{e}(t)\rangle$$
$$= \sin^2\theta_1\sin^2\theta_2\cos^2\theta_2[1 - \cos(\omega_+ - \omega_-)t]N_0, \tag{4}$$

which leads to $N_e(t) = 0$ for the conditions of $(\omega_+ - \omega_-)t = 2n\pi$ $(n = 0, 1, 2, ...)$. The detuning δ should be large generally in order to avoid the incoherent processes. Similarly we also can obtain

$$N_g(t) = \sin^2\theta_1\cos^2\theta_1[1 - \cos(\omega_+ t)]N_0. \tag{5}$$

Interestingly, we see that under the adiabatic approximations there can exist two dynamical regimes: (i) for $\omega_+\tau = 2n\pi$, we have an optical transmission regime in which the coherent PA in fact does *not* happen ($N_g(\tau) = 0$); (ii) only for $\omega_+\tau = (2n+1)\pi$, we can observe a coherent molecular

output, with an average flux of particles

$$N_g(\tau) = \left[\frac{2\omega\Omega}{\omega^2 + \Omega^2}\right]^2 N_a(0) \le N_a(0). \qquad (6)$$

And the complete quantum conversion from light to molecules happens for the conditions of $\omega \approx \Omega$ (or for the mixing angle, $\theta_1 = \pi/4$). This indicates the possibility to realize a nonclassical molecule laser. Although for a sample of trapped molecules the photons even can be coherently released again for the conditions of $\lambda\tau: (2n+1)\pi \to 2m\pi$, it is very difficult due to the realistic configurations in any actual experiment, such as the phase diffusion of trapped condensate, large inelastic atom-molecule scattering and other nonideal or dissipated factors especially in a dense condensate. This is the reason we consider a coherent molecular beam here instead of memory process in a trapped sample.

Now we proceed to study the quantum statistic of the output molecular field for an input PA light in a squeezed state [22]: $|\alpha\rangle_s = \hat{S}(\xi_1)|\alpha\rangle$, where the squeezed operator $\hat{S}(\xi_1) = \exp[\xi_1(\hat{a}^\dagger)^2 - \xi_1^*\hat{a}^2]$ with $\xi_1 = (r_1/2)e^{-i\phi_1}$, as a unitary transformation on the Glauber coherent state $|\alpha\rangle$ ($\alpha \equiv |\alpha|e^{i\varphi_1}$). The simplest case happens for the coherent molecular output regime, i.e., $\omega_+\tau = (2n+1)\pi$, and the Mandel's Q parameter for the output molecules is obtained as

$$Q_g^s(\tau) \equiv \frac{\langle\Delta\hat{N}_g^2(\tau)\rangle_s}{\langle\hat{N}_g(\tau)\rangle_s} - 1 = \langle\hat{N}_g(\tau)\rangle_s$$
$$= 4\cos^2\theta_1\sin^2\theta_2 Q_a(0) > 0,$$

with $Q_a(0) = \sinh^2 r_1$ and $\hat{g}(\tau) = -2\cos\theta_1\sin\theta_1\hat{a}(0) - \cos 2\theta_1\hat{g}(0)$. This indicates a complete quantum conversion (the super-Poisson distribution) from the input light to the output molecular fields. In addition, the squeezed angle of the input light also can affect the quantum statistics of the output molecular field. To see this clearly, we consider the quadrature squeezed coefficients [23]

$$\mathcal{S}_{ig} = \frac{\langle(\Delta\hat{\mathcal{G}}_i)^2\rangle - \frac{1}{2}|\langle[\hat{\mathcal{G}}_1, \hat{\mathcal{G}}_2]\rangle|}{\frac{1}{2}|\langle[\hat{\mathcal{G}}_1, \hat{\mathcal{G}}_2]\rangle|}, \qquad i = 1, 2 \qquad (7)$$

with $\hat{\mathcal{G}}_1 = \frac{1}{2}(\hat{g} + \hat{g}^\dagger)$, $\hat{\mathcal{G}}_2 = \frac{1}{2i}(\hat{g} - \hat{g}^\dagger)$. For the most interested case: $\omega_+\tau = (2n+1)\pi$, we can obtain

$$\mathcal{S}_{1g,2g}^{\xi_a}(\tau) = 2\sin^2(2\theta_1)\sinh r_1(\sinh r_1 \mp \cosh r_1\cos\phi_1),$$
$$(8)$$

which indicates a squeezed-angle-dependent molecular squeezed effect ($r_1 > 0$): (i) for $\phi_1 = 2n\pi$ ($n = 0, 1, 2, ...$), we have $\mathcal{S}_{1g} = 4\sin^2\theta_1\cos^2\theta_1(e^{-2r_1} - 1) < 0$ and $\mathcal{S}_{2g} = 4\sin^2\theta_1\cos^2\theta_1(e^{2r_1} - 1) > 0$, which means that the component $\mathcal{S}_{1g}$ is squeezed; but for $\phi_1 = (2n+1)\pi$, we have $\mathcal{S}_{1g} = \mathcal{S}_{2g}(\phi_1 = 2n\pi) > 0$ and $\mathcal{S}_{2g} = \mathcal{S}_{1g}(\phi_1 = 2n\pi) < 0$, which means that the quantum squeezing transfers to $\mathcal{S}_{1g}$ component; (ii) for $\phi_1 = (n+1/2)\pi$, we have $\mathcal{S}_{1g} = \mathcal{S}_{2g} > 0$, a squeezing-free effect for the output molecules [24].

In order to analytically study the impacts of the depletions of trapped condensate on the coherence of the output

molecular field, we firstly write the Heisenberg equations of motion of the excited molecules from equation (1) and, by assuming $|\delta|$ as the largest evolution parameter [11] in the system, we have: $\hat{e}/\delta = 0$, or $\hat{e} \simeq -(\epsilon\hat{b}\hat{b}\hat{a} + \Omega\hat{g})/\delta$. Substituting this into equation (1) to adiabatically eliminate the quasi-bound molecular state, yields the effective Hamiltonian as the following

$$\hat{H}_3 = -\Gamma\hat{g}^\dagger\hat{g} - \mu(\hat{a}\hat{a}^\dagger\hat{b}\hat{b}\hat{b}^\dagger\hat{b}^\dagger + H.c.) - \chi(\hat{g}^\dagger\hat{b}\hat{b}\hat{a} + H.c.), \quad (9)$$

where $\Gamma = 2\Omega^2/\delta$, $\mu = \epsilon^2/\delta$ and $\chi = 2\Omega\epsilon/\delta$. Now we choose $\Omega \gg \epsilon$, or $\chi \gg \mu$, then it would be good enough to consider the third term in equation (9) as the main couplings of the system. This observation greatly simplifies our analysis and the Heisenberg equations of motion for the three different states can be written as

$$i\frac{\partial}{\partial t}\hat{a} = -\chi\hat{b}^\dagger\hat{b}^\dagger\hat{g}, \quad i\frac{\partial}{\partial t}\hat{b} = -2\chi\hat{a}^\dagger\hat{b}^\dagger\hat{g},$$
$$i\frac{\partial}{\partial t}\hat{g} = -\Gamma\hat{g} - \chi\hat{b}\hat{b}\hat{a}. \qquad (10)$$

The analytical solutions of these equations can be obtained in the second-order approximation of the evolution time as the following simple forms:

$$\delta\hat{a}(t) = it\chi\hat{b}_0^\dagger\hat{b}_0^\dagger\hat{g}_0 + t^2\chi^2\hat{a}_0(2\hat{N}_{b0}\hat{N}_{g0} - \hat{M}_{b0}),$$
$$\delta\hat{b}(t) = 2it\chi\hat{a}_0^\dagger\hat{b}_0^\dagger\hat{g}_0 + t^2\chi^2[\hat{N}_{b0}\hat{b}_0(\hat{N}_{g0} - \hat{N}_{a0})$$
$$+ 2\hat{b}_0\hat{N}_{g0}(\hat{N}_{a0} + 1)],$$
$$\delta\hat{g}(t) = it\chi\hat{b}_0\hat{b}_0\hat{a}_0 - t^2\chi^2\hat{g}_0[2\hat{N}_{a0}(\hat{N}_{b0} + 1)$$
$$+ \hat{M}_{b0} + 2\hat{N}_{b0} + 1], \qquad (11)$$

where $\delta\hat{K}(t) \equiv \hat{K}(t) - \hat{K}_0$, $\hat{N}_{K0} = \hat{K}_0^\dagger\hat{K}_0$, $\hat{M}_{b0} = \hat{b}_0^\dagger\hat{b}_0^\dagger\hat{b}_0\hat{b}_0/2$ and $\hat{K}_0 = \hat{a}_0, \hat{b}_0, \hat{g}_0$. Therefore, firstly by using a coherent factorized structure of the initial state of the system, i.e., $|in\rangle = |\alpha\rangle_a|\mu\rangle_b|0\rangle_g$, with $\hat{b}|\mu\rangle = |\mu|e^{i\varphi_2}|\mu\rangle$, the squeezed coefficients for the output optical and molecular states are obtained as

$$\begin{pmatrix} S_{1a}(t) \\ S_{2a}(t) \end{pmatrix} = 3|\alpha|^2(1 - |\mu|^2\chi^2t^2)\begin{pmatrix} \cos^2\varphi_1 \\ \sin^2\varphi_1 \end{pmatrix}, \qquad (12)$$

and

$$\begin{pmatrix} S_{1g}(t) \\ S_{2g}(t) \end{pmatrix} = 3|\alpha|^2(1 - |\alpha|^2|\mu|^2\chi^2t^2)\begin{bmatrix} \sin^2(\varphi_1 + 2\varphi_2) \\ \cos^2(\varphi_1 + 2\varphi_2) \end{bmatrix}. \qquad (13)$$

This means that there is *no* squeezing for these fields even with the depletions of the trapped atoms. For the trapped atoms, however, the dynamical quadrature-squeezed effect always happens except for the specific case of $|\cos\varphi_2| = |\sin\varphi_2|$.

To examine the quantum state transfer process in the depleted case, we now use an input light in squeezed vacuum (with nonzero mean number of photons) [25] for a coherent atomic condensate in the magnetic trap, and the molecular squeezed coefficients can be obtained as

$$\mathcal{S}_{1g,2g}^{\xi_a}(t) = 2|\mu|^4\chi^2t^2\sinh r_1(\sinh r_1 \mp \cosh r_1\cos\phi_1),$$
$$(\varphi_{1,2} = 0) \quad (14)$$

from which, comparing with the simplest linear case, we can see that the squeezed behaviors for the output molecules are also dependent on the squeezed angle of the input light. However, if the initial atomic condensate is already prepared in a squeezed state by, for example, the nonlinear Kerr-type atomic collisions (see, e.g., Ref. [24]), but the input light is now in an ordinary Glauber coherent state, i.e., $|in\rangle = |\alpha\rangle_a |\mu_s\rangle_b |0\rangle_g$, $|\mu_s\rangle_b = \hat{S}(\xi_2)|\mu\rangle_b$, with $\hat{S}(\xi_2) = \exp[\xi_2(\hat{b}^\dagger)^2 - \xi_2^*\hat{b}^2]$, and $\xi_2 = (r_2/2)e^{-i\phi_2}$, then we get the different squeezed behaviors for the output molecular field as

$$\begin{pmatrix} \mathcal{S}_{1g}^{\xi_b}(t) \\ \mathcal{S}_{2g}^{\xi_b}(t) \end{pmatrix} = |\alpha|^2\chi^2 t^2 \sinh^2 r_2$$

$$\times \left[11\cosh^2 r_2 \begin{pmatrix} \sin^2\phi_2 \\ \cos^2\phi_2 \end{pmatrix} - 4 \right]. \quad (15)$$

Obviously, the squeezed effects really can appear except for the case of $|\cos\phi_2| = |\sin\phi_2|$. This may indicate a quantum transfer from the trapped atoms to the output molecules. However, it is more difficult for it to be observed in practice since the dynamical evolutions generally can affect the squeezed parameters of the atomic field itself (except for some special cases, e.g., $|\cos\varphi_2| = |\sin\varphi_2|$), and the initial detection of the atomic squeezed parameters also can be very challenging for current experiments of the ultracold atoms. Thereby, it is only accessible at present to consider the realization of the optical control of the output coherent molecular waves by using a dilute trapped condensate.

Finally we can study the mutual coherence of the output optical and molecular fields by calculating the second-order cross-correlation function $g_{ag}^{(2)}(t)$ [22]

$$g_{ag}^{(2)}(t) = \frac{\langle \hat{a}^\dagger(t)\hat{a}(t)\hat{g}^\dagger(t)\hat{g}(t)\rangle}{\langle \hat{a}^\dagger(t)\hat{a}(t)\rangle\langle \hat{g}^\dagger(t)\hat{g}(t)\rangle}. \quad (16)$$

Using $\langle \hat{a}^\dagger(t)\hat{a}(t)\hat{g}^\dagger(t)\hat{g}(t)\rangle = 2\chi^2 t^2 \langle \hat{N}_{a0}(\hat{N}_{a0}-1)\hat{M}_{b0}\rangle$, it is easily verified that this function is independent of the trapped atomic state and completely determined only by the input optical field. For the concrete examples, we can obtain the very simple results $g_{ag}^{(2)}(t) = 1 - 1/|\alpha|^2 < 1$ (anti-correlated states) for a coherent input light, and $g_{ag}^{(2)}(t) = 3$ (correlated states) for an input light in squeezed vacuum (with nonzero mean number of photons, see, e.g., reference [25] for the creation and properties of a light field in squeezed vacuum). This means that both the optical and molecular fields can exhibit the correlated or anti-correlated properties with or without the initial optical squeezing. Note that, these results hold only for the short-time limits and in this limits, there exists no violation of the Cauchy-Schwarz inequality (CSI) or nonlocal entanglement of the two output fields [22]. And it is interesting to further consider the dynamics of the system in a long time scale and the possibility of creating entangled molecule-light system via the quantum description of the two-color PA process.

In conclusion, we theoretically revisit the process of coherent two-color PA by using a quantized input light.

Under the single-mode approximations, we show that the quantum state transfer from light to molecules is possible without or with the atomic depletions. This indicates a possible optical control of the molecular fields. Of course, the intrinsic molecular collision in the output beam should be considered, which was shown for an atom laser also relevant to the squeezing effect [13]. The most serious simplification here, however, is a single-mode treatment of the light since our main purpose is about the possible mechanism of quantum conversion from light to molecules in this nonlinear system. Although this is suitable for current PA experiment conditions [4], a multi-mode or spatial structure analysis can be very significant for any quantum conversion process, by considering many nonideal factors like the phase diffusion of the condensate, optical damping and inelastic two-body scattering. Also, it is difficult to treat the problem of quantizing a PA light due to the complex microscopic many-body dynamics [26], and our simple model here should be taken only as the first step or qualitative probe of the possible features and applications of this novel nonlinear molecule-light system.

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