Quantum atom-heteronuclear molecule dark state: Role of population imbalance

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Recently, the finite-number effect of initial atoms in coherent atom-molecule conversion was investigated by Zhao *et al.* [Phys. Rev. Lett. **101**, 010401 (2008)]. Here, by extending to the atom-heteronuclear molecule dark state, we find that the initial populations imbalance of the atoms plays a significant role in quantum conversion rate and adiabatic fidelity. In particular, even for the finite total number of *imbalanced* two-species atoms, the mean-field conversion rate, contrary to the general belief, still can be remarkably close to the exact quantum results.

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The concept of a coherent population trapping (CPT) state or dark state is well known in quantum optics with many important applications [1]. Recently, an intriguing ultracold atom-molecule CPT dark state was experimentally observed by Winkler *et al.* [2] through coherent two-color photoassociation (PA)

$$A + A + \gamma_p \rightarrow A_2^*, \quad A_2^* \rightarrow A_2 + \gamma_d,$$

where A, A_2^*, or A_2, and $\gamma_{p,d}$ denote the entrance-channel atoms, the intermediate excited or the closed-channel molecules, and the pumping or dumping photons in counterintuitive pulse sequences. This process, also referred to as the stimulated Raman adiabatic passage (STIRAP) [3–6], has been studied quite extensively as an efficient technique for enhanced atom-molecule conversion rate. Various properties of this process were probed within the classical mean-field approach (MFA), such as the stability of the atom-molecule dark state [7], the formation of molecular matter-wave vortex [8], and the nonlinear adiabatic condition of the STIRAP [9].

Recently, Zhao *et al.* used a fully quantum approach to study the atom-molecule conversion system with a *finite* number N of initial atoms [10]. They found that the particles populations exhibit interesting quantum deviations from the MFA results, which turns out to be significant for the few-N cases (the classical MFA completely breaks down). The availability of exact quantum solutions also allows one to study the higher-order matter-wave statistical properties. We note that in the absence of the atom-molecule CPT dark state, quantum counting statistics were already proposed by Meiser and Meystre to diagnose the (coherent or chaotic) bosonic molecules created from *bosonic* or *fermionic* atoms [11].

In this Brief Report, we study the exact quantum solutions of the atom-hetermuclear molecule dark state by considering the two-color PA process

$$A + B + \gamma_p \rightarrow AB^*, \quad AB^* \rightarrow AB + \gamma_d,$$

here A, B denotes the entrance-channel atoms of different species AB^* and AB are the intermediate excited or the closed-channel ground-state molecules. Our purpose here is to find the possible role of initial populations imbalance $\mathcal{M} \neq 0$ of the two-species atoms in quantum conversion rate and second-order matter-wave correlations [12]. The main results of this work are (i) even for the finite total number of *imbalanced* two-species atoms, the mean-field conversion rate still can be remarkably close to the exact quantum results, a new feature which is impossible to exist in creating homonuclear dimers; (ii) for $N_a > N_b$, it is not the total particles number N but N_b which determines the molecular correlations; and (iii) the adiabatic fidelity can be improved by increasing the imbalance $\mathcal{M}$ (even with zero optical detuning or large N).

Quantum conversion rate with $\mathcal{M} \neq 0$.—Without any loss of generality, we take the Rabi frequency of pumping or dumping laser $\Omega_{p,d}$ to be real and positive whose phase factor can be absorbed by a global gauge transformation of the field operators [8]. Δ and δ are the single-photon and two-photon detunings of the frequencies of two lasers, respectively. Here, to compare with that of Ref. [10], we focus on the role of coherent atom-molecule-light couplings by ignoring the collisions of a dilute or Feshbach-resonance tuned gas [1], a safe approximation for short molecular lifetime [2,10]. In the interaction picture under the two-photon resonant condition, the Hamiltonian of this system can be written in the simplest level as ($\hbar = 1$)

$$\hat{H} = \Delta \hat{\psi}_e^\dagger \hat{\psi}_e + \tfrac{1}{2}(\Omega_p \hat{\psi}_e^\dagger \hat{\psi}_a \hat{\psi}_b + \Omega_d \hat{\psi}_e^\dagger \hat{\psi}_g + \text{H.c.}), \quad (1)$$

where $\hat{\psi}_i$ or $\hat{\psi}_i^\dagger$ is the bosonic annihilation or creation operator ($i = a,b,e,g$). The standard MFA is valid if the number of particles is significantly large and then the operators can be replaced by the c-numbers ψ_i or ψ_i^*. Then the mean-field dark state is computed

$$|D\rangle_{MF} = \left(\psi_a^0, \psi_b^0, \psi_e^0, \psi_g^0\right)^T = \left(\psi_a^0, \psi_b^0, 0, \psi_g^0\right)^T,$$

with

$$\left|\psi_a^0\right|^2 + \left|\psi_b^0\right|^2 + 2\left|\psi_g^0\right|^2 = N, \quad \left|\psi_a^0\right|^2 - \left|\psi_b^0\right|^2 = \mathcal{M} > 0,$$
$$2\left|\psi_{a,b}^0\right|^2 = -1/\chi^2 \pm \mathcal{M} + \sqrt{(N + 1/\chi^2)^2 - (N^2 - \mathcal{M}^2)},$$

and $\chi = \Omega_p/\Omega_d$. Clearly, by tuning $\chi(t)$, the atoms can be converted into the ground-state molecules without any population in the excited state $|e\rangle$. This picture is also true for *quantum* CPT state $|D\rangle_Q = |0\rangle_e |\Psi\rangle_{\mu\in(a,b,g)}$. From the eigenequation $\hat{H}|D\rangle_Q = 0$, we have

$$(\chi \hat{\psi}_a \hat{\psi}_b + \hat{\psi}_g)|\Psi\rangle_\mu = \hat{\psi}_g e^{\chi \hat{\psi}_g^\dagger \hat{\psi}_a \hat{\psi}_b} |\Psi\rangle_\mu = 0. \quad (2)$$

The four-mode quantum dark state is then

$$|D\rangle_Q = C(\chi, N, \mathcal{M}) e^{-\chi \hat{\psi}_g^\dagger \hat{\psi}_a \hat{\psi}_b} |N_a, N_b, 0, 0\rangle, \quad (3)$$

with the normalization factor $C(\chi, N, \mathcal{M})$ and the Fock-state basis $|N_a, N_b, N_e, N_g\rangle$. By expanding the exponential operator,

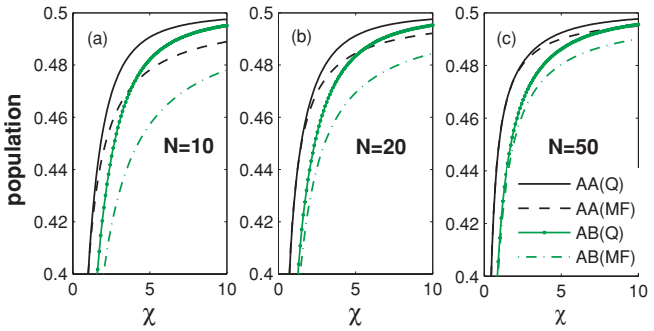

FIG. 1. (Color online) The populations of ground-state molecules A_2 or AB as the function of Rabi frequencies ratio χ, for different initial atomic number $N = 10, 20, 50$.

we have another form of the superposition state (i.e., $|D\rangle_Q = \sum c_n |N_a - n, N_b - n, 0, n\rangle$).

The comparisons between the quantum and the MFA results about the populations of closed-channel molecules A_2 or AB are shown in Fig. 1, for $\mathcal{M} = 0$ (but with different values of N). In this case, as one expected, quantum effects are significant in the few-N situations and the conversion rate is always *higher* than the mean-field results, for both heteronuclear and the homonulclear cases. We note that the quantum deviations are always *larger* in creating heteronuclear molecules AB.

Figure 2(a) shows the results for a small number $N = 10$ but $\mathcal{M} \neq 0$. Clearly, even for the finite total number of *imbalanced* two-species atoms, the mean-field conversion rate still can be remarkably close to the exact quantum results. This interesting new feature is impossible to be observed in creating homonuclear dimers. In Fig. 2(b), it also can be seen that for both large $N = 100$ and small $N = 10$, the mean-field and exact quantum lines are almost overlapped ($N_b = 4$ for both cases). In fact, for large N_a, this system is reduced to a linear one since the operator $\hat{\psi}_a$ can be replaced by a c-number. We note that the populations imbalance $\mathcal{M}$ also plays a significant role in the ground-state stability properties [13].

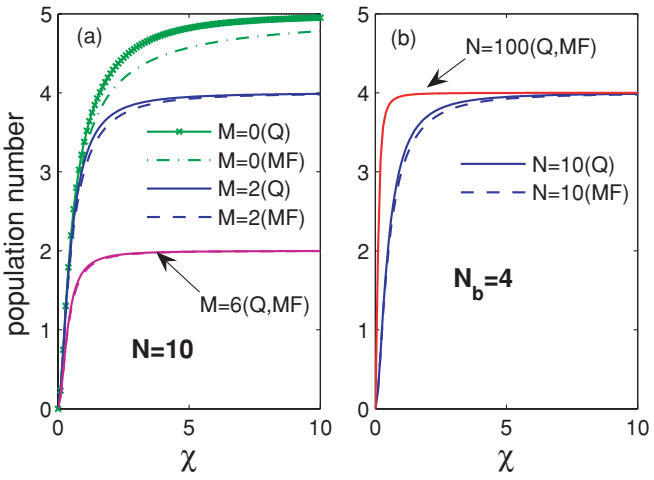

FIG. 2. (Color online) Panel (a) is the population number of molecules AB as the function of χ, for different initial number of N_b, at $N = 10$. Panel (b) adds a line of $N = 100, N_b = 4$. In all figures both the quantum and mean-field solution is presented.

Quantum correlations with $m \neq 0$.—The second-order correlation functions are defined as [11]

$$g_i^{(2)} = \frac{\langle \hat{\psi}_i^\dagger \hat{\psi}_i^\dagger \hat{\psi}_i \hat{\psi}_i \rangle}{\langle \hat{\psi}_i^\dagger \hat{\psi}_i \rangle^2} = \frac{\langle \hat{\psi}_i^\dagger \hat{\psi}_i^\dagger \hat{\psi}_i \hat{\psi}_i \rangle}{n_i^2}, \quad \mathcal{C}_{ig} = \frac{\langle \hat{n}_i \hat{n}_g \rangle}{n_i n_g}.$$

For $\mathcal{M} = 0$, we have confirmed that for large N the classical Poissonian distribution can be reached for both the homonuclear and the heteronuclear cases [11]. Nevertheless, we can also observe an unexpected and somewhat surprising result for the atoms A,

$$A_2 : g_a^{(2)} > 1 (\text{super-Poissonian}),$$
$$AB : g_a^{(2)} < 1 (\text{sub-Poissonian}),$$

for even particles number N. This indicates that quantum second-order coherence may be used to distinguish the bosonic heteronuclear and homonuclear molecules both created from *bosonic* atoms [11]. We emphasis that this interesting feature in the quantum regime has *not* been noticed in the previous literature [10,11].

For the general case $\mathcal{M} \neq 0$, we have

$$\lim_{\chi \to 0} g_{a,b}^{(2)} = 1 - \frac{1}{N_{a,b}}, \quad \lim_{\chi \to 0} g_{AB}^{(2)} = \frac{(N_a - 1)(N_b - 1)}{N_a N_b}, \tag{4}$$

and

$$\lim_{\chi \to \infty} g_a^{(2)} = 1 - \frac{1}{\mathcal{M}}, \quad \lim_{\chi \to \infty} g_{AB}^{(2)} = 1 - \frac{1}{N_b},$$
$$\lim_{\chi \to \infty} g_b^{(2)} = 1 - \frac{N + \mathcal{M} + 2}{(N - \mathcal{M})(\mathcal{M} + 2)}. \tag{5}$$

Figures 3(a) and 3(b) shows the numerical results for evolutions of quantum matter-wave correlations with a finite small number $N = 10$. We can observe that, by increasing the imbalance value $\mathcal{M}$, $g_a^{(2)}$ can be significantly increased and tends to the classical value $g_a^{(2)} \to 1$. In contrast, the nonclassical feature becomes more evident for the created molecules (i.e., $g_{AB}^{(2)} \to 0$). The most interesting result turns

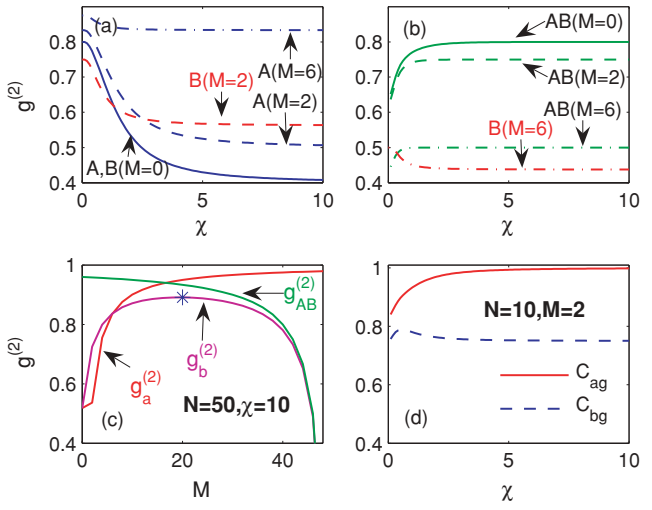

FIG. 3. (Color online) Quantum correlation functions $g^{(2)}$ (a–b) as a function of χ for $N = 10$, and (c) as a function of populations imbalance $\mathcal{M}$ for $N = 50, \chi = 10$; (d) the atom-molecule correlation $\mathcal{C}_{ag,bg}$ as a function of χ.

out to be that of $g_b^{(2)}$, which first increases to some maximum value for

$$\mathcal{M}_{\max} = \sqrt{2N(N+2)} - (N+2),$$

and then decreases to 0. This feature also can be seen in Fig. 3(c) for $N = 50$. Clearly, $\mathcal{M}_{\max} \sim 4$ for $N = 10$ and $\mathcal{M}_{\max} \sim 20$ for $N = 50$.

The role of populations imbalance also can be observed by probing the atom-molecule quantum correlations. In the two limits of χ,

$$\lim_{\chi\to 0} \mathcal{C}_{ag} = 1 - \frac{1}{N_a}, \quad \lim_{\chi\to\infty} \mathcal{C}_{ag} = 1,$$
$$\lim_{\chi\to 0} \mathcal{C}_{bg} = 1 - \frac{1}{N_b}, \quad \lim_{\chi\to\infty} \mathcal{C}_{bg} = 1 - \frac{1}{N_b} < 1. \tag{6}$$

The distinction between $\mathcal{C}_{ag}$ and $\mathcal{C}_{bg}$ is obvious, since one tends to the classical value 1 but the other tends to a nonclassical value relying on the initial atomic number. This feature, as numerically confirmed in Fig. 3(d), does *not* exist both in the homonuclear case or in the heteronuclear (balanced) case with $\mathcal{M} = 0$.

Adiabatic fidelity with $\mathcal{M} \neq 0$.—The exact quantum solutions also allows us to study the full energy spectrum and the adiabatic fidelity of the atom-heteronuclear molecule conversion. As shown in Fig. 4(a), we plot the eigenvalues of Hamiltonian (1) in a Fock-state basis as the function of Δ, where the solid or dotted lines represent the fully quantum or the MFA results. To make the figure more clear, here we set $N_a = 5$, $N_b = 3$, and $\Omega_{p,d} = 1$ (the nondegenerate MF spectrum has four lines but two of them overlap with the horizon line). All the lines increase monotonically with Δ, except for the zero-energy one (dark state). The quantum spectrum has *two* degeneracies for $\Delta = 0, \pm 0.7$, which allows the system to evolve from dark state to other states (adiabatic breakdown). Figure 4(b) shows the number of lines cross the zero-energy line at $\Delta = 0$, for larger N. For example, no such line for $N_b = 1$, but one for $N_b = 2,3$ and *two* for $N_b = 4,5$, indicating more possibilities to departure from the dark state. In our simulation, the existence of $\mathcal{M}$ makes the energy spectrum clearly separated.

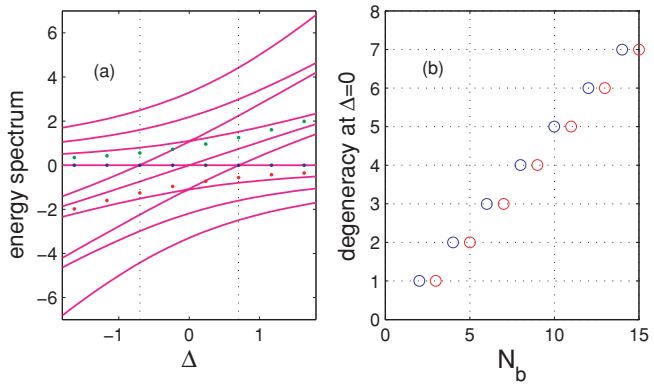

FIG. 4. (Color online) (a) The energy spectrum as the function of single-photon detuning Δ, the initial atomic numbers are $N_a = 5$, $N_b = 3$. (b) The degree of degeneracy at $\Delta = 0$ as the function of initial number N_b.

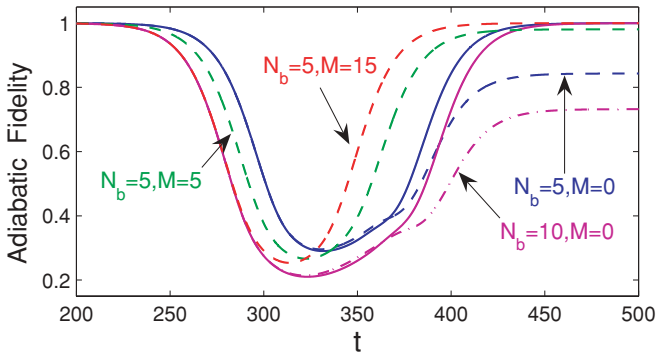

FIG. 5. (Color online) The adiabatic fidelity $|\langle\psi(t)|D(t)\rangle_Q|^2$ as the function of time, for $\Delta = 0$ (dashed lines, $N_b = 5$, $\mathcal{M} = 0,5,15$) and $\Delta = 0.5$ (indistinguishable solid lines for different values of N and $\mathcal{M}$). Other parameters: $\Omega_0 = 5$, $T = 100$, $t_d = 250$, and $t_p = 450$.

To confirm this point, we performed the simulations by choosing two Gaussian pulses (with width T, centered at $t_{p,d}$, $t_d < t_p$)

$$\Omega_{p,d}(t) = \Omega_0 e^{-(t-t_{p,d})^2/T^2}. \tag{7}$$

Figure 5 shows the adiabatic fidelity between $|\psi(t)\rangle$ and the instantaneous dark state $|D(t)\rangle_Q$. In the ideal case, the fidelity should be close to 1. However, for $\Delta = 0$, the system departures from $|D(t)\rangle_Q$ even for finite number of atoms N. This problem can be overcome either by choosing a larger detuning (i.e., $\Delta = 0.5$, see Fig. 5), or more interestingly, by choosing a larger populations imbalance [e.g., $\mathcal{M} = 15$, $\Delta = 0$ (with a fixed value of N_b)]. We note that larger total particles number N (with a fixed value of N_b) leads to an improved adiabatic fidelity. This is in contrast to that of the homonuclear system (the adiabatic fidelity is worse for larger N [10]). Clearly, the parameter $\mathcal{M}$ provides an additional freedom to control quantum dark-state evolutions of the system.

In conclusion, we have constructed an exact quantum atom-heteronuclear molecule dark state and confirmed that for nonzero two-species populations imbalance $\mathcal{M}$, the quantum atom-molecule dark state possesses some new features that are absent for the homonuclear case: (i) even for finite total number of atoms, the mean-field conversion rate still can be remarkably close to the exact quantum results; (ii) for $N_a > N_b$, it is not the total particles number N but N_b which determines the molecular correlations; and (iii) the adiabatic fidelity can be improved by increasing the imbalance $\mathcal{M}$ (with a fixed N_b). This means that, in contrast to the homonuclear case, both the total particles number N and the populations imbalance $\mathcal{M}$ play the important roles in controlling quantum dynamics of coherent atom-molecule system. In our future work, we plan to study the exact quantum solutions of the laser-catalyzed bimolecular reaction [14–16]. We also plan to study the role of populations imbalance in creating fermionic dimers from a two-species Bose-Fermi mixture [17] or molecular trimers from a three-species atomic mixture [18].

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