Improvement of conversion efficiency of atom-molecule Bose-Einstein condensate

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We investigate the stimulated Raman adiabatic passage in two-color photoassociation for a atom-molecule Bose-Einstein condensate. By applying two time-varying Guassian laser pulses that fulfill generalized two-photon resonance condition, we obtain highly efficient atom-molecule conversion. The efficiency depends on the free-bound detuning and the delay time between the two pulses. By adjusting the parameters optimally, we achieve 92% conversion efficiency.

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I. INTRODUCTION

Possible realization of molecular Bose-Einstein condensate (BEC) from atomic condensate has attracted wide attentions [1, 2, 3, 4, 5, 6]. A pair of atoms in open channel can be coupled to a weakly bound molecule by using either photoassociation process [2, 3, 4] or Feshbach resonance method [3, 4]. Such a coherent conversion of two chemically different species is of fundamental physical interests, and can be regarded as matter-wave analog of second harmonic generation. It was shown that a direct conversion via one-color free-bound photoassociation (Feshbach resonance) creates the molecules in highly excited electronic (vibrational) levels. Moreover, statistical property of the molecules exhibits sub-Poissonian only at initial stage [7], and it will be transformed into super-Poissonian due to inherent nonlinearity of the BEC [8].

To overcome all these problems, the second laser field is employed to couple the excited molecules to long-live ground-state molecules. Two-color free-bound-bound mechanism based on the stimulated Raman adiabatic passage (the STIRAP) was proposed as a promising route to achieve the high efficiency of atom-molecule conversion [9, 10, 11, 12, 13, 14, 15]. The STIRAP relies on the coherent population trapping (CPT) state, which is a superposition of two long-lived levels which decouple from the excited levels [10, 11, 12]. In the atom-molecule BEC, the CPT state (also called as the dark state) is a superposition of the atomic and molecular ground states, and has been observed in recent experiments [16, 17, 18].

In principle, one can realize a complete and reversible atom-molecule conversion as long as the system is kept in the CPT state. However, it was shown that the mean-field collisions set a limit to conversion efficiency by about 46% [12, 13].

In Ref. [13], Ling et al. recognized the CPT condition in the linear atomic system differs from those in the atom-molecule BEC. They found that high conversion efficiency, namely about 83%, is achievable provided that the CPT condition is satisfied during whole time evolution. In their work a single time-varying laser pulse is used for the bound-bound transition, while a constant Feshbach magnetic field is applied in the free-bound transition. The Feshbach resonance method was thought to present relatively large free-bound coupling than that of the photoassociation. However, both methods are actually equivalent in theoretical point of view. In addition, we note that the highest-conversion efficiency in their work appears near the exact Feshbach resonant, which unfortunately leads to strong atomic loss.

In this paper, we study the STIRAP in the atom-molecule BEC by using the frequency modulation scheme [15], where two time-varying Guassian laser pulses are employed. Our scheme has its advantage to provide additional parameters to optimize the conversion efficiency so that an extreme high conversion efficiency, namely about 92%, can be obtained. Moreover, the highest conversion occurs at far off-detuned region for the free-bound transition, which avoids the loss of atomic BEC in the Feshbach resonance version of the STIRAP. Our paper is organized as follows. In Sec. II, we present a simple three-level model for the atom-molecule BEC system, and investigate the effects of the mean-field collisions on the CPT condition and dynamical instability. In Sec. III, we present our numerical results and some discussions. Finally, a summary of our paper is presented.

II. THEORETICAL MODEL

We consider a Λ-type atom-molecule system, as shown in Fig. 1 (a), where a pair of free colliding atoms in level |a⟩ is converted to a bound molecule in level |g⟩ via an intermediate molecular level |b⟩. The free-bound transition |a⟩ → |b⟩ is characterized by Rabi frequency Ω1, while the coupling strength Ω2 corresponds to the bound-bound transition |b⟩ → |g⟩. In practice, the atoms in level |a⟩ is a BEC gas with density denoted as ρ. Due to the Bose enhancement, the free-bound Rabi frequency Ω1 ∝ √(ρ) [12].

The detuning for the free-bound and bound-bound transitions are denoted as ∆1 = ωb − 2ωa − ν1 and ∆2 = ωb − ωg − ν2, respectively, with νσ the central frequencies of the Laser fields σ = 1, 2. Thus the two-photon (Raman) detuning is δ = ∆1 − ∆2 = (ωg − 2ωa) − (ν1 − ν2).

Following Refs. [9, 10, 11, 12, 13, 14, 15], we assume that the atoms and the molecules can be described solely by three bosonic fields. For an atom-molecule BEC sys-
tem with high density, we employ further the standard mean-field treatments, i.e., replacing the field operators by the normalized field amplitudes $a$, $b$, and $g$. From Heisenberg equation of the field operators, we obtain the following coupled equations:

$$i\dot{a} = (\Lambda_{aa}|a|^2 + \Lambda_{ag}|g|^2)a - \Omega_1a^*b,$$  \hspace{1cm} (1)

$$i\dot{b} = (\Delta_i - i\gamma_b/2)b - \frac{1}{2}(\Omega_1a^2 + \Omega_2g),$$  \hspace{1cm} (2)

$$i\dot{g} = (\Lambda_{ag}|a|^2 + \Lambda_{gg}|g|^2)g + \delta g - \frac{1}{2}\Omega_2b,$$  \hspace{1cm} (3)

where $\Lambda_{ij} = \rho U_{ij}$ represent two-body collision rate with $U_{ii} = 4\pi \hbar a_i/m_i$ and $U_{ij} = U_{ji} = 2\pi \hbar a_i$ for $i \neq j$. Here $a_{*ij}$ are s-wave scattering lengths, and $\mu_{ij}$ are the reduced masses between states $i$ and $j$. We neglect mean-field interactions related to the excited molecular state due to small occupation of this state [12]. The decay rate $\gamma_b$ is added phenomenologically to take into account the loss of the excited molecules.

In the absence of collisions $\Lambda_{ij} = 0$, Eq. (3) is just the ordinary two-photon resonance condition. For two Gaussian laser pulses in a counterintuitive order, $\delta$ varies as shown in the inset of Fig. 3.

The mean-field collisions result in dynamical instability of the CPT solutions. To investigate it, we employ the standard perturbation theory, i.e., adding small perturbations to the CPT solutions as $\alpha = (a(0) + \delta a)e^{-i\omega t}$, where $\delta a = \eta_a e^{-i\omega t} + \nu_{\alpha} e^{i\omega t}$ (for $\alpha = a, b, g$), and $\omega$ is the eigenfrequency of the perturbations. The dynamical instability of the CPT solution takes place when the eigenfrequency is imaginary. In this case, a weak perturbation may induce an exponential growth of the excited molecules, which in turn lowers remarkably the fraction of ground-state molecules. After some tedious calculations, one can obtain the eigenfrequency of the perturbations.

FIG. 2: The dynamically unstable regions in the $(\Omega_2/\Omega_1, \Delta_1/\Omega_1)$ plane. Other parameters are taken as (in unit of $\Omega_1 = 2.1$ MHz): $\Lambda_{aa} = 1.02 \times 10^{-2}$, $\Lambda_{ag} = -1.32 \times 10^{-2}$ and $\Lambda_{gg} = 0.51 \times 10^{-2}$. Inset: the unstable region II near $\Delta_1 = 0$.

It was shown that in a linear $\Lambda$ three-level system, the CPT state occurs if the two-photon resonance condition $\delta = 0$ is satisfied [10, 17, 18]. However, the mean-field collisions probably modify the CPT condition [15]. To obtain the new condition, we neglect temporarily the particle losses, which implies the conservation of particle number i.e. $|a|^2 + |b|^2 + |g|^2 = 1$. By introducing $\alpha = a(0)e^{-i\omega t}$ (for $\alpha = a, b, g$) with the chemical potentials $\mu_{a, b, g} = 2\mu_a$, we can obtain the stable solution of Eq. (3). The CPT steady state corresponds to $b(0) = 0$, and

$$|a(0)|^2 = 1 - 2|g(0)|^2 = \frac{2}{1 + \sqrt{1 + 8(\Omega_1/\Omega_2)^2}},$$  \hspace{1cm} (4)

which depends only explicitly on the two coupling strengths. For $\Omega_1/\Omega_2 \to 0$, $|a(0)|^2 = 1$; for $\Omega_1/\Omega_2 \to \infty$, $|g(0)|^2 = 1/2$. The dotted lines of Fig. 3 present the CPT solutions $|a(0)|^2$ and $|g(0)|^2$ for two time-varying laser fields (see Eq. 5). The CPT solution appears under generalized two-photon resonance condition [13]:

$$\delta = (2\Lambda_{aa} - \Lambda_{ag})|a(0)|^2 + (2\Lambda_{ag} - \Lambda_{gg})|g(0)|^2.$$  \hspace{1cm} (5)

FIG. 1: (a) Schematic picture of three-level atom-molecule system; (b) Rabi frequencies as a function of time, generated by counterintuitive order of the two laser pulses.

FIG. 3: The dynamically unstable regions in the $(\Omega_2/\Omega_1, \Delta_1/\Omega_1)$ plane. Other parameters are taken as (in unit of $\Omega_1 = 2.1$ MHz): $\Lambda_{aa} = 1.02 \times 10^{-2}$, $\Lambda_{ag} = -1.32 \times 10^{-2}$ and $\Lambda_{gg} = 0.51 \times 10^{-2}$. Inset: the unstable region II near $\Delta_1 = 0$.

In Fig. 2, the instability regions calculated numerically are shown in the $(\Delta_1, \Omega_2)$ plane, where both $\Delta_1$ and $\Omega_2$ are in unit of $\Omega_1$. Similar to the previous work [13], there are two unstable areas: the area I takes place only in positive $\Delta_1$ (i.e., red-detuned region) for small $\Omega_2/\Omega_1$; while unstable region II is a thin area centered at $\Delta_1 = \Lambda_{ag}$ for small $\Omega_2/\Omega_1$. Unlike the previous work [13], part of area II in the small $\Omega_2/\Omega_1$ limit appears in the blue-detuned region due to negative $\Lambda_{ag}$ for $^{87}$Rb atom-molecule system.

III. NUMERICAL RESULTS

The atom-molecule conversion efficiency is defined as $\eta = 2|g(\infty)|^2$, which measures how many atoms in level
|a⟩ converted to the stable molecule state |g⟩. It was shown the mean-field collisions set a limit to conversion efficiency, and the highest conversion efficiency is thus about 46% \(^{12}\). Ling et al. found however that higher efficiency (83\%) can be obtained as long as the CPT condition, Eq. \((5)\), is satisfied during whole time evolution \(^{15}\). Following their scheme, we restudied the STIRAP in the atom-molecule BEC system. Two time-varying laser pulses are employed in our model, as shown in Fig. 1 (b). The Rabi frequencies are taken as

\[
\Omega_a(t) = \Omega_0 \exp[-(t-t_a)^2/\tau^2],
\]

with \(\sigma = 1, 2\). To fulfill the adiabatic condition, we take \(\Omega_0/\sigma = 5 \times 10^3\). The two Gaussian pulses are designed in counterintuitive order: the bound-bound laser field centered at \(t_2\) is applied earlier than the free-bound laser field centered at \(t_1\). The delay time between two pulses is defined as \(T = t_1 - t_2\). Compared with previous work \(^{15}\), our scheme provides additional parameters \(\Delta_1\) and \(T\), which is useful for optimizing the atom-molecule conversion.

Following Refs. \(^{12,14}\), we consider a dilute \(^{87}\)Rb atomic gas. The peak of Rabi frequency takes the form of \(\Omega_0 = 2.1\sqrt{\rho/\rho_0}\) MHz, where \(\rho_0\) is the peak atomic density. We consider high atomic density with \(\rho = \rho_0 = 4.3 \times 10^{14}\) cm\(^{-3}\), so that \(\Omega_0 = 2.1\) MHz and the pulse duration \(\tau = 5 \times 10^3/\Omega_0 \approx 2.4\) ms. The decay rate of the excited molecules is chosen as \(\gamma_b = 74\) MHz. The s-wave scattering strengths for \(^{87}\)Rb atoms are taken as \(^{12}\): \(U_{aa} = 4.96 \times 10^{-17}\) MHz cm\(^3\), \(U_{ag} = -6.44 \times 10^{-17}\) MHz cm\(^3\) and \(U_{gg} = 2.48 \times 10^{-17}\) MHz cm\(^3\). Considering the density \(\rho = 4.3 \times 10^{14}\) cm\(^{-3}\), we obtain \(\Lambda_{aa} = 21.328\) kHz = 1.02 \times 10^{-2}\(\Omega_0\), \(\Lambda_{ag} = -27.692\) kHz = 1.32 \times 10^{-2}\(\Omega_0\), and \(\Lambda_{gg} = 10.664\) kHz = 0.51 \times 10^{-2}\(\Omega_0\), respectively.

In Fig. 3, we calculate time evolution of the three-component populations for a fixed free-bound detuning \(\Delta_1 = -1.4\gamma_b\), where a blue-detuned laser is adopted to avoid dynamical unstable state. Mean-field collision terms result in dynamical instability of the CPT state for positive \(\Delta_1\), which in turn leads to lower conversion efficiency \(^{15}\). As shown by the inset of Fig. 3, with time evolution \(\delta\) decreases slightly according to the CPT condition, Eq. \((5)\). In practice, such laser frequency modulation can be realized by setting \(\Delta_2 = -1.4951\gamma_b\) initially, and then increasing it adiabatically to \(-1.3554\gamma_b\). Numerical results of \(|a|^2\) (\(a = a, b, g\)) are shown by solid curves, which are consistent with the CPT solutions \(|a|^{(0)}|^2\) and \(|g|^{(0)}|^2\). We find that the final population of the stable molecules \(|g|^{(\infty)}|^2\) = 0.46, which means that 92% atoms in level |a⟩ are converted into the stable molecules.

We investigate further the relation between the delay time \(T\) and \(|g|^{(\infty)}|^2\) with the free-bound detuning \(\Delta_1\), where we fix \(t_2 = 2.5\tau\) and increase \(T\), so that \(t_1 = 3.0\tau, 3.77\tau,\) and \(4.5\tau\). As shown by the solid line of Fig. 4, the optimal conversion occurs at \(t_1 = 3.77\tau\). The molecular population \(|g|^{(\infty)}|^2\) (also \(\eta\)) varies slowly with \(\Delta_1\) in the blue-detuned region (negative \(\Delta_1\)), while it decreases quickly to zero for positive \(\Delta_1\). This asymmetric variation is aroused from dynamical instability of the CPT in the red-detuned region \(^{15}\). For the case of \(t_1 = 3\tau\) (dashed line), however, \(|g|^{(\infty)}|^2\) begins to decrease in the blue-detuned region since for certain parameters the instability can also occur as shown by inset of Fig. 2. It should be mentioned that in previous work \(^{15}\) the highest conversion occurs at the exact resonance (\(\Delta_1 = 0\)). If the free-bound transition is realized by using Feshbach resonance, strong atomic losses may take place near the resonant point. In our scheme, however, the highest conversion occurs at \(\Delta_1 = -1.4\gamma_b\), far-off resonance to the Feshbach transition. Therefore, condensate atomic losses in state |a⟩ can be ignored safely.

In the above theoretical treatments, we have neglected the decay of stable molecules, \(\gamma_b\). In recent experiment \(^{19}\), however, it was found that \(\gamma_b\) is relatively large and depends on the intensity of the first laser, which may lead to very low conversion rate. There are several reasons that lead to dramatic losses of the ground-state molecules. Firstly, the counter-rotating couplings of the two Raman lasers give contribution to incoherent loss of the stable molecules. Secondly, the coupling of other vibrational molecular levels near level |b⟩ break the ideal three-level system, and provide addition loss channels. Finally, the rogue dissociation of the molecule itself also give negative contributions to the atom-molecule conversion. In fact, the obtained molecular fraction \(|g|^{(\infty)}|\sim 10^{-4}|\), i.e., only hundreds of ground-state molecules are prepared from the \(^{87}\)Rb BEC of about \(10^5\) atoms \(^{19}\). Such a low conversion is caused not only from the decay of the stable molecules mentioned above, but also because of an extremely weak free-bound cou-
pling used in the experiment. To overcome the latter problem a large Franck-Condon overlap integral for the free-bound transition is crucially needed. By carefully choosing suitable molecular levels, one can increase the conversion efficiency by several orders of magnitude.

IV. CONCLUSION

In summary, the efficient conversion between atomic BEC’s and the stable molecular BEC’s has been investigated by using the frequency modulation STIRAP scheme. The effects of the mean-field collisions on the CPT condition and dynamical instability are also discussed. We find that dynamical instability of the CPT solution can also occur in the blue detuned region due to negative atom-molecule interactions. A very high conversion efficiency about 92% can be obtained by choosing optimally the free-bound detuning and time delay of the two Raman lasers. We also find that the highest conversion occurs at the far off-detuned free-bound transition which is important for Feshbach resonance version of the STIRAP in the atom-molecule BEC.

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