Creating a Stable Molecular Condensate Using a Generalized Raman Adiabatic Passage Scheme

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We study the Feshbach resonance assisted stimulated adiabatic passage of an effective coupling field for creating stable molecules from an atomic Bose condensate. By exploring the properties of the coherent population trapping state, we show that, contrary to the previous belief, mean-field shifts need not limit the conversion efficiency as long as one chooses an adiabatic passage route that compensates the collision mean-field phase shifts and avoids the dynamical unstable regime.

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Molecules offer a whole new dimension in the study of ultracold atomic physics. In particular, resonant photoassociation or magnetoassociation (Feshbach resonance [1]) of cold atoms to molecules represents a matter-wave analog of second harmonic generation and has become a new paradigm of coupled macroscopic quantum systems. Because of energy conservation, such processes generally produce molecules in a vibrationally and/or electronically excited quasibound level and hence are not energetically stable. This does not seem to be a serious problem in the case of fermionic atoms due to the suppression of molecular decay by Pauli blocking. In fact, several groups have now successfully Bose condensed so-formed molecules [2]. In contrast, although evidence of macroscopic coherence has been observed in several experiments, excited molecules formed by a pair of bosonic atoms have a very short lifetime ($\lesssim$ a few ms) and the atom-molecule conversion efficiency is limited to $\lesssim 10\%$ [3,4]. Hence, it is very important to be able to create deeply bound ground state molecules from atomic Bose condensates with high efficiency, which is the focus of the current work.

First proposed for photoassociating nondegenerate atoms into stable molecules [5], stimulated Raman adiabatic passage (STIRAP) aided by Feshbach resonance is considered a more efficient way of converting atomic condensates into molecular ones [6,7] than the bare STIRAP implementation of photoassociation [5,8]. In this scheme, the free atomic, the quasibound, and the ground molecular states form the three-level Λ system to which STIRAP can apply [9]. The success of STIRAP relies on the existence of the coherent population trapping (CPT) state [10]. In a linear Λ system, the CPT state exists when the two-photon resonance condition is satisfied; hence STIRAP can be straightforwardly implemented by appropriately choosing the laser frequencies. In the case of condensate, collisions between particles give rise to nonlinear mean-field shifts, which dynamically change when population is transferred from the atomic state to the molecular state. This poses a serious problem for STIRAP as collisions shift the system out of the two-photon resonance. For typical experimental parameters, a conversion efficiency of only $\sim 20\%$ is predicted with a somewhat complicated laser sequence containing seven Raman pulses [6]. A possible remedy is to use a low-density atomic condensate where the effect of collisions can be minimized [11]. This, however, creates new problems as stronger coupling fields and a longer time scale are required.

The purpose of our work is to show that, despite the presence of the mean-field collisions, the ''two-photon'' or the CPT condition [see Eq. (4) below] can be *dynamically maintained* and the high atom-molecule conversion rate can be achieved in condensates of typical density. The nonlinear collisions, however, may induce dynamically unstable regimes in the parameter space. It is crucial for the success of STIRAP to avoid these unstable regimes when designing the route of adiabatic passage.

Our model system is schematically sketched in Fig. 1, where the relevant energy levels are denoted by $|a\rangle$ (free atom), $|m\rangle$ (quasibound molecule), and $|g\rangle$ (ground state molecule). Levels $|a\rangle$ and $|m\rangle$ are coupled by a magnetic field through Feshbach resonance with coupling strength α' and detuning ϵ (ϵ is experimentally tunable via external magnetic field), while $|m\rangle$ and $|g\rangle$ are coupled by a laser field with Rabi frequency and detuning Ω and Δ, respectively [12]. Without loss of generality, we take both α' and Ω to be real as their phase factors can be absorbed by a

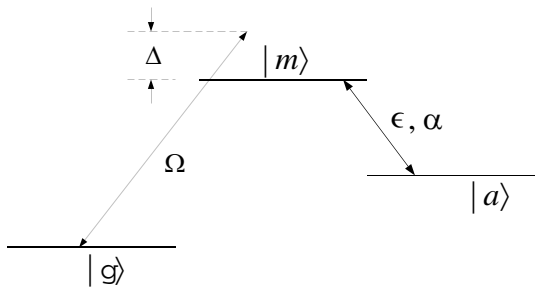

FIG. 1. The energy diagram of three-level atom-molecule system involving free-quasibound-bound transitions.

trivial global gauge transformation of the field operators. In the interaction picture, the Hamiltonian describing the system reads

$$\hat{H} = \hbar \int d\mathbf{r} \Big\{ \frac{1}{2} \sum_{i,j} \lambda'_{ij} \hat{\Psi}_i^\dagger(\mathbf{r}) \hat{\Psi}_j^\dagger(\mathbf{r}) \hat{\Psi}_j(\mathbf{r}) \hat{\Psi}_i(\mathbf{r}) + \epsilon \hat{\Psi}_m^\dagger(\mathbf{r}) \hat{\Psi}_m(\mathbf{r}) + \frac{\alpha'}{2} [\hat{\Psi}_m^\dagger(\mathbf{r}) \hat{\Psi}_a(\mathbf{r}) \hat{\Psi}_a(\mathbf{r}) + \text{H.c.}] + (\Delta + \epsilon) \hat{\Psi}_g^\dagger(\mathbf{r}) \hat{\Psi}_g(\mathbf{r}) - \frac{\Omega}{2} [\hat{\Psi}_m^\dagger(\mathbf{r}) \hat{\Psi}_g(\mathbf{r}) + \text{H.c.}] \Big\},$$

where $\hat{\Psi}_i$ ($i = a$, m, and g) is the annihilation field operator for state $|i\rangle$, the terms proportional to λ'_{ij} represent two-body collisions with $\lambda'_{ii} = 4\pi\hbar a_i/m_i$ and $\lambda'_{ij} = \lambda'_{ji} = 2\pi\hbar a_{ij}/\mu_{ij}$ for $i \neq j$ (a_i and a_{ij} are s-wave scattering lengths, and μ_{ij} is the reduced mass between states i and j) characterizing the intrastate and interstate interaction strengths, respectively. Here we consider a uniform system and hence have dropped the usual kinetic and trapping terms.

From the Hamiltonian we can easily derive the equations of motion of the field operators. We adopt the standard mean-field treatment to replace the field operators $\hat{\Psi}_i/\sqrt{n}$ with c number ψ_i, where n is the density of the total particle number. The mean-field approach ignores high order quantum correlations, but is expected to be valid for systems with a sufficiently large number of particles [13]. The set of the mean-field equations is

$$\frac{d\psi_a}{dt} = -i(\lambda_a|\psi_a|^2 + \lambda_{am}|\psi_m|^2 + \lambda_{ag}|\psi_g|^2)\psi_a - i\alpha\psi_m\psi_a^*, \tag{1a}$$

$$\frac{d\psi_m}{dt} = -\gamma\psi_m - i\epsilon\psi_m - i(\lambda_m|\psi_m|^2 + \lambda_{am}|\psi_a|^2 + \lambda_{mg}|\psi_g|^2)\psi_m - i\frac{\alpha}{2}\psi_a^2 + i\frac{\Omega}{2}\psi_g, \tag{1b}$$

$$\frac{d\psi_g}{dt} = -i(\lambda_g|\psi_g|^2 + \lambda_{ag}|\psi_a|^2 + \lambda_{mg}|\psi_m|^2)\psi_g - i(\Delta + \epsilon)\psi_g + i\frac{\Omega}{2}\psi_m, \tag{1c}$$

where $\lambda_i = \lambda'_{ii}n$, $\lambda_{ij} = \lambda'_{ij}n$, $\alpha = \alpha'\sqrt{n}$ are the renormalized quantities, and the term proportional to γ in Eq. (1b) is introduced phenomenologically to simulate the decay of the quasibound molecules. Implied in Eqs. (1) is the assumption that this decay dominates all the other particle loss mechanisms. Hence, Eqs. (1) hold only in a time scale much shorter than the characteristic times associated with other loss mechanisms, although the time scale itself can be much longer than γ^{-1}.

The technique of STIRAP allows the system to evolve in a coherent superposition of stable particle states ($|a\rangle$ and $|g\rangle$) over time, effectively eliminating the loss of particles through highly unstable states ($|m\rangle$). It is impossible to apply the STIRAP in the usual sense directly to our model because our model contains nonlinear interaction terms. Another difference with the conventional STIRAP is that while the coupling strength of the $|m\rangle \leftrightarrow |g\rangle$ transition can be controlled by the optical pulses, the coupling strength of the $|a\rangle \leftrightarrow |m\rangle$ transition α is fixed not by optical means but by the hyperfine interaction and is hence independent of time [14].

We now proceed to show that despite these important differences, Eqs. (1) support a CPT steady state with $\psi_m = 0$. To this end, let us first neglect the particle loss by taking $\gamma = 0$ and treat all the other parameters as time independent. Then the total particle number is conserved, i.e., $|\psi_a|^2 + 2|\psi_m|^2 + 2|\psi_g|^2 = 1$, so that we can introduce the atomic chemical potential μ through the following steady-state ansatz:

$$\psi_a = |\psi_a|e^{i\theta_a}e^{-i\mu t}, \qquad \psi_{m,g} = |\psi_{m,g}|e^{i\theta_{m,g}}e^{-i2\mu t}. \tag{2}$$

Putting (2) into (1) and taking the time derivative to be zero, one finds that the following CPT solution exists:

$$|\psi_a^0|^2 = \frac{2}{\sqrt{1 + 8(\alpha/\Omega)^2} + 1} = 1 - 2|\psi_g^0|^2, \tag{3a}$$

$$|\psi_m^0|^2 = 0, \tag{3b}$$

with $\theta_g = 2\theta_a$ and the corresponding chemical potential $\mu = \lambda_a|\psi_a^0|^2 + \lambda_{ag}|\psi_g^0|^2$, under the condition

$$\Delta = -\epsilon + (2\lambda_{ag} - \lambda_g)|\psi_g^0|^2 + (2\lambda_a - \lambda_{ag})|\psi_a^0|^2. \tag{4}$$

Equation (4) is the generalized "two-photon" resonance condition that incorporates the nonlinear collisional phase shifts. This CPT solution is thus a generalization of the one found in the collisionless limit [8].

Two crucial conclusions regarding the utility of this CPT state in the adiabatic atom-molecule conversion can be arrived at by observing (3) and (4): (i) Population is concentrated in states $|a\rangle$ and $|g\rangle$ under the respective limit $\alpha/\Omega \to 0$ and $\alpha/\Omega \to \infty$, which facilitates adiabatic coherent population transfer between the atoms and the ground state molecules by tuning the optical Rabi frequency Ω (α, as we mentioned, is regarded as fixed in time). (ii) Since the temporal dependence of Ω uniquely determines the time evolution of the population, we can design *a priori* the temporal evolution of the laser detuning Δ in accordance with $\Omega(t)$ such that the resonance condition (4) is maintained dynamically at any time. (A similar idea can be found in a study of the vortex coupler [15].) In so doing, we eliminate the need to minimize the effect of collisions by reducing the density of the system [11] or by other means [16].

The existence of the CPT state, however, does not guarantee that this state can be followed adiabatically. We have to study the stability properties of the state. This has been neglected in all the previous studies on this problem. For this purpose, we adopt the linear perturbation approach, adding small fluctuations (in both amplitudes and phases) to the steady-state CPT solutions and linear-

izing the equations of motion. The excitation frequencies of the system other than the zero frequency Goldstone mode can be found analytically as

$$\omega = \pm\sqrt{(b \pm \sqrt{b^2 - 4c})/2}, \tag{5}$$

where

$$b = \Omega^2/2 + 2\alpha^2|\psi_a^0|^2 + A^2,$$

$$c = (\Omega^2/4 + \alpha^2|\psi_a^0|^2)^2 + \frac{\alpha^2}{2}|\psi_a^0|^4(\lambda_g - 4\lambda_{ag} + 4\lambda_a)A,$$

with $A = \lambda|\psi_a^0|^2 + \lambda_{ag} - 0.5\lambda_{mg} - \epsilon$ and $\lambda = 2\lambda_a - \lambda_{ag} - \lambda_{am} + 0.5\lambda_{mg}$. When ω becomes complex, the corresponding CPT state is *dynamically unstable.* Hence the unstable regime is given by either $c < 0$ or $c > b^2/4$. The instability here is caused by collisions as it can easily be seen that, in the absence of collisions, ω is always real. The typical results from the stability analysis based on the parameters of our interest are summarized in Fig. 2, where the (Ω, ϵ) space is divided into the stable (white) and the unstable (dark) regions.

In our calculations, we have taken the parameters for ^{23}Na. The s-wave scattering length for the sodium atom is well known and is about $a_a = 3.4$ nm [17], which yields $\lambda'_{aa} = 1.18 \times 10^{-16}\ \mathrm{m^3\,s^{-1}}$. We choose $\alpha' = 4.22 \times 10^{-6}\ \mathrm{m^{3/2}\,s^{-1}}$, corresponding to the atom-molecule coupling strength for the sodium Feshbach resonance at a magnetic field strength of 85.3 mT [18]. We take the condensate density n to be $5 \times 10^{20}\ \mathrm{m^{-3}}$. This gives rise to $\alpha = 9.436 \times 10^4\ \mathrm{s^{-1}}$, and $\lambda_a = 5.9 \times 10^4\ \mathrm{s^{-1}} = 0.625\alpha$. There are so far no good estimates on molecular scattering lengths. So we take the collisional coefficients involving the molecular levels to have the same magnitudes as λ_a. Generally speaking, there are two unstable regions: region I is thin along the ϵ dimension and centers at $\epsilon = \lambda_{ag} - \lambda_{mg}/2$ in the small Ω limit and at $\epsilon = 2\lambda_a - \lambda_{am}$ in the large Ω limit; region II occurs at small Ω and lies either above or below region I in the ϵ direction depending on whether $\lambda_g - 4\lambda_{ag} + 4\lambda_a$ is positive or negative. We show that it is very important to avoid these unstable regions in order to convert atoms into stable molecules.

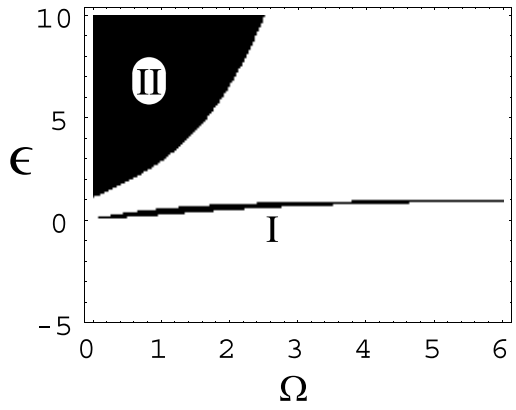

FIG. 2. Stability diagram in (Ω, ϵ) space. Ω and ϵ are in units of α. Other parameters: $\lambda_a = 0.625$, $\lambda_m = \lambda_g = 0.1875$, $\lambda_{am} = \lambda_{ag} = \lambda_{mg} = 0.1875$, all in units of α.

Samples of our results on atom-molecule conversion are displayed in Fig. 3, where we have numerically solved the full set of Eqs. (1) including the loss term with a time-varying Rabi frequency given by

$$\Omega(t) = \frac{\Omega_{\max}}{2}\left[1 - \tanh\left(\frac{t - t_0}{\tau}\right)\right], \tag{6}$$

and Δ changes accordingly as determined by (4). Although in principle the Feshbach detuning ϵ can be varied temporally to optimize the conversion efficiency, we find it not quite necessary. Fixing ϵ at a finite value not only simplifies the experimental procedure, but more importantly it also allows us to avoid strong condensate losses near the exact Feshbach resonance ($\epsilon = 0$) [19]. An experimental implementation of our scheme may thus take the following steps. First, turn on the coupling field and fix its amplitude at $\Omega_{\max}$ while the Feshbach transition is far-off resonance. Next, we bring the Feshbach transition suddenly into near resonance. This is followed by the adiabatic ramp of the amplitude and the frequency of the coupling field accord-

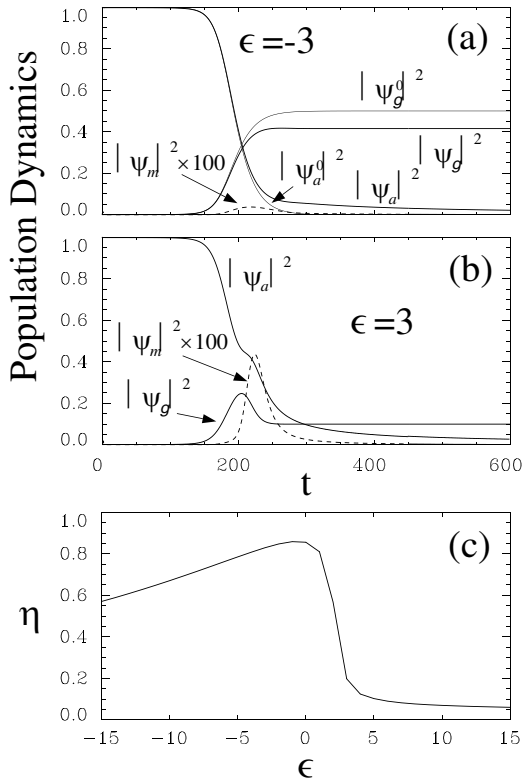

FIG. 3. (a),(b) Population as functions of time for $\epsilon = -3$ and 3, respectively. Also plot in (a) are the CPT solutions of Eq. (5). Here $\gamma = 1$, $\Omega_{\max} = 40$, $\tau = 40$, $t_0 = 120$, and all the other parameters are the same as in Fig. 2. Time is in units of $1/\alpha$, all other quantities are in units of α. (c) Conversion efficiency $\eta = 2|\psi_g(t = \infty)|^2$ as a function of ϵ.

ing to Eqs. (6) and (4), respectively. This implementation represents a counterintuitive sequence as the optical field coupling the initially empty state $|g\rangle$ and $|m\rangle$ is turned on before the interaction on the transition involving the initially populated state $|a\rangle$.

Figure 3(a) shows the time evolution of the population $|\psi_{a,g,m}(t)|^2$. Here ϵ is chosen such that the system remains in the stable regime (see Fig. 2). Also plotted in the figure is the analytical CPT solutions of Eqs. (3). As can be seen, the exact population dynamics follows closely the prediction of the CPT solutions. The small discrepancies at later times can be attributed to the fact that the system cannot maintain adiabaticity completely [20]. The final population in the stable molecular state is $|\psi_g(\infty)|^2 = 0.415$, corresponding to a conversion efficiency of $\eta = 2|\psi_g(\infty)|^2 = 83\%$. The conversion efficiency can be further improved if the optical field is ramped down more slowly. In contrast, in the dynamics depicted in Fig. 3(b), initially the populations follow the CPT solutions, but significant deviation starts to occur at about $t = 200/\alpha$ when the system enters the unstable regime. The final conversion efficiency is less than 20%, which cannot be improved by using a more slowly varying optical field as one does in the stable regime. Figure 3(c) summarizes the conversion efficiencies for various ϵ. The marked asymmetry between positive and negative ϵ is a dramatic manifestation of the dynamical instability.

In conclusion, we have presented here an efficient method to convert atomic condensate into stable molecules using a single optical pulse without the need to sweep through the Feshbach resonance. Our method generalizes the conventional STIRAP scheme in that the laser is frequency chirped to compensate for the mean-field shift arising from the particle collisions, so that, at least in principle, the two-photon resonance is maintained at all times. We also point out that the presence of particle collisions also gives rise to potential dynamically unstable regimes in parameter space. A high conversion efficiency can be reached only if one designs the STIRAP route such that these unstable regimes are avoided.

Note that we do not expect the presence of the trap to change our results significantly as long as the trapping frequencies are made sufficiently small. An estimate in the Thomas-Fermi limit and for a spherical trap of frequency $\omega_0 = 2\pi \times 41$ Hz indicates that more than 90% of a total of 5×10^6 sodium atoms will occupy the central part of the trap where the variation of the mean-field shift due to density inhomogeneity is less than $\alpha/10$. A numerical calculation based on the same parameters as in Fig. 3 but with the two-photon resonance condition violated by an amount of $\alpha/10$ indicates that a conversion efficiency as high as 76% is still attainable.

Finally, we mention that, although we specifically studied a Feshbach resonance situation, due to the formal resemblance of the governing equations, our method can be straightforwardly applied to a photoassociation situation. In fact, we solved Eqs. (3) of Ref. [11] with our laser frequency modulation scheme and obtained a final stable molecular population of 0.42 at density $n = 4.3 \times 10^{14}$ cm^{-3} using the parameters for their Fig. 2(c). Compared to 0.08 obtained in Ref. [11], the advantage of our method is quite obvious.

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