CHIN.PHYS.LETT. Vol. 25, No. 3 (2008) 847

Generation of Coherent Trimers from Bose-Condensed Atoms via a Generalized Stimulated Raman Adiabatic Passage *

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We propose to use a generalized technique of stimulated Raman adiabatic passage to create an atom–molecule dark state that permits the enhanced coherent creation of triatomic molecules in a repulsive atomic Bose–Einstein condensate. As an interesting comparison, the similar cases of creating heteronuclear (bosonic or fermionic) trimers are also briefly discussed.

PACS: 03. 75. Pp, 42. 50. −p, 03. 70. +k

Recently there has been much interest in creating an ultracold molecular condensate (MBEC) via a magnetic Feshbach resonance (FR)[1,2] or an optical photo-association (PA) technique[3] within an atomic Bose–Einstein condensate (BEC). As an atom-optics analogue of the simplest nonlinear quantum optical system (second-harmonic generation),[4] the quantum properties of the hybrid BEC-MBEC have been extensively studied. For example, Herbig *et al.*[5] created a pure molecular quantum gas spatially separated from its atomic partner by adding a magnetic levitation field to an ordinary Feshbach resonance technique within a caesium BEC. Winkler *et al.* [6] experimentally studied the coherent two-colour PA process, a process first dubbed by Heinzen *et al.* as 'superchemistry' or Bose-enhanced free-quasi-bound–bound transition.[7] Very recently, by combining these magneto-optic techniques, Ling *et al.*[8] proposed a generalized atom–dimer dark state method to achieve an efficient and stable conversion of atoms to diatomic molecules in a trapped BEC by minimizing the impact of negative factors like the mean-field shift and the vibrational relaxations (also see Ref. [9]).

So far, the focus of superchemistry has been largely on the atom–dimer coupling process ($a+a \to d$),[10−12] but it certainly includes more complex three-body coupling, i.e., $a + a + a \to g$ or $a + a + b \to g$,[13] where a (b) and d (g) denote atom a (b) and dimer (trimer), respectively. In fact, some remarkable advances have been witnessed in the assembly of ultracold objects of increasing complexity, including the evidence of Efimov triatomic resonances[14,15] or tetramer molecules Cs_4.[16] Thereby, the assembly and quantum control of complex molecules is of particular interest as it may lead to intriguing developments such as the realization of a new type of molecular matter-wave amplifier,[17] the study of the quantum phases of an ultracold trimer or tetramer gas in an optical lattice,[18] as well as the coherent control of, e.g., the trimer-state-intermediated displacement reaction.[19]

The first direct observation of Efimov-type triatomic resonance was achieved by Kraemer *et al.* in 2006 at Innsbruck.[14] In that landmark experiment, the three-body bound-state was observed both for large negative two-body scattering lengths and for small positive values of collisions. There is no dimer state for the former case, but for the latter the trimer state can decay into deeply bound dimer state plus a free atom and hence the signature of trimer formation was observed to be much weaker than the former case.[14] The lifetime of this type of trimer was studied by Nielsen *et al.* in detail.[20]

In this Letter, it is attempting to demonstrate theoretically that a generalized atom–molecule dark-state technique can in principle be used to realize the coherent atom-trimer conversion. The basic idea is to first create highly excited dimers via a standard FR, and then to couple them to a bound trimer via PA. Since a coherent atom–molecule dark state is exploited to prevent the dimer population from becoming significant throughout the conversion process, the formation of trimers is thus expected to be enhanced via this technique. Such a scheme has previously been proposed for the creation of molecular dimers, and has been theoretically demonstrated to be stable for a broad range of conditions. While the proposed scheme is experimentally challenging, recent progress in the manipulation of dimer–atom or even dimer–dimer resonances [16,20−22] indicates that it might become realizable in the near future. It is also noted that this atom–trimer conversion process is quite different from a FR-induced dimer–trimer mixture in a resonant condensate.[23]

Now we consider the coherent creation of homonuclear trimer A_3 in a repulsive atomic BEC, which in practice can be realized through, e.g., an FR-assisted coherent PA method.[8,10−12,24] The direct three-atom coupling is safely ignored by starting from a repulsive atomic BEC below three-body interference minima near zero scattering length.[14] The occupation of intermediate dimers can be minimized by creating an atom–molecule dark state that permits the direct association of atoms into trimers without creating a substantial dimer population in the process. In fact,

*Supported by the Talented-Youth Found of Henan Province and the National Natural Science Foundation of China.
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this atom–molecule dark-state idea also can be used to create, e.g., the molecular tetramer Cs_4[16] or even more complex molecules.

Our model system consists of a Bose condensate of atoms coupled to molecular dimers via an FR, these dimers being in turn photoassociated to the atoms to form bound trimers. Denoting the strength of the atom–dimer coupling with detuning δ by λ', the Rabi frequency of the photoassociation laser by Ω' and its detuning by Δ, the dynamics of the system is described at the simplest level by the model Hamiltonian

$$\begin{aligned}\widehat{\mathcal{H}} = &-\hbar\int dr\Big\{\sum_{i,j}\chi_{ij}\hat{\psi}_i^\dagger(r)\hat{\psi}_j^\dagger(r)\hat{\psi}_j(r)\hat{\psi}_i(r)\\ &+\delta\hat{\psi}_d^\dagger(r)\hat{\psi}_d(r)+\lambda'\big[\hat{\psi}_d^\dagger(r)\hat{\psi}_a(r)\hat{\psi}_a(r)+\text{H.c.}\big]\\ &+(\Delta+\delta)\hat{\psi}_g^\dagger(r)\hat{\psi}_g(r)\\ &-\Omega'\big[\hat{\psi}_d^\dagger\hat{\psi}_a^\dagger\hat{\psi}_g(r)+\text{H.c.}\big]\Big\},\end{aligned} \tag{1}$$

where the annihilation operators $\hat{\psi}_i(r)$, where the indices $i,j = a,d,g$ stand for atoms, dimers and trimers, satisfy bosonic commutation relations: $[\hat{\psi}_i(r),\hat{\psi}_j^\dagger(r')] = \delta_{ij}(r-r')$, and the collision terms proportional to χ_{ij} describe s-wave collisions between these species. The dimers could be formed via rf association of pairs of atoms close to a Feshbach resonance, or by applying a magnetic pulse across the resonance. The nonlinear coupling between dimers and trimers is typically induced by a narrow-frequency, continuous-wave PA laser, for which the Franck–Condon factor can possibly be calculated by, e.g., the diatomics-in-molecules description of potential energy surfaces[21] or other simulations.[20,25]

We assume in the following that the main features of the dynamics of this system are adequately described by a mean-field analysis, $\hat{\psi}_i \to \sqrt{n}\psi_i$, where n is the initial atomic density. In this limit, the system is described by the equations of motion ($\hbar = 1$)

$$\begin{aligned}\frac{d\psi_a}{dt} &= 2in\sum_j\chi_{aj}|\psi_j|^2\psi_a + 2i\lambda\psi_d\psi_a^* - i\Omega\psi_d^*\psi_g,\\ \frac{d\psi_d}{dt} &= -(\gamma-i\delta)\psi_d + 2in\sum_j\chi_{dj}|\psi_j|^2\psi_d + i\lambda\psi_a^2\\ &\quad - i\Omega\psi_a^*\psi_g,\\ \frac{d\psi_g}{dt} &= 2in\sum_j\chi_{gj}|\psi_j|^2\psi_g + i(\Delta+\delta)\psi_g - i\Omega\psi_d\psi_a,\end{aligned} \tag{2}$$

where $\lambda = \lambda'\sqrt{n}$ (l = 1,2), $\Omega = \Omega'\sqrt{n}$ and the decay rate γ accounts for the loss of untrapped dimers. To reduce these losses we exploit a coherent population trapping (CPT) technique that relies on the existence of an approximate atom–molecule dark state.[26]

For the specific case of trimer formation that we consider here it is straightforward to use the following steady-state ansatz (μ_a is the atomic chemical potential)

$$\begin{aligned}&\psi_a = |\psi_{a,s}|e^{i\theta_a}e^{-i\mu_a t}, \quad \psi_d = |\psi_{d,s}|e^{2i\theta_a}e^{-2i\mu_a t},\\ &\psi_g = |\psi_{g,s}|e^{3i\theta_a}e^{-3i\mu_a t},\end{aligned} \tag{3}$$

to show that under the generalized two-photon resonance condition

$$\begin{aligned}&\Delta = -\delta + (6\chi_{ag}-\chi_{gg})nN_{g,s} + (6\chi_{aa}-2\chi_{ag})nN_{a,s},\\ &\mu_a = 2(\chi_{aa}N_{a,s}+\chi_{ag}N_{g,s}).\end{aligned} \tag{4}$$

Equations (2) admit a steady-state solution with no dimer population,

$$N_{g,s} = \frac{1}{3}(1-N_{a,s}) = \frac{(\lambda/\Omega)^2}{1+3(\lambda/\Omega)^2}, \tag{5}$$

where we have used the condition of conserved particles number: $N_{g,s} + 2N_{d,s} + 3N_{g,s} = 1$. This suggests that approximate CPT dynamics such that the dimer population N_d remains small at all times can be achieved for an appropriate 'counter-intuitive' time dependence of the laser detuning Δ[8,24] (also see Ref. [27]). Additionally, it may be possible to exploit an optimal feedback technique[28] to further stabilize this process.

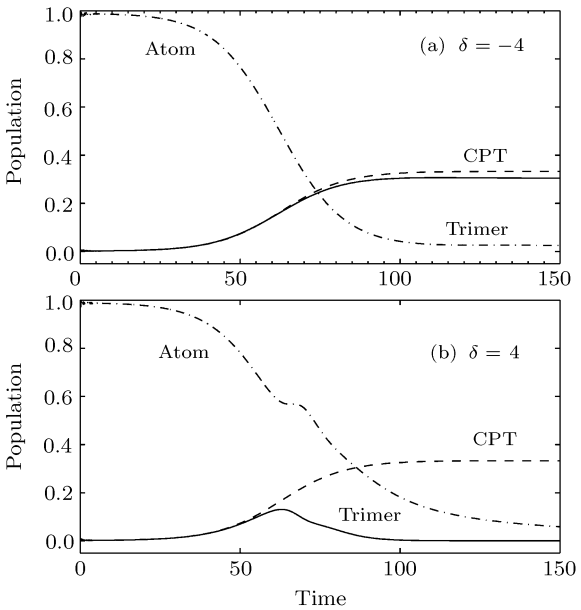


Fig. 1. Time dependence of the normalized atom and homonuclear trimer populations: (a) $\delta = -4$ and (b) $\delta = 4$, for a system initially composed of atoms only, all in units of λ/n, the parameters $\chi_{aa} = 0.5303$ and the other collisions are taken as 0.0938. Time is in units of λ^{-1} and $\gamma = 1$. The dimer population remains essentially zero (not shown). The line labelled by CPT shows the ideal trimer population.

Figure 1 shows the result of a numerical solution of Eq. (2). In this example the various parameters are appropriate for ^{87}Rb, with the s-wave scattering length 5.77 nm, $\lambda = 4.718\times10^4\,\text{s}^{-1}$, and

$$\Omega(t) = \Omega_0\text{sech}(t/\tau), \tag{6}$$

with $\Omega_0/\lambda = 20$, $\lambda\tau = 20$. Very little is known about the scattering lengths of collisions involving molecular trimers, so we have carried out simulations with several sets of plausible parameters, even for other samples of bosonic atoms ^{23}Na (with the s-wave scattering length 3.4 nm or $\chi_{aa} = 0.3125$) or ^{41}K (with $\chi_{aa} = 0.3214$). We found that the stable formation of trimers is always possible for a range of values of the detuning δ.

The curves in Fig. 1(a) give such an example for $\delta = -4$. Their general features resemble those of Fig. 2 in Ref. [26], which corresponds however to the creation of dimers rather than trimers. In particular, we observe a similarly increasing departure of the population transfer from the CPT solution as time evolves. Just as the case for dimer formation,[8] the association of atoms into trimers is characterized by the existence of regions with unstable dynamics. This is for instance the case for $\delta = 4$ as shown in Fig. 1(b).

It is interesting to compare the homonuclear and the heteronuclear cases in creating coherent trimers by starting from the same total number of atoms. In fact, very recently, we have studied the interesting situation of heteronuclear trimer formation A_2B via the intermediate dimers A_2 or AB.[24] For the single-path cases, we obtain the steady-state number of trimers A_2B as

$$N_{g,s} = \frac{1}{3}\Big(\frac{k(\lambda_i/\Omega_i)^2}{1+k(\lambda_i/\Omega_i)^2}\Big), \tag{7}$$

where $i = 1$ and $k = 4$ for the path AA, and $i = 2$ and $k = 1$ for the path AB.[24] Thereby, for a given initial number of atoms we find that

$$N_{g,s}^{\mathrm{AB}} < N_{g,s} < N_{g,s}^{\mathrm{AA}}, \tag{8}$$

where $N_{g,s}$ corresponds to the single-channel situation discussed above. From this result, we know that the more efficient single-path assembly of heteronuclear trimer A_2B is through the intermediate homonuclear dimer A_2 instead of the heteronuclear dimer AB.

Finally we remark that the present method can also be extended to the interesting case of creating the fermionic trimer A_2B by starting from some two-species boson-fermi mixture, such as the bosonic atoms ^{87}Rb (A) and the fermionic atoms ^{40}K (B). In particular, if we consider only the dominated kinetic energy and ignore the s-wave collisions of fermionic particles,[29,30] we can write a single evolution equation for each kind of particles and then find that the steady-state number of fermionic trimers A_2B in the CPT regime remains the same as in the bosonic case. The fact that we found the similar results as in bosonic cases is due to the statistics-independent cooperating many-body effect instead of the bosonic enhancement effect, as firstly revealed in creating fermionic dimers or four-wave mixing.[12,31]

In conclusion, we have shown that a stimulated Raman adiabatic passage (STIRAP) scheme based on FR-assisted photoassociation, which has previously been shown to result in the production of ultracold molecular dimers, can be extended to the generation of molecular trimers. The efficient atom-trimer conversion is realized by minimizing the intermediate dimers via the atom–molecule dark state. Our work suggests that coherent trimer gas holds the promise to become a new playground for the research of coherent matter-wave optics, such as a new type of molecular matter-wave amplifier.[17] Future work will study the quantum statistics of the trimer field,[12] the trimer gas in an optical lattice [17,18] and the possible geometric phases.

This work is related to Ref. [32] which, rather than probing the atom–molecule dark state and the mean-field long-time trimer creation rate, studied the short-time quantum statistics of trimer fields.

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