Coherent Atom-Trimer Conversion in a Repulsive Bose-Einstein Condensate

H. Jing,[1,2] J. Cheng,[3] and P. Meystre[1]

[1]*B2 Institute and Department of Physics, The University of Arizona, Tucson, Arizona 85721, USA*
[2]*Center for Cold Atoms (Wuhan) and Henan Normal College, People's Republic of China*
[3]*School of Physical Science and Technology, South China University of Technology, Guangzhou 510640, People's Republic of China*

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We show that the use of a generalized atom-molecule dark state permits the enhanced coherent creation of triatomic molecules in a repulsive atomic Bose-Einstein condensate, with further enhancement being possible in the case of heteronuclear trimers via the constructive interference between two chemical reaction channels.

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The experimental realization of Bose-Einstein condensates (BECs) in dilute atomic vapors has led to a number of advances with implications well past the confines of traditional atomic, molecular, and optical physics [1]. A development of particular relevance for the present study is the generation and probing of ultracold molecular samples and of molecular condensates from atomic BECs, using magnetic Feshbach resonances (FRs) and photoassociation (PA) [2–5] to control the dynamics of the system. In particular, it has recently been reported experimentally that these techniques can be applied to create both homonuclear [2,5] and heteronuclear [6] ultracold molecules.

The controlled association of ultracold heteronuclear molecules into deeply bound states is a subject of considerable current interest [6,7], in particular, in view of their permanent electric dipole moment that makes them ideal candidates for the study of quantum gases with strong anisotropic interactions [6]. So far, the focus has been largely on the creation of heteronuclear dimers and of their transfer to deeply bound internal states [6], but increasing efforts are now directed toward the assembly of ultracold objects of increasing complexity, including the recent evidence of tetramer molecules Cs_4 [5]. The formation and quantum control of heteronuclear trimers is of particular interest, as it may lead to intriguing developments such as the realization of a triatomic molecular matter-wave amplifier [8], the study of the quantum phases of an ultracold trimer gas in an optical lattice [6,9], and the coherent control of a variety of chemical reactions [10].

The goal of this Letter is to demonstrate theoretically that techniques originally developed for the creation of dimers can, in principle, be extended to the generation of molecular trimers but with an important difference that can be exploited to significant advantage: The creation of heteronuclear trimers can be markedly enhanced by *constructively interfering* the two quantum paths leading to their formation from a two-species atomic BEC [11].

The basic idea of our scheme is to first create highly excited dimers via a standard FR and to then couple them to a bound trimer via PA. The dimer occupation is kept close to zero during the transfer to the trimer state by using a coherent population trapping (CPT) technique. This proposed extension of ''superchemistry'' [3] manipulations from dimers to trimers exploits the existence of three-body bound states in ultracold atomic samples as the scattering length for two-body collisions becomes infinite. As discussed in Ref. [12], this occurs not only for identical particles, a situation first considered by Efimov [13,14], but also for two identical and one different particle [12]. In particular, in the case of two heavy particles and one light particle, these authors found that the heavy-heavy interaction does not matter and that there is an infinite number of bound trimer states as long as the light-heavy scattering length becomes infinite. In the case of the interaction between one heavy and two light particles, by contrast, there is no infinite number of bound states unless both two-body scattering lengths become simultaneously infinite. While the proposed scheme is experimentally challenging, recent progress in the manipulation of dimer-atom resonances [7,15–17] indicates that it might become realizable in the near future. Note also that this process is quite different from a FR-induced dimer-trimer mixture in a resonant condensate [18].

We mentioned that a point of particular interest in the formation of heteronuclear trimers A_2B is the role of quantum interferences between two different reaction paths that involve intermediate dimers A_2 (path AA) and AB (path AB), respectively. To set the stage, we consider first the two different reaction paths independently. Our model system consists of an atomic BEC coupled to molecular dimers via a FR, these dimers being in turn photoassociated to the atoms to form bound trimers. Denoting the strength of the Feshbach coupling with detuning δ by λ_1', the Rabi frequency of the PA laser by Ω_1', and its detuning by Δ, the dynamics of the system is described

at the simplest level by the model Hamiltonian

$$\hat{\mathcal{H}} = -\hbar \int dr \Big\{ \sum_{i,j} \chi_{ij} \hat{\psi}_i^\dagger(r) \hat{\psi}_j^\dagger(r) \hat{\psi}_j(r) \hat{\psi}_i(r) + \delta \hat{\psi}_d^\dagger(r) \hat{\psi}_d(r) + \lambda_1' [\hat{\psi}_d^\dagger(r) \hat{\psi}_a(r) \hat{\psi}_a(r) + \text{H.c.}] + (\Delta + \delta) \hat{\psi}_g^\dagger(r) \hat{\psi}_g(r) - \Omega_1' [\hat{\psi}_g^\dagger(r) \hat{\psi}_d \hat{\psi}_b + \text{H.c.}] \Big\}.$$

Here the annihilation operators $\hat{\psi}_i(r)$, where the indices $i, j = a, b, d, g$ stand for atoms (A and B), dimers, and trimers, satisfy bosonic commutation relations, and the collision terms proportional to χ_{ij} describe s-wave collisions between these species. (Trimer formation via the path AB only can be investigated in a similar fashion.) As discussed by Ospelkaus *et al.* [6], the dimers could, for example, 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 [15] or other simulations [16,19].

We assume in the following that the main features of the dynamics are adequately described by a standard mean-field analysis [4] $\hat{\psi}_i \to \sqrt{n}\psi_i$, where n is the initial atomic density. In this limit, the system is described by the mean-field equations of motion ($\hbar = 1$)

$$\begin{aligned}
\frac{d\psi_a}{dt} &= 2in \sum_j \chi_{aj} |\psi_j|^2 \psi_a + 2i\lambda_1 \psi_d \psi_a^*, \\
\frac{d\psi_b}{dt} &= 2in \sum_j \chi_{bj} |\psi_j|^2 \psi_b - i\Omega_1 \psi_d^* \psi_g, \\
\frac{d\psi_d}{dt} &= -(\gamma - i\delta)\psi_d + 2in \sum_j \chi_{dj} |\psi_j|^2 \psi_d + i\lambda_1 \psi_a^2 - i\Omega_1 \psi_b^* \psi_g, \\
\frac{d\psi_g}{dt} &= 2in \sum_j \chi_{gj} |\psi_j|^2 \psi_g + i(\Delta + \delta)\psi_g - i\Omega_1 \psi_d \psi_b,
\end{aligned} \tag{1}$$

where $\lambda_1 = \lambda_1' \sqrt{n}$, $\Omega_1 = \Omega_1' \sqrt{n}$, and the decay rate γ accounts for the loss of untrapped dimers. To reduce these losses, we exploit a CPT technique that relies on the existence of an approximate atom-molecule dark state. Such techniques are well known in the case of linear systems, where they permit the transfer of population from an initial to a final state via an intermediate state that remains unpopulated at all times. This is the basis for stimulated Raman adiabatic passage (STIRAP), which achieves this goal via a so-called counterintuitive sequence of pulses [2–4].

CPT and STIRAP rely explicitly on the validity of the adiabatic theorem, which applies only to linear systems. Unfortunately, in the situation at hand, two-body collisions render the system nonlinear, and it is not immediately obvious that STIRAP still works in that case. This problem has recently been investigated in Ref. [4], which shows that an approximate adiabatic condition can still be achieved by linearizing the nonlinear system around the intended adiabatic evolution. If the eigenfrequencies of the linearized system are real, the associated "normal modes" do not grow in time, and the system is stable. Hence, a system initially prepared in a CPT state will approximately remain in that state at all times, although the adiabaticity condition may be difficult to fulfill at the later stages of the evolution. In contrast, we show later on that the association efficiency of trimers can be further enhanced by constructively interfering the two paths leading to the final molecule.

For the single-path case of trimer formation, it is easily shown that, under a generalized "two-photon" resonance condition [20], the steady-state solutions with no dimer population *do* exist, i.e.,

$$N_{g,s} \equiv |\psi_{g,s}|^2 = \frac{1}{3} \left(\frac{k(\lambda_i/\Omega_i)^2}{1 + k(\lambda_i/\Omega_i)^2} \right), \tag{2}$$

where $k = 4$ ($i = 1$). Here we have used $|\psi_{d,s}|^2 = 0$ and $|\psi_{a,s}|^2 + |\psi_{b,s}|^2 + 3|\psi_{g,s}|^2 = 1$. The indices of λ and Ω refer to the two different channels. For the path AB, Eq. (2) remains the same, but with $k = 1$ and $i = 2$, so that $N_{g,s}^{AB} < N_{g,s}^{AA}$ for the same external parameters.

This suggests that approximate CPT dynamics such that the dimer population N_d remains small at all times can be achieved for an appropriate "counterintuitive" time dependence of the laser detuning Δ [20] (see also Ref. [21]). Additionally, it may be possible to exploit a feedback technique [22] to further stabilize this process.

Figure 1 shows a numerical simulation of the formation of the heteronuclear trimers A_2B, when considering the single-path (AA) case separately. In this specific example, atom A is ^{41}K and atom B is ^{87}Rb, two atoms for which good scattering parameters are available [11]: $\lambda_l = 4.718 \times 10^4\ \text{s}^{-1}$ ($l = 1, 2$), and $\Omega_l(t) = \Omega_{l,0}\,\text{sech}(t/\tau)$, with $\Omega_{l,0}/\lambda_l = 20$, $\lambda_l \tau = 20$. All in units of λ_l/n, the parameters $\chi_a = 0.3214$, $\chi_b = 0.5303$, and $\chi_{ab} = 0.8731$ and the other collisions parameters are taken as 0.0938 [11]. The scattering lengths of collisions involving molecular trimers, which depend on the details of the interatomic potential, are not yet known, and we actually have carried out simulations with a large number of sets of parameters [23]. We found that the stable formation of trimers is *always* possible by controlling the detuning value δ.

The curves labeled (a) in Fig. 1 give one such example for $\delta = -3$. Their general features resemble those of Fig. 2 in Ref. [4], 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 is the case for dimer formation [3,4], the association of atoms

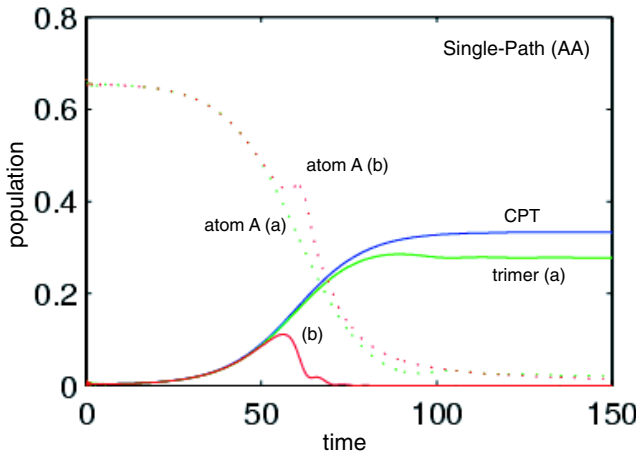

FIG. 1 (color online). Single-path (AA) heteronuclear trimer production for (a) $\delta = -3$ (in green) or (b) $\delta = 3$ (in red) and the corresponding numbers of atom A. Time is in units of λ_l^{-1}, and $\gamma = 1$ (δ and γ are in units of λ_l). The dimer population remains zero at all times and is not shown. The line labeled "CPT" shows the ideal, analytically derived trimer population. The evolution of the atoms B is also not shown.

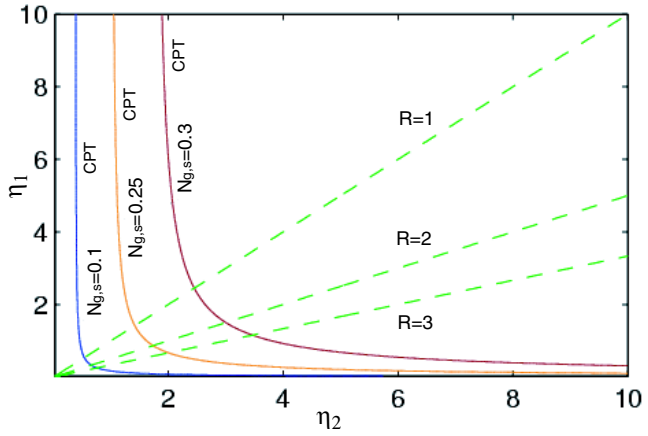

FIG. 2 (color online). Normalized collisionless CPT trimer numbers ($N_{g,s} = 0.1, 0.25, 0.3$) as a function of $\eta_l = \lambda_l/\Omega_l$, $l = 1, 2$. Also shown are three values of $R = \eta_2/\eta_1$. At constant ratios $\Omega_2(t)/\Omega_1(t)$, the counterintuitive evolution of the system is along the lines of constant R starting from the origin.

into trimers is characterized by the existence of regions with unstable dynamics. This is, for instance, so for $\delta = 3$ [curves (b) in Fig. 1]. The dynamics of the AB channel is similar, although it leaves a significantly larger number of atoms A in the sample at the end of the conversion process and, hence, results in a lower yield of trimers.

We have also studied the full quantum dynamics of heteronuclear trimer formation in the short-time limit using a c-number positive-P representation approach [3]. In that limit, as predicted by Hope and Olsen for dimer production [3], the quantum dynamics reproduces the mean-field behavior of trimer creation, and quantum noise-induced trimer damping occurs only near total atom-trimer conversion [3,24].

We now demonstrate that, when acting in concert, the two channels can yield a significantly larger conversion rate and approach the *ideal* CPT yield of $1/3$ (Fig. 2) even in the presence of collisions. Here the coexistence of the two channels provides considerable additional flexibility in approaching the ideal CPT value for trimer formation; see Fig. 3. Note, however, that this approach either requires an accidental coincidence of FRs for the A_2 and AB dimer formation or might be realizable in other cases by applying a magnetic field gradient [25] across the coexisting A and B condensates.

This double-path situation can be described by a straightforward extension of the single-path case. Requiring as before that the number of dimers remains equal to zero, the CPT steady-state number of trimers is then

$$N_{g,s} = \frac{(\lambda_1/\Omega_1)(\lambda_2/\Omega_2)^2}{\lambda_1/\Omega_1 + \lambda_2/\Omega_2 + 3(\lambda_1/\Omega_1)(\lambda_2/\Omega_2)^2}, \qquad (3)$$

where the asymmetry between the two channels results from the fact that the intermediate dimer involves two undistinguishable particles in the first one and two distinguishable particles in the second case.

Figure 2 plots the contour lines of steady-state trimer population $N_{g,s}$ as a function of $\eta_l = \lambda_l/\Omega_l$, $l = 1, 2$, as

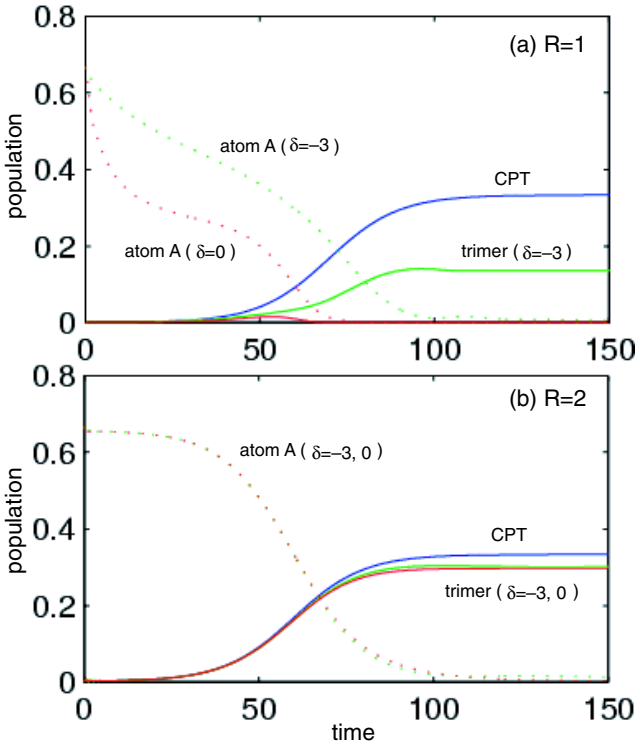

FIG. 3 (color online). (a) Double-path generation of heteronuclear trimers. The time dependence of the populations of the trimers (solid curve) and of atom A (dotted curve) are shown for $\delta = -3$ and $\delta = 0$ and (b) $R = 1$ or (c) 2 . The CPT value of trimers is also plotted. $\chi_{d_1 d_2} = 0$, and $d_{1,2}$ denotes A_2 or AB.

well as the parameter

$$R = \eta_2/\eta_1. \quad (4)$$

(Note that there is no CPT solution for $R < 0$.) As the STIRAP PA pulses $\Omega_1(t)$ and $\Omega_2(t)$ are applied, η_1 and η_2 increase, the system evolving along a line of constant R if the ratio of their amplitudes remains constant.

Figure 2 implies that an efficiency of 0.3 can be reached for any value of R when considering only the coherent coupling of the matter waves. However, it is not obvious that this should still be the case in the presence of collisions. In fact, each of the individual paths leading to the formation of trimers (see Fig. 1 or Ref. [4]) is characterized by an increasing departure of the population transfer from the CPT solution as time evolves. This is where quantum interferences come into play: The freedom of choice of R in Fig. 2, which is a unique feature of the two-channel scheme, provides us with additional flexibility in attempting to approach the ideal CPT value via the interference of the two paths.

To determine whether this is indeed the case, and whether an optimum choice of R permits us to approach a normalized trimer population of $1/3$ under the nonideal STIRAP conditions of our nonlinear system, we have solved numerically the mean-field equations of motion of the system for various values of $R \in [1, 3]$ (see Fig. 3), using the same parameters as in the single-channel case. We found numerically that $R = 2$ leads to a trimer production that most closely approaches the ideal CPT solution and is significantly larger than in the single-channel case of Fig. 1. This is illustrated in Fig. 3(b) for $\delta = 0$ and $\delta = -3$—note the insensitivity of trimer production on the detuning here. For $\delta = -3$, the single-path approach already results in a conversion close to the ideal CPT value; hence, no significant improvement is obtained by using the optimized double-path scheme. This is, however, not generally the case, the situation being drastically different for other detunings. For example, Fig. 3(a), for $R = 1$, shows an almost complete absence of trimer production for $\delta = 0$, and, in that case, optimizing the interference between the two paths improves things dramatically. Similar results hold for the other values of R and collisional parameters [23] that we have considered.

We observe also that in the two-channel case the trimer population can reach a transient value that is larger than its final value. This suggests that maintaining a constant ratio R during the evolution of the system may not be optimal. Future work will use genetic algorithms to determine the optimum time dependence of $R(t)$ for maximum trimer production.

In conclusion, we have shown that a STIRAP scheme based on Feshbach-assisted photoassociation can be applied to creation ultracold molecular trimers and that in the case of heteronuclear molecules the constructive interference between two reaction paths can lead to a significant enhancement of trimer production. We stress that this two-path interference is not dependent on the details of the potential functions of the trimer (which is without a doubt difficult and still remains to be explored in both theoretical simulations [15] and experimental PA). Future work will probe the role of quantum fluctuations in the early stages of trimer production and the quantum statistics of the trimer field. We also plan to study coherent displacement reaction [10,26] and a trimer gas in an optical lattice [8,9].

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