Coherent generation of triatomic molecules from ultracold atoms

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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. The creation of fermionic trimers is also briefly discussed.

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I. INTRODUCTION

The realization of Bose-Einstein condensates (BEC) in ultracold atomic gases has led to a profound revolution in modern physics, from low-temperature physics to atom optics [1]. In particular there has been much recent interest in creating a molecular BEC (MBEC) via a magnetic Feshbach resonance (FR) [2] or optical photoassociation (PA) [3] within an atomic BEC. Herbig *et al.* [4] 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 cesium BEC. Winkler *et al.* [5] experimentally studied the coherent two-color PA process, a process first called "superchemistry" by Heinzen *et al.* [6]. Very recently, by combining these magneto-optic techniques, Ling *et al.* proposed a generalized atom-dimer dark state method to achieve the 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 [7] (see also Ref. [8]). The quantum properties of the hybrid BEC-MBEC have been extensively studied as an atom-optics analog of the simplest nonlinear quantum optical system, second-harmonic generation [9].

So far, the focus of superchemistry has concentrated largely on the atom-dimer coupling process $a+a\rightarrow d$ [10–12], but it certainly includes more complex three-body coupling, i.e., $a+a+a\rightarrow g$ or $a+a+b\rightarrow g$ [13], where a and b denote atoms, d is a dimmer, and g is a trimer. 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] and of tetramer molecules Cs_4 [16]. In view of their novel anisotropic properties [17,18], 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 [19], the study of the quantum states of an ultracold trimer gas in an optical lattice [17,20], as well as the coherent control of the trimer-mediated displacement reaction [21] or the (Bose-enhanced) dissociating paths of some heteronuclear trimers [22].

The goal of this paper is to demonstrate theoretically that the atom-molecule dark state technique can in principle be extended to the generation of molecular trimers. An important result is that the creation of *heteronuclear* trimers can be significantly enhanced by the constructive interference of two quantum channels leading to their formation. 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. A coherent atom-molecule dark state is exploited to prevent the dimer population from becoming significant throughout the conversion process. 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.

This proposed extension of superchemistry 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. [23], this occurs not only for three identical particles [15], but also for two identical and one different particle. While the proposed scheme is experimentally challenging, recent progress in the manipulation of dimer-atom or even dimer-dimer resonances [6,18,24–26] indicates that it might become realizable in the near future. Note also that this atom-trimer conversion process is quite different from a FR-induced dimer-trimer mixture in a resonant condensate [27].

The remainder of this paper is organized as follows: Section II sets the stage for the discussion by introducing a mean-field model of Feshbach resonance assisted photoassociation. We review how stimulated Raman adiabatic passage (STIRAP) techniques can be generalized to nonlinear systems as considered here, and present results for the production of homonuclear trimers. Heteronuclear trimers are discussed in Sec. III, which shows how the existence of two distinct channels for the creation of these molecules offers additional flexibility, and how the quantum interference between these channels can be exploited to reach a level of trimer production close to the ideal, linear STIRAP prediction. Finally, Sec. IV briefly discusses the possible creation of fermionic trimers. Section V is a summary and conclusion.

II. HOMONUCLEAR TRIMERS

As illustrated in Fig. 1, we focus on the simplest three cases of coherent trimer creation in a repulsive atomic BEC, including the homonuclear (I) and heteronuclear cases

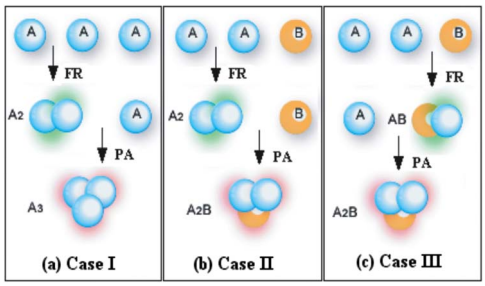

FIG. 1. (Color online) The simplest three cases for coherent atom-trimer conversion in a weakly repulsive atomic BEC, including the homonuclear case (I) [10] and the heteronuclear cases (II–III) where the *same* type of trimers are created through two *different* reaction channels or intermediate dimers.

(II-III). Clearly, the heteronuclear trimer A_2B can be created via two different chemical reaction channels that involve either the intermediate homonuclear dimer A_2 or the heteronuclear dimer AB. In practice, these combination processes can be realized, e.g., through a FR-assisted coherent PA method [7,10–12,28]. Direct three-atom coupling can be safely ignored by starting from a repulsive atomic BEC below the three-body interference minima near zero scattering length [14].

We consider first the creation of homonuclear trimers. The essence of the idea is to minimize the occupation of intermediate dimers 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. We show that this idea, which was previously exploited to create a diatomic molecular condensate [7], can be extended to the more complicated situation at hand.

Our model system consists of a Bose condensate of atoms 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 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

$$\hat{\mathcal{H}}_{\mathrm{I}} = -\hbar \int dr \Big\{ \sum_{i,j} \chi_{ij} \hat{\psi}_i^\dagger(r) \hat{\psi}_j^\dagger(r) \hat{\psi}_j(r) \hat{\psi}_i(r) + \delta \hat{\psi}_d^\dagger(r) \hat{\psi}_d(r) + \lambda' [\hat{\psi}_d^\dagger(r) \hat{\psi}_a(r) \hat{\psi}_a(r) + \mathrm{H.c.}] + (\Delta + \delta) \hat{\psi}_g^\dagger(r) \hat{\psi}_g(r) - \Omega' [\hat{\psi}_d^\dagger \hat{\psi}_a^\dagger \hat{\psi}_g(r) + \mathrm{H.c.}] \Big\}. \quad (1)$$

Here, the annihilation operators $\hat{\psi}_i(r)$, where the indices $i,j=a,d,g$ stand for atoms, dimmers, 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. As discussed by Ospelkaus *et al.* [17], 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 (DIM) description of potential energy surfaces [24] or other simulation methods [25,29].

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 ($\hbar = 1$)

$$\frac{d\psi_a}{dt} = 2in \sum_j \chi_{aj} |\psi_j|^2 \psi_a + 2i\lambda \psi_d \psi_a^* - i\Omega \psi_d^* \psi_g,$$

$$\frac{d\psi_d}{dt} = -(\gamma - i\delta)\psi_d + 2in \sum_j \chi_{dj} |\psi_j|^2 \psi_d + i\lambda \psi_a^2 - i\Omega \psi_a^* \psi_g,$$

$$\frac{d\psi_g}{dt} = 2in \sum_j \chi_{gj} |\psi_j|^2 \psi_g + i(\Delta + \delta)\psi_g - i\Omega \psi_d \psi_a, \quad (2)$$

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. 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.

Recently, Pu *et al.* [30] showed that an approximate adiabatic condition can be achieved for the atom-molecule coupling process by linearizing the nonlinear system around the intended adiabatic evolution. If the eigenfrequencies of the linearized system are real, the associated"normal modes" will not grow in time, and the system is stable. 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 later stages of the evolution. In contrast, if the eigenvalues of the linearized problem are complex, the system is dynamically unstable for some parameter values under which adiabaticity will break down.

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

$$\psi_a = |\psi_{a,s}| e^{i\theta_a} e^{-i\mu_a t},$$

$$\psi_d = |\psi_{d,s}| e^{2i\theta_a} e^{-2i\mu_a t},$$

$$\psi_g = |\psi_{g,s}| e^{3i\theta_a} e^{-3i\mu_a t}, \quad (3)$$

to show that under the generalized two-photon resonance condition

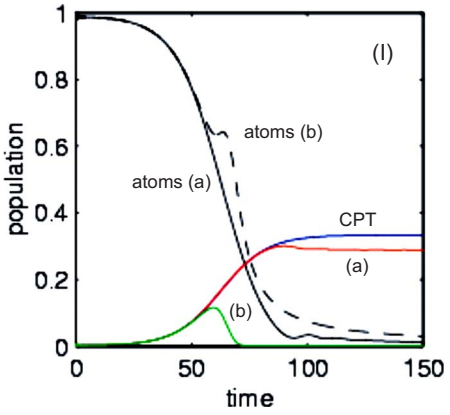

FIG. 2. (Color online) Time dependence of the normalized atom and homonuclear trimer populations. (a) $\delta=3$ (colored red online) and (b) -3 (in green), for a system initially composed of atoms only. Time is in units of λ^{-1} and $\gamma=1$. The dimer population remains essentially zero at all times and is not shown. The line labeled "CPT" shows the ideal, analytically derived trimer population.

$$\Delta_{\mathrm{I}} = -\delta + (6\chi_{ag} - \chi_{gg})nN_{g,s} + (6\chi_{aa} - 2\chi_{ag})nN_{a,s},$$

$$\mu_a^{\mathrm{I}} = 2(\chi_{aa}N_{a,s} + \chi_{ag}N_{g,s}), \tag{4}$$

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

$$N_{g,s} = \frac{1}{3}(1 - N_{a,s}) = \frac{(\lambda_a/\Omega_a)^2}{1 + 3(\lambda_a/\Omega_a)^2},$$

$$N_{d,s} = 0, \tag{5}$$

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 "counterintuitive" time dependence of the laser detuning Δ_I [7,28] (see also Ref. [31]). Additionally, it may be possible to exploit an optimal feedback technique [32] to further stabilize this process.

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

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

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 *many* sets of plausible parameters, such as the example of collisions $\chi_{aa}=0.5303$, and the other collisions are taken as 0.0938 (all in units of λ/n, time is in units of λ^{-1} and $\gamma=1$), for $|\delta|=3$ or $|\delta|=4$. We have also considered the cases of ^{23}Na, with the s-wave scattering length 3.4 nm or $\chi_{aa}=0.3125$, and of ^{41}K (with $\chi_{aa}=0.3214$). We found that the stable formation of trimers was always possible for some range of values of the detuning δ.

The curves labeled (a) in Fig. 2 give one such example for $\delta=3$. Their general features resemble those of Fig. 2 in Ref. [30], 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 [7], the association of atoms into trimers is characterized by the existence of regions with unstable dynamics. This is, for instance, the case for $\delta=-3$ and the parameters of Fig. 2.

We conclude the discussion of the homonuclear case by noting that we have also studied the full quantum dynamics of trimer formation in the short-time limit using a c-number positive-P representation approach [33]. We find that in that limit the quantum dynamics reproduces the CPT dimer production predicted by the mean-field theory, and quantum noise-induced trimer damping occurs only near total atom-trimer conversion [11,34].

III. HETERONUCLEAR TRIMERS

We now turn to the more interesting situation of heteronuclear trimer formation. A point of particular interest is to determine the role of quantum interferences between the two paths that result in the production of the heteronuclear trimer A_2B via the intermediate dimers A_2 and AB. In the following we compare the two single-path cases and then show that the coexistence of these two paths provides considerable additional flexibility in approaching the ideal CPT value for coherent trimer formation.

For the single-path cases II and III (see Fig. 1), the dynamics of the system can be described by the model Hamiltonian

$$\begin{aligned}\hat{\mathcal{H}}_{\mathrm{II,III}} = -\int dr \Big\{ &\sum_{m,n} \chi_{m,n}\hat{\psi}_m^\dagger(r)\hat{\psi}_n^\dagger(r)\hat{\psi}_n(r)\hat{\psi}_m(r) \\ &+ \delta\hat{\psi}_{d_i}^\dagger(r)\hat{\psi}_{d_i}(r) + \lambda_i'[\hat{\psi}_{d_i}^\dagger(r)\hat{\psi}_a(r)\hat{\psi}_{a,b}(r) + \mathrm{H.c.}] \\ &+ (\Delta+\delta)\hat{\psi}_g^\dagger(r)\hat{\psi}_g(r) - \Omega_i'[\hat{\psi}_{d_i}^\dagger\hat{\psi}_{b,a}^\dagger\hat{\psi}_g(r) + \mathrm{H.c.}]\Big\},\end{aligned} \tag{7}$$

where $m,n=a,b,d_i,g$ and $i=1$ or 2 for the case I or II. By using the steady-state ansatz

$$\psi_a = |\psi_{a,s}|e^{i\theta_a}e^{-i\mu_a t},$$

$$\psi_{d_1} = |\psi_{d_1,s}|e^{2i\theta_a}e^{-2i\mu_a t},$$

$$\psi_b = |\psi_{b,s}|e^{i\theta_b}e^{-i\mu_b t},$$

$$\psi_{d_2} = |\psi_{d_2,s}|e^{i(\theta_a+\theta_b)}e^{-i(\mu_a+\mu_b)t},$$

$$\psi_g = |\psi_{g,s}|e^{i(2\theta_a+\theta_b)}e^{-i(2\mu_a+\mu_b)t}, \tag{8}$$

it is easily shown that under the generalized two-photon resonance condition

$$\Delta_{\mathrm{II}} = \Delta_{\mathrm{III}} = -\delta + (4\chi_{ag} + 2\chi_{bg} - 2\chi_g)N_g^0 + (4\chi_a + 4\chi_{ab} - 2\chi_{ag} + \chi_b - \chi_{bg})N_a^0,$$

$$\mu_{a;b}^{\mathrm{II}} = \mu_{a;b}^{\mathrm{III}} = 2(\chi_{aa;bb}N_{a,s;b,s} + \chi_{ab}N_{b,s;a,s} + \chi_{ag;bg}N_{g,s}), \tag{9}$$

the steady-state number of trimers in the CPT regime is given by

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

$$N_{d_i,s} = 0, \tag{10}$$

where $i=1$ and $k=4$ for the path AA, and $i=2$ and $k=1$ for the path AB. In obtaining this result we have used the fact that $2N_{b,s}-N_{a,s}=\varepsilon$, where the c-number ε denotes the initial imbalance between the population of atoms A and twice the population of atoms B. For simplicity we take $\varepsilon=0$ here, but we obtain a similar result for $\varepsilon \neq 0$, with essentially no new physics.

It is interesting to compare the single-path cases (I–III) by starting from the same total number of atoms. For a given initial number of atoms we find that

$$N_{g,s}^{\mathrm{AB}} < N_{g,s} < N_{g,s}^{\mathrm{AA}}, \tag{11}$$

where $N_{g,s}$ corresponds to the single-channel situation of Sec. II. This indicates that the more efficient single-path production of the heteronuclear trimer A_2B is through the intermediate homonuclear dimer A_2 rather than the heteronuclear dimer AB.

One can show both numerically and also via an approximate analytical treatment that the increase in deviation from the optimal CPT values as a function of time is different for the two single-path cases (II–III). To demonstrate this point we numerically simulated the creation of the heteronuclear trimers A_2B, including two-body collisions. Figure 3 shows that results of numerical simulations that consider the channels AA and AB separately. In the specific example of the figure atom A is ^{41}K and atom B is ^{87}Rb, two atoms for which good scattering parameters are available [35], $\lambda_i = 4.718 \times 10^4$ s^{-1} $(i=1,2)$, and

$$\Omega_i(t) = \Omega_{i,0}\,\mathrm{sech}(t/\tau), \tag{12}$$

with $\Omega_{i,0}/\lambda_i = 20$, $\lambda_i\tau = 20$. In units of λ_i/n, the parameters $\chi_a=0.3214$, $\chi_b=0.5303$, $\chi_{ab}=0.8731$, and the other collisions parameters are taken as 0.0938 [35]. The scattering lengths of collisions involving molecular trimers, which depend on the details of the interatomic potential, are not known at this time, leading us to carry out simulations with a large number of sets of parameters. We also considered other atomic species such as ^{23}Na (A) and ^{87}Rb (B), with the s-wave scattering length for Na-Rb collisions taken as half the critical value $\kappa(a_{\mathrm{Na}}a_{\mathrm{Rb}})^{1/2}$ [10], as required for the stable coexistence of Na and Rb condensates in the Thomas-Fermi limit. Here

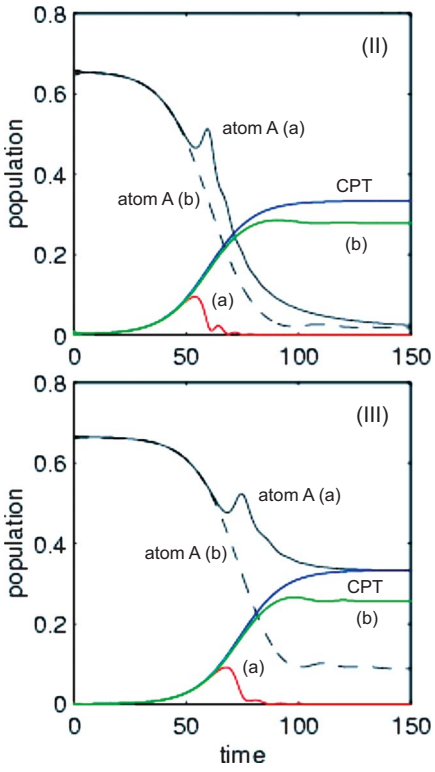

FIG. 3. (Color online) Heteronuclear trimer population for (a) $\delta=3$ (colored red online) or -3 (in green) and their corresponding numbers of atom A. The CPT trimer value is also plotted for the same field parameters as in Fig. 2. The evolution of the population of atoms B and the nearly zero populations of dimer A_2 (II) or AB (III) are not shown.

$$\kappa = (M_{\mathrm{Na}}M_{\mathrm{Rb}})^{1/2}/(M_{\mathrm{Na}} + M_{\mathrm{Rb}}),$$

and $M_{\mathrm{Na,Rb}}$ and $a_{\mathrm{Na,Rb}}$ denote the mass and scattering length of the sodium and rubidium atoms, respectively. This criterion leads to $\chi_a=0.3125$, $\chi_b=0.5303$, and $\chi_{ab}=0.0832$. The other collisions parameters are assumed to be those of atom A, except for $\chi_{kl}=-1.4583$ ($k=a,b$and $l=d,g$). We find that stable trimer creation is always possible for $\delta=-3$ but the system can be unstable in both cases for $\delta=3$. As expected, the two reaction paths do lead to different dynamical behaviors. In particular, the AB channel 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 heteronuclear trimers.

We now proceed to 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, see Fig. 4. Note, however, that this approach either requires an accidental coincidence of Feshbach resonances for the A_2 and AB dimer formation, or might be realizable in other cases by applying a magnetic field gradient [28] across the coexisting A and B condensates.

We describe the two-path situation by the mean-field equations of motion

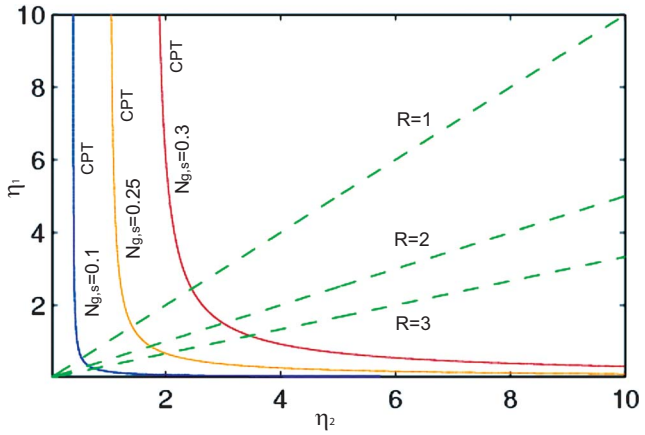

FIG. 4. (Color online) Normalized collisionless CPT trimer numbers ($N_{g,s}$=0.1, 0.25, and 0.3) as a function of $\eta_l=\lambda_l/\Omega_l$, $l=1,2$. Also shown are three values of $R=\eta_2/\eta_1$. At constant ratios $\Omega_2(t)/\Omega_1(t)$ the counterintuitive evolution of the system is along lines of constant R starting from the origin.

$$\frac{d\psi_a}{dt} = 2in\sum_j \chi_{aj}|\psi_j|^2\psi_a + 2i\lambda_1\psi_a^*\psi_{d_1} + i\lambda_2\psi_b^*\psi_{d_2} - i\Omega_2\psi_{d_2}^*\psi_g,$$

$$\frac{d\psi_b}{dt} = 2in\sum_j \chi_{bj}|\psi_j|^2\psi_b + i\lambda_1\psi_a^*\psi_{d_2} - i\Omega_1\psi_{d_1}^*\psi_g,$$

$$\frac{d\psi_{d_1}}{dt} = 2in\sum_j \chi_{d_1j}|\psi_j|^2\psi_{d_1} - (\gamma - i\delta)\psi_{d_1} + i\lambda_1\psi_a^2 - i\Omega_1\psi_b^*\psi_g,$$

$$\frac{d\psi_{d_2}}{dt} = 2in\sum_j \chi_{d_2j}|\psi_j|^2\psi_{d_2} - (\gamma - i\delta)\psi_{d_2} + i\lambda_2\psi_b\psi_a - i\Omega_2\psi_a^*\psi_g,$$

$$\frac{d\psi_t}{dt} = 2in\sum_j \chi_{tj}|\psi_j|^2\psi_t + i(\Delta+\delta)\psi_t - i\Omega_1\psi_{d_1}\psi_b - i\Omega_2\psi_{d_2}\psi_a, \tag{13}$$

where $d_{1,2}$ denote as before the dimers AA and AB, respectively, and the coefficients χ_{ij} account for the two-body collisions between species i and j. Using the steady-state ansatz Eq. (8), we have

$$\lambda_1\psi_{a,s}^2 = \Omega_1\psi_{b,s}\psi_{g,s},$$

$$\lambda_2\psi_{b,s} = \Omega_2\psi_{g,s}. \tag{14}$$

The condition of conserved particles number can now be written as $N_{a,s}+N_{b,s}+2(N_{d_1,s}+N_{d_2,s})+3N_{g,s}=1$. Then it is easy to show that, under the *same* generalized two-photon resonance condition and with the *same* chemical potentials as in the single-path cases [see Eq. (9)], the CPT steady-state solutions is

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

$$N_{d_1,s} = N_{d_2,s} = 0,$$

$$N_{b,s}/N_{a,s} = (\lambda_1\Omega_2)/(\lambda_2\Omega_1), \tag{15}$$

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

Figure 4 plots the steady-state trimer number $N_{t,s}$ as a function of $\eta_i=\lambda_i/\Omega_i$, $i=1,2$, as well as the parameter

$$R = \eta_2/\eta_1. \tag{16}$$

(Note that there is no CPT solution for $R<0$.) As the STIRAP photoassociation pulses $\Omega_1(t)$ and $\Omega_2(t)$ are applied, η_1 and η_2 increase and if the ratio of their amplitudes remains constant the system evolves along a line of constant R. We see 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. 3 or Ref. [30]) 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 an optimum choice of R permits one to approach the ideal trimer population of 1/3 under the nonideal STIRAP conditions of the nonlinear system we have solved numerically the mean-field Eqs. (13) for various values of R between 1 and 3, using the same parameters as in the single-channel case, with $\chi_{d_1d_2}=\chi_{aa}$.

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 situation of Fig. 3. This is illustrated in Fig. 5 for $\delta=0$ and -3. For $\delta=-3$, the trimer fraction also approaches the ideal CPT solution by slightly increasing from ~0.28 to ~0.3. (Note the insensitivity of trimer production to the detuning here.) These results should be contrasted to Fig. 3(a), which shows the trimer number in case $R=1$, again for $\delta=0$ or -3. Here, the trimer production is very significantly reduced and depends strongly on the value of the detuning. Similar results have been obtained for the other values of R and the other sets of collisions parameters 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.

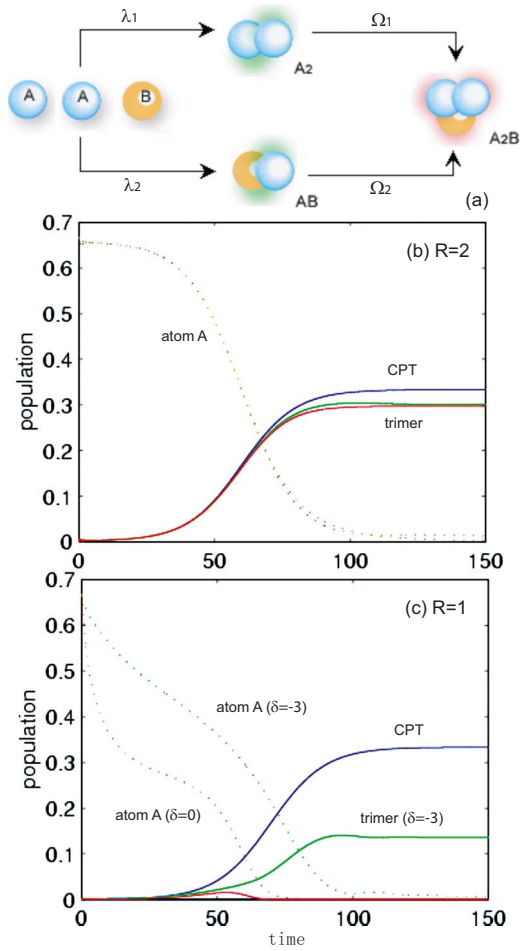

FIG. 5. (Color online) (a) Two-channel generation of heteronuclear trimers. The time dependence of the populations of the trimers and of atom A are shown for (b) $R=2$ and (c) $R=1$ with $\delta=-3$ or $\delta=0$. The CPT value of the trimer is also plotted.

IV. FERMIONIC TRIMERS

Finally we remark that this method can be extended to the creation of the fermionic heteronuclear 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 dominant kinetic energy and ignore the s-wave collisions of fermionic particles [36,37], the energy density corresponding to the two-path Hamiltonian is written in the Hartree approximation as

$$E_{\text{total}} = E_{\text{II}} + E_{\text{III}},$$

$$E_{\text{II,III}} = -\left\{ \sum_{m,n} \chi_{m,n} |\psi_m|^2 |\psi_n|^2 + \delta |\psi_{d_i}|^2 + (\Delta + \delta) |\psi_g|^2 + \sum_f \frac{(6\pi^2)^{2/3}}{10 M_f / 3} |\psi_f|^{10/3} + \lambda_i' [\psi_{d_i}^* \psi_a \psi_{a,b} + \text{H.c.}] - \Omega_i' [\psi_{d_i}^* \psi_{b,a}^* \psi_g + \text{H.c.}] \right\}, \tag{17}$$

where the fermionic components $f=b,d_2,g$ and $\chi_{ff}=0$, the other indexes are the same as in Eq. (7). The method adopted in studying purely bosonic system is not applicable to the present system due to the existence of fermionic particles [36]. We thus study the system in terms of the mean-field Lagrangian density,

$$\mathcal{L} = \frac{i}{2} \sum_j \left(\psi_i^* \frac{\partial \psi_i}{\partial t} - \psi_i \frac{\partial \psi_i^*}{\partial t} \right) - E. \tag{18}$$

Inserting this Lagrangian density into the Euler-Lagrange equation:

$$\frac{\partial \mathcal{L}}{\partial \psi_i^*} - \partial_\nu \left(\frac{\partial \mathcal{L}}{\partial (\partial_\nu \psi_i^*)} \right) = 0, \tag{19}$$

we obtain a set of differential equations for the c-numbers probability amplitudes ψ_a, ψ_b, ψ_{d_1}, ψ_{d_2}, and ψ_g. These single evolution equations for each type of particles turn out to be also in the form of Eqs. (13) but with the following substitution:

$$\chi_{j,j} |\psi_j|^2 \to A_j |\psi_j|^{4/3}, \tag{20}$$

where $A_j=(6\pi^2)^{2/3}/4M_j$ and $j=b,d_2,g$ (M denotes the particle mass, and d_2 is the fermionic dimers AB). The steady-state number of fermionic trimers A_2B in the CPT regime remains the same as in the bosonic cases [see Eqs. (10) and (15)]. The generalized two-photon resonance conditions are also similar to Eqs. (9) [with the above substitution Eq. (20)].

Hence it is straightforward to numerically simulate this process by using a method similar to the purely bosonic case. We find that if one only cares about the particle populations, the results are as in the purely bosonic case [34]. This feature is a manifestation of a statistics-independent cooperative rather than bosonic enhancement, as first discussed in the context of the generation of fermionic dimers and of fermionic four-wave mixing [12,38].

V. CONCLUSION

In conclusion, we have shown that a STIRAP scheme based on Feshbach-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, and that in the case of heteronuclear molecules the interference between two formation channels can lead to a significant enhancement of bosonic or fermionic trimer production.

This two-path constructive interference does not depend on the details of the potential functions of the trimer (which are challenging to determine and still remain to be explored in both theoretical simulations [24] and experimental photoassociation experiments). Future work will study further ways to optimize this interfering-for-the-good scheme and will probe the role of quantum fluctuations in the early stages of trimer production and the quantum statistics of the bosonic or fermionic trimer fields [12,39]. We also plan to study an ultracold trimer gas in an optical lattice [19,20], the

geometric phase in the atom-trimer conversion process, and the trimer-state-intermediated coherent displacement reaction [21,40]. Finally, by starting from a three-species quantum degenerate Fermi-Fermi-Bose or ^{6}Li-^{40}K-^{87}Rb mixture as Taglieber *et al.* created very recently [41], one can even study the creation of heteronuclear trimers *ABC* and their further superchemistry manipulations [22].

While the experimental "bottom-up" realization of atom-trimer conversion promises to be challenging, recent progress in trimer creation [14] and in manipulating dimer-atom resonances [18,24–26] indicates that they may become possible in the not too distant future. This suggests that ultracold triatomic molecular gases hold much promise as a future playground for research in coherent matter-wave optics, such as a new type of molecular matter-wave amplifier [19] or interferometer [42].

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