Density-optimized efficiency for magneto-optical production of a stable molecular Bose–Einstein condensate

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Abstract. Although photoassociation and the Feshbach resonance are feasible means in principle for creating a molecular Bose–Einstein condensate (MBEC) from an already quantum-degenerate gas of atoms, collision-induced mean-field shifts and irreversible decay place practical constraints on the efficient Raman delivery of stable molecules. Focusing on stimulated Raman adiabatic passage, we propose that the efficiency of both mechanisms for producing a stable MBEC can be improved by treating the density of the initial atom condensate as an optimization parameter.

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1. Introduction

Photoassociation occurs when a pair of colliding ultracold atoms absorb a laser photon [1], thereby jumping from the two-atom continuum to a bound molecular state. If the initial atoms are Bose condensed [2], then the subsequent molecules will also form a Bose–Einstein condensate (BEC) [3, 4]. On the other hand, the process known as the Feshbach resonance [5], which we refer to as magnetoassociation, occurs when two ultracold atoms collide in the presence of a magnetic field, whereby the spin of one of the colliding atoms flips, and the pair might then jump from the two-atom continuum to a bound molecular state. As with photoassociation, the so-formed molecules will form a condensate, a molecular Bose–Einstein condensate (MBEC), if the incident atoms are themselves Bose condensed [6, 7]. With the recent observation of atom–molecule coherence [8], it becomes clear that these two mechanisms, or a combination of them, could become the method of choice for creating a stable BEC of molecules, circumventing the need to develop buffer gas cooling [9] and Stark-deceleration [10] techniques.

The catch to photoassociation is that it generally occurs to an excited state, and the subsequent irreversible losses defeat the purpose of molecular coherence. Adding a second laser to drive the molecular population to a stable electronic state allows stimulated Raman adiabatic passage (STIRAP) [11] in the photoassociation [12, 13] of a BEC [15]–[17] to be proposed as a means of avoiding radiative decay. The hallmark of STIRAP is of course the counterintuitive pulse sequence [11], which amounts to adjusting the two lasers so that at the beginning, when almost all of the population is in the initial level (atomic or quasibound BEC), the strongest coupling is the intermediate ↔ final level (excited and stable MBEC), while at the end, when effectively everything is in a stable state, the initial ↔ excited BEC coupling is strongest. Hence, as the population is transferred between the initial and final condensates, the state with the larger population is always weakly coupled to the electronically excited condensate, and the subsequently low (ideally zero) population reduces (eliminates) radiative losses. However, mean-field shifts due to collisions between condensate particles (atom–atom, atom–molecule and molecule–molecule) make it difficult to achieve adiabaticity by moving the system off the required two-photon resonance [16, 17], which can limit the practical efficiency of free–bound–bound STIRAP.

Adiabatic following in magnetoassociation involves only two levels, and occurs because the ground state of the atom–molecule system is all atoms for large positive detunings, yet all molecules for extremely negative detunings. A slow sweep of the magnetic field across the Feshbach resonance thus results in ‘rapid’ adiabatic passage from an atomic to a molecular BEC [18, 19]. The catch now is that the resulting molecular condensate, while translationally and rotationally ultracold, is vibrationally highly excited, and thus susceptible to collision-induced vibrational relaxation. Tacking a pair of lasers onto this scheme allows for what we call Feshbach-stimulated Raman photoproduction of a stable molecular condensate, but the practical efficiency of a concurrent sweep-STIRAP scheme is limited [20], because here adiabaticity is ruined by a time-dependent quasibound molecular energy incurred by the changing magnetic field. Although a static magnetic field cures this deficiency [21], implying that STIRAP will trump radiative losses, an explicit calculation of the effect of decay collisions has not yet, to our knowledge, been performed.

5 Incidentally, STIRAP in the photoassociation of a non-degenerate gas [12, 13] is a matter of ongoing controversy [14].

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Besides radiative and relaxational losses, coupling to the continuum of non-condensate modes occurs because dissociation of a zero-momentum \((k = 0)\) MBEC molecule need not take the atoms back to the \(k = 0\) atomic condensate, but may just as well end up with two atoms with opposite momenta \((\pm k)\), i.e. due to rogue \([13, 22, 23]\) or unwanted \([24, 25]\) dissociation. There are two sides to this story: first, when varying the intensity \(I\) (Feshbach width \(\Delta_B\)), the BEC–MBEC coupling will scale like \(\sqrt{I} (\sqrt{\Delta_B})\), whereas the coupling to rogue modes scales like \(I(\Delta_B)\); second, when changing the density \(\rho\), the BEC–MBEC coupling scales like \(\sqrt{\rho}\), while the rogue coupling is density independent. The standard wisdom is that when the BEC–MBEC coupling (Rabi frequency) exceeds the characteristic value \(\omega_\rho = \hbar \rho^{2/3}/m\), then rogue transitions become not only possible but also favourable over molecular transitions \([13, 22, 23]\), an understanding that led us to deduce \([26]\) rogue dissociation as the mechanism behind the experimentally observed \([8]\) \(^{85}\)Rb BEC–MBEC Ramsey fringes \([27]\)–\([29]\). In the present case, however, rogue dissociation is something to be avoided.

The purpose of this paper is to suggest that the efficiency of STIRAP for a stable molecular BEC can be improved by treating the density of the initial atomic condensate as an optimization parameter. For both free–bound–bound photoassociation and free–bound–bound–bound Feshbach-stimulated photoproduction, we explicitly include the effects of spontaneous decay from the electronically excited molecules and the relevant mean-field shifts, and for the free–bound–bound–bound transitions we also include collision-induced vibrational relaxation. As expected, when the density is lowered from a typical condensate value \((10^{14} \text{ cm}^{-3})\), mean-field and vibrational-relaxation effects play a smaller role, and the efficiency of STIRAP to a stable MBEC can be improved.

Our work is outlined as follows. Section 2 discusses STIRAP in free–bound–bound photoassociation, filling in the gaps in our more abbreviated previous work \([30]\). Section 3 covers the scheme where the atoms first Rabi flop to quasibound molecules under magnetoassociation, which is followed by STIRAP to a stable molecular condensate. We close with the brief discussion of section 4.

### 2. Bose-stimulated Raman adiabatic passage in photoassociation

STIRAP from an atomic to a molecular condensate \([15]\) in the presence of particle–particle interactions has been recently investigated \([16, 17, 30]\). For typical initial BEC densities \((\sim 10^{14} \text{ cm}^{-3})\), it appears that, while atom–molecule STIRAP is feasible in principle, the conversion efficiency of STIRAP can be limited in practice to less than 50%, a limit imposed by two-photon dephasing caused by particle–particle interactions. The purpose of this section is to illustrate how the interactions can be downplayed simply by reducing the condensate density to modest values \((\rho \sim 10^{12} \text{ cm}^{-3})\), so that STIRAP from an atomic to a molecular condensate will proceed with a significantly improved efficiency.

Turning to the situation in figure 1, we assume that \(N\) atoms have Bose condensed into the same one-particle state, e.g. a plane wave with wavevector \(k = 0\). Photoassociation then removes two atoms from this state \(|1\rangle\), creating a molecule in the excited state \(|2\rangle\). By including a second laser, bound–bound transitions remove excited molecules from state \(|2\rangle\) and create stable molecules in state \(|3\rangle\). In second quantized notation, boson annihilation operators for atoms, primarily photoassociated molecules, and stable molecules are denoted, respectively, by \(a\), \(b\) and \(g\). The laser–matter interactions that drive the atom–molecule and molecule–molecule transitions are characterized by their respective Rabi frequencies \(\kappa\) and \(\Omega\). The two-photon
and intermediate detunings are denoted by $\Delta$ and $\delta$. We also introduce atom–atom, molecule–molecule and atom–molecule contact interactions with respective coupling strengths:

$$\lambda_1 = \frac{2\pi a_{11}}{m V_{11}}, \quad \lambda_3 = \frac{2\pi a_{33}}{MV_{33}}, \quad \lambda_{13} = \lambda_{31} = \frac{2\pi a_{31}}{\mu V_{31}},$$

(1)

where $a_{ij}$ is the particle–particle scattering length, $V_{ij}^{-1} = \int d^3r |\psi_i(r)|^2 |\psi_j(r)|^2$ is the inverse effective volume of the single particle wavefunctions ($i, j = 1, 3$), $m (M = 2m)$ is the mass of the atom (molecule), and $\mu = 2m/3$ is the reduced mass of the atom–molecule pair. Anticipating the generally large value for the spontaneous decay rate of the excited molecular condensate (appearing momentarily), the mean-field shifts for the electronically excited molecular state $|2\rangle$ are therefore omitted.

The three-mode Hamiltonian for the above system is

$$\hbar^{-1} H = -\Delta g^\dagger g + (\delta - \Delta) b^\dagger b - \frac{1}{2}(\kappa b^\dagger a a + \kappa^* a^\dagger a^\dagger b) - (\Omega g^\dagger b + \Omega^* b^\dagger g) + \lambda_1 a^\dagger a^\dagger a a + \lambda_3 g^\dagger g^\dagger gg + 2\lambda_{13} a^\dagger g^\dagger ag.$$  

(2)

Before proceeding, we do a bit of rearranging to reduce the number of parameters, and to be consistent with our previous work [15, 30]. Since $N = a^\dagger a + 2(b^\dagger b + g^\dagger g)$ is a conserved quantity, we may add the term $\frac{1}{2}\kappa N^2$ to the Hamiltonian (2) without altering the physics; hence, the time evolution of the system is given also by

$$\hbar^{-1} H = \frac{1}{2}\Delta a^\dagger a + \delta b^\dagger b - \frac{1}{2}(\kappa b^\dagger a a + \kappa^* a^\dagger a^\dagger b) - (\Omega g^\dagger b + \Omega^* b^\dagger g) + \lambda' g^\dagger g^\dagger gg + \lambda a^\dagger g^\dagger ag,$$  

(3)

where the parameters $\lambda' = \lambda_3 - 4\lambda_1$ and $\lambda = \lambda_{13} - 4\lambda_1$ now characterize the mean-field shifts due to collisional interactions. Using a $c$-number approximation to the Heisenberg equations of motion, the Hamiltonian (3) gives the mean-field equations for collective two-colour photoassociation of a quantum degenerate gas:

$$i\dot{a} = (\frac{1}{2}\Delta + \Lambda |g|^2)a - \chi a^* b,$$

(4)

$$i\dot{b} = (\delta - \frac{1}{2}i\chi) b - \frac{1}{2}(\chi a a + \Omega g),$$

(5)

$$i\dot{g} = (\Lambda |a|^2 + \Lambda' |g|^2)g - \frac{1}{2}\Omega b.$$  

(6)
As usual, the semiclassical amplitudes $a$, $b$ and $g$ have been scaled by a factor of $\sqrt{N}$, which makes them of order unity and yields the Bose-enhanced coupling strengths $\chi = \sqrt{N} \kappa$, $\Lambda = N \lambda$ and $\Lambda' = N \lambda'$. Additionally, we have included irreversible spontaneous decay from the electronically excited molecular condensate at the rate $\gamma_s$.

The laser–matter interactions that drive STIRAP from the initial atoms to stable molecules are taken as having the time-dependent form $\chi(t) = \chi_0 \exp[-(t - D_1)^2/T^2]$ and $\Omega(t) = \Omega_0 \exp[-(t - D_2)^2/T^2]$. We focus on explicit numbers for $^{87}$Rb [17]: $\gamma_s = 7 \times 10^7$ s$^{-1}$, $\chi_0 = 2 \times 10^6 \sqrt{\rho/\rho_0}$ s$^{-1}$, $\rho_0 = 5 \times 10^{14}$ cm$^{-3}$, $\lambda_{13} = -5 \times 10^{-11}$ cm$^3$ s$^{-1}$. Since the molecule–molecule scattering length is unknown, we impose $\lambda_3 = 4\lambda_1$, which conveniently gives $\Lambda' = 0$. Anticipating the minimized role of mean-field shifts that occurs for a modest density, we use $\rho = 5 \times 10^{12}$ cm$^{-1}$ as an example, so that $\chi_0 = 2 \times 10^5$ s$^{-1}$ and $\Lambda = -375$ s$^{-1}$.

Offhand, we see that the mean-field shift is roughly three orders of magnitude lower than the peak Bose-enhanced free–bound coupling $\chi_0$, and since it is $\chi_0$ that sets the timescale for atom–molecule STIRAP [15], we expect particle interactions to have a diminished role. Similarly, because STIRAP can generally give an excited condensate fraction of order $10^{-7}$, spontaneous decay should be roughly negligible on a timescale $\tau_s = (10^{-7} \gamma_s)^{-1} \approx 0.15$ s. This intuition is confirmed by the results shown in figure 2. However, in order to achieve efficient conversion for the shortest possible pulse width, it turns out that asymmetric pulses [16, 17] are indeed an advantage. Efficient conversion to a stable $^{87}$Rb$_2$ BEC should be achievable on a timescale $T = 5 \times 10^3/\chi_0 \sim 10$ ms, where $\rho = 10^{13}$ cm$^{-3}$ from figure 2(b).
It is important to note that the above results are actually somewhat sensitive to the assumed collisional coupling $\lambda_3$, since the typically small Rabi coupling means that collisions readily occur on a faster timescale than STIRAP, limiting the final efficiency. However, $\lambda_3 = 4\lambda_1$ could be achieved in practice by tuning the atom–atom scattering length with a Feshbach resonance. Short of an unfortunate accident in the size of the molecule–molecule scattering length, the required change in scattering length ought to be of an order of magnitude or less, so that the required magnetic field will be sufficiently off-resonant (see section 3), and there is no need explicitly to include the Feshbach resonance in the present formalism.

We have neglected the presence of any additional molecular levels, as well as bound–bound transitions induced by the photoassociation laser [17]. The intermediate detuning consider here ($\delta = \chi_0 = 2 \times 10^5$ s$^{-1}$) is trivial compared to the molecular level spacing ($\sim 10^{10}$ s$^{-1}$), and the lower density allows for exact two-photon resonance. Moreover, there is the fact that the so-formed ‘stable’ molecules are still highly excited vibrationally [17], meaning that molecule–molecule and atom–molecule collisions may foster relaxation to lower-lying vibrational levels. There is no known handle on this quenching rate. If, by unfortunate coincidence, a lower density is insufficient on this account (like the mean-fields hifts, the quenching rate is proportional to $\rho$), one might consider targeting lower-lying vibrational levels. Omitting these effects should not be unreasonable.

3. Feshbach-stimulated photoproduction of stable MBEC

We now consider Feshbach-stimulated photoproduction, in which the quasibound state formed in the presence of a static magnetic field is coupled via a Raman laser configuration to a stable molecular state. Again, a version of this scheme, in which STIRAP-inducing lasers are run concurrently with a sweep of the magnetic field across the Feshbach resonance, has been previously proposed [20]; however, the efficiency of STIRAP is limited to a few per cent by the time dependence of the quasibound energy arising from the changing magnetic field$^6$, and large system densities ($\sim 10^{14}$ cm$^{-3}$) allow collision-induced mean-field shifts and vibrational relaxation to dominate atom–molecule conversion. We thus considered previously a modest-density gas of atoms interacting with a static magnetic field, which seemingly enables near-unit efficient ($\sim 95\%$) production of a stable molecular condensate [21]. Here we explicitly include the effects of spontaneous decay, mean-field shifts and vibrational relaxation. As with photoassociation alone, we treat the density of the initial condensate as a variable parameter, and find that the efficiency of converting atoms to stable molecules is improved, this time more dramatically. The source of the drama lies in the strong coupling to the electronically excited state that occurs for the Feshbach-resonance state [20]. This bit of serendipity enables Rabi pulses that are both large and equal, by which STIRAP occurs on a timescale faster than collisions.

Referring to figure 3, we model a gas of $N$ atoms that has Bose condensed into the state $|0\rangle$, and is coupled via a Feshbach resonance to a condensate of quasibound molecules $|1\rangle$. Additionally, one laser drives quasibound $\leftrightarrow$ excited–bound molecular transitions, thus creating a condensate of electronically excited molecules in state $|2\rangle$, and another laser induces excited–bound $\leftrightarrow$ ground transitions to a stable molecular condensate in state $|3\rangle$. The initial atoms are denoted by the unit-order amplitude $a$, the quasibound molecules by $b_1$, the excited molecules by $b_2$ and, finally, the ground state molecules by $g$. Moving right to a semiclassical approximation, the $c$-number equations of motion that govern the evolution of the atom–molecule system are

$^6$ The 20% final efficiency in [20] is obtained by applying a series of seven STIRAP pulses.
Figure 3. Few-level diagram corresponding to Feshbach-stimulated photoproduction of a stable molecular condensate. A gas of atoms has assumedly Bose condensed into state $|0\rangle$. The collective-enhanced Feshbach coupling is $\alpha$, and the corresponding detuning is $\omega_1$. Similarly, the laser 1 (2) coupling is $\Omega_{1(2)}$, and the two-photon (intermediate) detuning is $\delta$. 

\[ i\dot{a} = \tilde{\omega}_0 a + \alpha a^* b_1, \quad (7) \]
\[ i\dot{b}_1 = \tilde{\omega}_1 b_1 + \frac{i}{2} \alpha a a^* + \frac{1}{2} \Omega_{1} b_2, \quad (8) \]
\[ i\dot{b}_2 = \delta b_2 + \frac{1}{2} \Omega_{1} b_1 + \frac{1}{2} \Omega_{2} g, \quad (9) \]
\[ i\dot{g} = \tilde{\delta} g + \frac{i}{2} \Omega_2 b_2, \quad (10) \]

where the mean-field shifts and spontaneous decay are contained in the quantities $\tilde{\omega}_0 = 2(\Lambda_{01}|b_1|^2 + \Lambda_{03}|g|^2)$, $\tilde{\omega}_1 = \omega_1 + \Lambda_{11}|b_1|^2 + 2(\Lambda_{01}|a|^2 + \Lambda_{13}|g|^2)$, $\tilde{\delta} = \delta - \frac{1}{2} i \gamma_5$ and $\tilde{\delta} = \Delta + \Lambda_{13}|g|^2 + 2(\Lambda_{03}|a|^2 + \Lambda_{13}|b_1|^2)$. Collisions with electronically excited molecules are neglected as in section 2.

The Bose-enhanced strength of the Feshbach resonance is given by $\alpha = [2\pi \rho|a|\mu_{ma} \Delta_B/m]^{1/2}$, where $a$ is the off-resonance s-wave atom–atom scattering length, $\mu_{ma}$ is the difference in magnetic moments between the quasibound molecule and free-atom pair, and $\Delta_B$ is the resonance width in magnetic-field units. The molecular energies are $\hbar \omega_i$, with the quasibound energy given specifically in terms of the magnetic field $B$ by $\hbar \omega_1 = \text{sgn}[a] (B - B_0)\mu_{ma}/2$, where $B_0$ locates the resonance position. The Rabi frequency corresponding to laser L1 (L2), which has a frequency $\omega_{L1(2)}$, is denoted by $\Omega_{L1(2)}$; correspondingly, the respective intermediate and two-photon detunings are $\delta = \omega_2 - \omega_{L1}$ and $\Delta = \omega_3 - \omega_{L1} + \omega_{L2}$.

Since the numbers are not well known for atom–molecule and molecule–molecule collisions, we assume that the mean-field shift due to the atom–atom scattering length, $\Lambda = 2\pi \hbar \rho a_{00}/m$, approximates the effect of s-wave particle collisions in general, and write $\Lambda_{ij} \equiv \Lambda (i, j = 0, 1, 3)$. Similarly, collision-induced vibrational relaxation of the quasibound molecular state.
Table 1. Explicit Feshbach-stimulated photoproduction numbers for various systems. Resonance positions $B_0$ are given in G, densities $\rho$ in $10^{14}$ cm$^{-3}$, and collective-enhanced atom–molecule couplings $\alpha$ in ms$^{-1}$. The mean-field shift associated with s-wave collisions is approximated by 

$$\frac{\Lambda_1}{\Lambda_{10}} = \frac{4\pi\hbar}{\rho a_0} \frac{\alpha_{00}}{m}$$

where $a_{00}$ is the atom–atom scattering length. The vibrational relaxation rate $G/\rho = 4 \times 10^{-10}$ s$^{-1}$ is borrowed from sodium [7]. The ideal [21] atom–molecule conversion rate, $R = 0.95 \alpha/3$, is compared to the rogue-dissociation-imposed rate limit for converting atoms to molecules, $R_\rho = \frac{6\hbar}{m} \frac{\rho^{2/3}}{3}$. To estimate $\alpha$, we assume $\mu_{ma} = \mu_B$, where $\mu_B$ is the Bohr magneton.

| Atom  | $(B_0)$ | $\rho$ | $\alpha$ | $\Lambda/\alpha$ | $G/\alpha$ | $R/R_\rho$ |
|-------|---------|--------|----------|------------------|------------|------------|
| $^7$Li [31] | (800) | 1 | 3989 | 0.0008 | 0.0013 | 1.077 |
| $^{23}$Na [32] | (853) | 0.01 | 4.13 | 0.029 | 0.012 | 0.079 |
| $^{85}$Rb [33] | (155) | 0.01 | 142.4 | $-0.0013$ | 0.0004 | 10.1 |
| $^{87}$Rb [34] | (680) | 0.1 | 8 | 0.061 | 0.062 | 0.013 |

adds imaginary parts to the parameters $\Lambda_{1i}$ ($i = 0, 1, 3$), which are assumedly all equal to the value $\text{Im}(\Lambda_{10})$ appropriate for atom–quasibound collisions. In the end, the large coupling between the Feshbach and electronically excited molecular states [20] translates into large Rabi frequencies, which means that collisions, and thus the exact values of the collisional couplings (real or imagined), have little bearing on the results.

Finally, rogue dissociation of the quasibound molecules to non-condensate modes is in fact non-exponential, i.e. the Fermi Golden Rule and simple inclusion with a non-Hermitian term [7, 20] do not strictly apply [23]. Rogue dissociation imposes a rate limit on atom–molecule conversion given roughly by $R_\rho \approx \frac{6\hbar}{m} \frac{\rho^{2/3}}{3}$ [4, 22, 23]. For a ballpark comparison, we estimate from [21] that 95% of the atoms are converted into quasibound molecules in roughly $3/\alpha$ seconds, so that $R = 0.95 \alpha/3$ is to be compared with $R_\rho$.

Explicit numbers for various systems [31]–[34] are listed in table 1, where the densities (resonance widths) are chosen as a balance between being low (high) enough to enable safe neglect of s-wave and vibrational–quenching collisions, and large (small) enough to keep atom–molecule conversion below the rate limit. The table indicates that rogue dissociation is the main obstacle. Overall, the $^{23}$Na (853 G) and $^{87}$Rb (680 G) resonances appear as the strongest candidates. We choose to focus on $^{23}$Na, since this system is well characterized [20]. The remaining unspecified parameter is $\gamma_s = 6 \times 10^7$ s$^{-1}$.

Given that the collective-enhanced free–bound coupling $\alpha$ determines the appropriate frequency scale, $N$ atoms will coherently flop to $N/2$ quasibound molecules on a timescale given by $1/\alpha$, and we need only wait for a sufficient population to build before effecting STIRAP. As in the previous section, our model assumes Gaussian laser pulses of the form $\Omega_i = \Omega_0 \exp[-(t - D_i)^2/T^2]$, where $i = 1, 2$ and the pulse parameters are $\alpha T = 1/2$, $D_2 = 5T$ and $D_1 = 7T$. Adiabaticity is assured by selecting pulse heights such that $\Omega_0 T = 5 \times 10^3$. The atoms are tuned close to the Feshbach resonance with $\omega_1/\alpha = -0.1$, and the molecules onto (off) two-photon (intermediate) resonance with $\Delta = 0$ ($\delta = \Omega_0$). From table 1, $\alpha \sim 4 \times 10^3$ s$^{-1}$ for sodium, so that equal Rabi frequencies $\Omega_0 \sim 40 \times 10^6$ s$^{-1}$ are required, which is not unreasonable [20].
Figure 4. Creation of a stable molecular condensate via Feshbach-STIRAP. Frequency (time) is in units of $\alpha (\alpha^{-1})$. (a) For $\rho = 10^{11}$ cm$^{-3}$, we see that, as the collective conversion of atoms to quasibound molecules nears completion, the lasers transfer the population to the stable molecular condensate with about 85% efficiency. (b) The Gaussian laser pulses have equal widths and heights: $T = 1/2$ and $\Omega_0 = 2 \times 10^3$.

For $\rho = 10^{11}$ cm$^{-3}$, figure 4 shows magnetoassociation collectively converting $\sim 85\%$ of the initial condensate atoms into a quasibound molecular condensate, and the STIRAP pulses subsequently transferring the entire quasibound population to the stable molecular condensate. Figure 5 indicates that the optimum density is indeed $\rho \sim 10^{11}$ cm$^{-3}$. For such densities, 85% conversion occurs in about $3/\alpha \simeq 2$ ms, so that $R/R_{\rho} = (0.85\alpha/3)/R_{\rho} \approx 0.1$ makes rogue dissociation a borderline issue.

4. Discussion

There are, as usual, a few things that remain neglected. Beginning with trap dynamics, we note that, for a spherically symmetric trap in the Thomas–Fermi limit, our decrease in density from $\rho \sim 10^{14}$ to $10^{12}$ cm$^{-3}$ correspondingly reduces the trap frequency from $\omega_r/2\pi \sim 100$ to 1 Hz (presuming a fixed atom number), so that the motion of our atoms will occur on a timescale $\sim 500–1000$ ms. For atom–molecule conversion occurring over a few tens of milliseconds, the trap dynamics should have a negligible effect. To account for the variation of the density across the trap, one could in the leading order of approximation use the average density, which in the Thomas–Fermi limit is two-fifths of the maximum density.

Quantum fluctuations have been neglected, too. So-called beyond-mean-field theory effects can be split into one- and two-body considerations. For the one-body case, we note that the semiclassical theory of rapid adiabatic passage is accurate on the 1% level for as few as $N = 100$ particles [22], and similar results were recently found for photoassociation in general [35]. Neglecting two-body effects amounts to neglecting rogue dissociation. In photoassociation, this is not much of a stretch because photodissociation is generally a good deal slower than spontaneous decay. If STIRAP beats spontaneous decay, then it should easily
Figure 5. Density dependence of the efficiency of collective Feshbach–STIRAP. Similar to photoassociation, the timescale is set by the atom–molecule coupling ($\alpha\sqrt{\rho}$), so that the gain in efficiency that is achieved by marginalizing collisions at low density is undone when the conversion and spontaneous decay occur on comparable timescales.

beat photodissociation. On the contrary, we find that rogue dissociation cannot necessarily be avoided in free–bound–bound–bound transitions. However, even if the Feshbach coupling $\alpha$ is well above the rate limit, rapid adiabatic passage from atoms to molecules can still occur for a sweep directed from above to below threshold [36, 37]. If rogue dissociation is found to be an issue, a ‘sweep–dump’ scheme might therefore be employed, whereby STIRAP-inducing pulses are applied after a properly directed sweep across the Feshbach resonance is satisfactorily completed.

Experiments with photoassociation alone [38] are just on the verge of coherent conversion [22], and culmination requires a difficult-to-impossible balance of low density and high laser intensity simultaneously to minimize mean-field shifts and rogue dissociation. The advantage of the Feshbach-stimulated photoproduction is the improved Frank–Condon overlap with the intermediate molecular level, making it potentially easier to enact coherent conversion. For both of these mechanisms, we have shown here that mean-field shifts can be made reasonably negligible in the utilization of STIRAP, and the same goes for vibrational relaxation in free–bound–bound–bound transitions. This result is based on the observation that—along with the detuning, pulse width and pulse delay [17]—the density of the initial atomic condensate can be treated as an optimization parameter. Efficient production of a BEC of stable molecules via collective atom–molecule conversion could now appear to be a matter of experimental design.

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