

Stimulated Raman molecule production in Bose-Einstein condensates

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A Raman photoassociation process can be used to form translationally cold molecules from a Bose-Einstein condensate. Coupled-channels scattering calculations show that the rate coefficient for coherent production of molecules in specific target states, $>10^{-10}$ cm³/s, can greatly exceed the rate coefficient for heating and loss channels due to excited-state spontaneous decay. This implies the feasibility of rapid coherent conversion of atom pairs in a condensate to molecules on a time scale short compared to the trap oscillation time. [S1050-2947(98)50708-1]

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The successful experimental realization of Bose-Einstein condensates [1–3] makes possible their application as sources of coherent matter waves. A prototype “pulsed atom laser,” a matter analog of a pulsed photon laser, has been demonstrated experimentally with a Na atom condensate [4], using radio-frequency (rf) pulses to switch the atoms trapped in the $F=1, M=-1$ Zeeman sublevel to an untrapped sublevel, causing pulses of coherent matter waves to be ejected from the trap. Here we propose using a Raman photoassociation scheme, as indicated schematically in Fig. 1, to form translationally ultracold molecules from pairs of colliding atoms in a condensate. This offers the prospect of a “molecule laser” or even Bose-condensed molecules. The process could also be used to make molecules (1) in a cold, dense thermal gas that has not yet reached temperatures below the critical temperature for condensation or (2) in an optical lattice if it were possible to put two atoms in a single cell. Photoassociation, the process by which two colliding atoms are excited by light to form a bound molecule, has been suggested as a means of making cold molecules [5–7], and production of cold Cs₂ molecules has now been observed in a magneto-optical trap [8]; a nonoptical method of making cold molecules has also been proposed [9]. Our treatment is specifically developed for making molecules from condensates.

As long as the detuning Δ_A from atomic resonance is large compared to the atomic linewidth γ_A , or collective linewidth in the case of a condensate, the atoms in a cold thermal gas or a condensate scatter light of frequency ω_1 at the rate per atom, $\Gamma_A = \gamma_A(\Omega_A/\Delta_A)^2$, where $\hbar\Omega_A$ is the optical coupling matrix element. A condensate will have a prominent photoassociation spectrum [10], and the rate of light scattering per atom due to binary collisions of pairs of atoms is $\Gamma(p) = K(p)n = \langle \sigma_p v \rangle n$, where n is the atomic density, σ_p the cross section for the collisional process making

product p , v the relative collisional velocity, and the brackets indicate an average over the distribution of v . The cross section for s -wave collisions of identical atoms near $T=0$ is written in terms of the S -matrix element for the collisional process as

$$\sigma_p(k) = (2-x^2) \frac{\pi}{k^2} |\delta_{pg} - S_{pg}(k)|^2, \quad (1)$$

where $k = \mu v/\hbar$ is the relative collisional momentum of the atom pair with reduced mass μ and collision energy $E = \hbar^2 k^2/2\mu$, and $S_{pg}(k)$ is the S -matrix element for undergoing a transition from the colliding ground-state atoms (g) to the product p ; the factor $(2-x^2)$ represents the modification due to the presence of a condensate of fraction x [11]. For the case of one-photon photoassociation through level v_1 in Fig. 1, the products p_1 of the inelastic collision process result in loss of trapped atoms when the excited vibrational level in molecular potential $V_1(R)$ decays by spontaneous emission back to translationally hot ground-state atoms or to bound molecules distributed over a number of molecular states according to the Franck-Condon factors for emission. Reference [12] gives a resonant scattering expression for the probability of such loss: $P(\text{loss}) = |S_{p_1g}|^2$. Reference [10] applied this expression to the case of a condensate to show that the binary pair light scattering rate $\Gamma(p)$ typically exceeds the free atom light scattering rate Γ_A by several orders of magnitude, on the order of $(\Delta E_1/\hbar\gamma_1)$, when the light is tuned to resonance with an upper level v_1 : $E_{v_1} = E + \hbar\omega_1$. Here ΔE_1 is the vibrational spacing of the upper vibrational levels, and γ_1 is the molecular decay rate. When the detuning Δ_A is more than a few γ_A , ΔE_1 is much larger than γ_1 , and the resonant enhancement factor is much larger than unity.

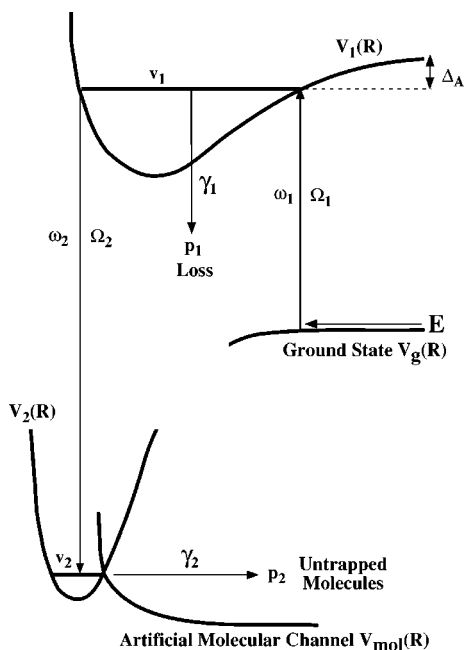


FIG. 1. Schematic molecular potential-energy curves for molecule formation via resonant photoassociative Raman transitions of two colliding ground-state atoms with potential $V_g(R)$ (R is interatomic separation) through the intermediate level v_1 in excited potential $V_1(R)$. The target molecular level v_2 can be any level in the potential $V_2(R)$. Level v_1 decays with rate γ_1 to loss products, and level v_2 decays with rate γ_2 to the artificial molecular channel with potential $V_{mol}(R)$, simulating ejection of the molecules from the optical excitation region where they are made.

Bohn and Julienne [13] generalized the resonant scattering expression of Napolitano *et al.* [12] to two colors, the case when a second frequency ω_2 is added in Fig. 1. The target molecular level v_2 is assumed to decay by some process, with decay rate γ_2 . In contrast to Ref. [13], where the level v_2 was an excited Rydberg state that decayed spontaneously to produce ground-state molecules, the target level v_2 in Fig. 1 represents a nondecaying stable bound-state molecule. Here the decay process represents the removal of the untrapped molecules by their falling out of the trap; thus $\gamma_2 = 1/\tau_{res}$, where τ_{res} is the residence time of the molecules in the region of the focused light beam. Reference [13] suggested that by selecting the laser frequencies so that the two bound states are in exact resonance with the collision energy E , namely $E_{v_1} = E + \hbar\omega_1$ and $E_{v_2} = E + \hbar\omega_1 - \hbar\omega_2$, and by choosing the two laser intensities appropriately, it should be possible to make the probability $P(\text{mol}) = |S_{p_{2g}}(k)|^2$ for formation of the ejected molecule products p_2 approach the unitarity limit, $P(\text{mol}) \rightarrow 1$, in which case $P(\text{loss})$ becomes very small. Thus, it should be possible to form molecules efficiently from a collision of two cold atoms without destructive loss processes being introduced by spontaneous decay of the intermediate state.

We test our proposed molecule production scheme by introducing a numerical model that sets up a coupled-channels scattering calculation for the atoms in the two light fields to represent the five channels in Fig. 1: (1) the colliding ground-state atoms with potential $V_g(R)$, (2) the excited state with potential $V_1(R)$, (3) the target channel with poten-

tial $V_2(R)$, (4) the decay channel for the excited state with decay rate γ_1 , and (5) the artificial molecular channel with potential $V_{mol}(R)$ representing the ejection of molecules from the trap. The excited-state channel is coupled to the ground state by the optical coupling matrix element $\hbar\Omega_1$, and the two bound-state channels are coupled by the optical coupling matrix element $\hbar\Omega_2$. These are proportional to the strengths of the corresponding electronic transition dipole matrix elements and the square root of the corresponding laser intensities I_1 and I_2 . The specific intermediate level $|v_1\rangle$ and final target level $|v_2\rangle$ are selected by tuning the laser frequencies ω_1 and ω_2 . The decay of the excited channel by spontaneous emission with rate γ_1 is included by using a complex potential, as in Refs. [5,12,13]. Thus, the decay channel for v_1 is not explicitly represented, but instead $P(\text{loss})$ is calculated from loss of unitarity of the S matrix. The open decay channel 2 is represented explicitly as in [5,13] by an arbitrary ‘‘artificial channel,’’ whose coupling matrix element $\hbar\Omega_{mol}$ is selected so that the desired γ_2 is given by the golden rule: $\gamma_2 = (2\pi/\hbar)|\langle v_2 | \hbar\Omega_{mol} | E_{mol} \rangle|^2$.

Although the model potentials were taken from the model of Ref. [5], the qualitative features of resonant Raman molecule production should be robust, scaling approximately as the laser intensities times the Franck-Condon factors involved, i.e., $I_1|\langle v_1 | E \rangle|^2$ and $I_2|\langle v_1 | v_2 \rangle|^2$. Therefore, our qualitative conclusions should be applicable to a variety of different molecular schemes with a wide range of Franck-Condon factors. For example, if F_1 and F_2 represent the lower and upper hyperfine spin states of an alkali-metal atom, collisions of doubly spin-polarized F_2 atoms only occur on the $a^3\Sigma_u^+$ potential, implying a gerade intermediate state for V_1 and $a^3\Sigma_u^+$ for V_2 . A distribution of trapped and untrapped Zeeman components of the $a^3\Sigma_u^+$ rotational state is likely, with some control over the distribution through the polarizations of the two Raman lasers. Possible intermediate states are $^3\Sigma_g^+(P_{1/2}+S)$ or the $0_g^-(P_{3/2}+S)$, where (P_j+S) indicates the separated atom limit to which the state correlates. The $0_g^-(P_{3/2}+S)$ intermediate state is known to produce molecules efficiently for Cs_2 photoassociation [8], and Almazor *et al.* [14] calculate that the same should be true for Rb_2 . By contrast, collisions of two F_1 atoms occur on a mixture of $a^3\Sigma_u^+$ and $X^1\Sigma_b^+$ potentials, implying an intermediate V_1 of either gerade or ungerade symmetry, such as $A^1\Sigma_u^+(P_{1/2}+S)$, so that V_2 could be $X^1\Sigma_g^+$ as well as $a^3\Sigma_u^+$.

The model potentials were selected according to the model of Ref. [5]. The specific detunings and optical couplings can be scaled for other schemes according to the energies and Franck-Condon factors for the levels involved; all of the schemes mentioned above have ranges of levels for which the two Franck-Condon factors are suitable. The level v_1 was given a decay rate of $\gamma_1 = 2\pi(10 \text{ MHz})$ and the level v_2 was given a decay rate of $\gamma_2 = 2\pi(0.8 \text{ kHz})$, corresponding to a residence time of $200 \mu\text{s}$, a typical time to leave the condensate due to two recoil units of photon momentum. The v_1 level in Ref. [5] is deeply bound, with Δ_A on the order of $2\pi(2.4 \text{ THz})$, and the target level v_2 is the vibrational ground state of its potential, $V_2(R)$. Schemes with v_1 and v_2 much higher in their respective potentials are more likely to be experimentally realistic.

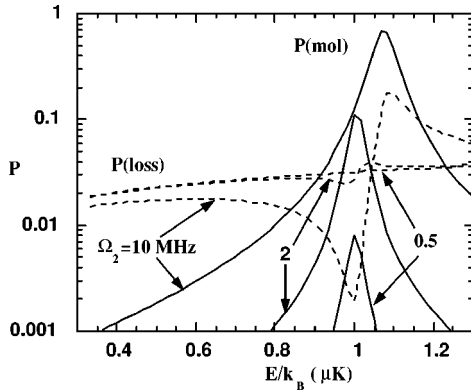


FIG. 2. Calculated molecule production and trap loss probabilities, $P(\text{mol})$ (solid) and $P(\text{loss})$ (dashed), versus collision energy for three cases of optical coupling between the bound levels v_1 and v_2 . Laser 1 is tuned on-resonance with level v_1 , and laser 2 is tuned to place the v_2 Raman resonance with a 0.8-kHz width at $E/k_B = 1 \mu\text{K}$. The listed $\Omega_2/2\pi$ does not include the Franck-Condon factor $|\langle v_1 | v_2 \rangle|^2 = 0.00205$.

Figure 2 shows $P(\text{mol})$ and $P(\text{loss})$ for several optical couplings Ω_2 and for a range of collision energies E near $E/k_B = 1 \mu\text{K}$, a relatively high collision energy range that might characterize an evaporatively cooled gas before reaching condensation. We represent collision energy in temperature units by dividing E by the Boltzmann constant k_B . The actual rate coefficients should average these energy-dependent probabilities over the distribution of collision energies in the gas. The Ω_2 value in Fig. 2 must be multiplied by the Franck-Condon factor $|\langle v_1 | v_2 \rangle|^2 = 0.00205$ to get the actual optical coupling. The optical coupling Ω_1 is fixed at $2\pi(6800 \text{ MHz})$, which is beyond the saturation limit where the free-bound transition would be driven to saturation according to the criterion of Ref. [13], that is, the stimulated rate, $\gamma_s = (2\pi/\hbar)|\langle v_1 | \hbar\Omega_1 | E \rangle|^2$, is larger than the spontaneous rate γ_1 . This large Ω_1 , implying a laser power on the order of 20 kW/cm^2 , is needed because of the very large $\Delta_A = 2\pi(2.4 \text{ THz})$ for the resonance level used. For other levels the intensity should scale inversely as $|\langle v_1 | E \rangle|^2$, and Ω_1 could easily be reduced by two orders of magnitude, and I_1 by four orders of magnitude, by selecting a resonance with $\approx 24 \text{ GHz}$ binding energy (such a v_2 implies a v_1 level close to its dissociation limit to have a favorable $|\langle v_1 | v_2 \rangle|^2$). For the lowest $\Omega_2 = 0.5 \text{ MHz}$, Fig. 2 shows that $P(\text{loss})$ is flat as a function of energy (the width 10 MHz of the level v_1 corresponds to a collision energy spread of $\hbar\gamma_1/k_B = 500 \mu\text{K}$); $P(\text{mol})$ exhibits a sharp Raman resonance with a width $\hbar\gamma_2/k_B = 40 \text{ nK}$, and $P(\text{mol}) \ll P(\text{loss})$. As Ω_2 is increased, the strength of the resonance increases linearly until it saturates and the resonance exhibits power broadening and a shift in its peak position. $P(\text{loss})$ clearly exhibits the effect of the Raman resonance, with $P(\text{loss}) \ll P(\text{mol})$ over a significant range of collision energies near the resonance position, in accordance with the qualitative predictions of Ref. [13]. Even if averaged over a Maxwellian velocity distribution that would characterize a thermal gas near $1 \mu\text{K}$, the molecule production rate can be driven to near its upper bound given by unitarity of the S matrix, and the rate coefficient $K(\text{mol})$ can be comparable to or larger than $K(\text{loss})$.

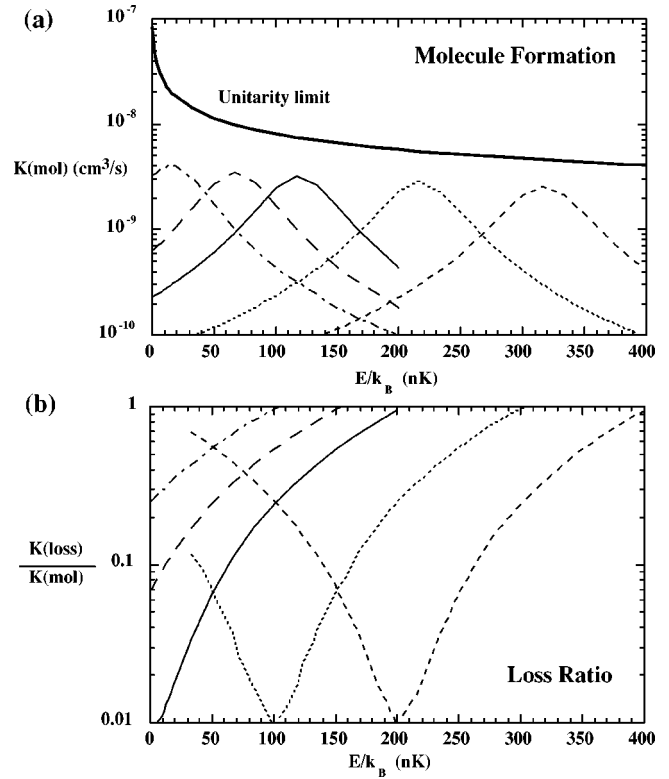


FIG. 3. Calculated rate coefficients versus E/k_B : (a) $K(\text{mol})$ for molecule production and (b) $K(\text{loss})$ for trap loss for five different cases of ω_2 detuning that place the Raman resonance close to the $E=0$ collision energy. Ω_1 and Ω_2 are kept constant at the values used in Fig. 2 for the largest $P(\text{mol})$ case. The solid line case exhibits the lowest $K(\text{loss})/K(\text{mol})$ ratio in the 1-nK region.

The position of the resonance depends on the detuning $\Delta\omega = \omega_1 - \omega_2$. Figure 3(a) shows how $K(\text{mol})$ varies as ω_2 is tuned to place the resonance close to zero collision energy, appropriate to a condensate, where the order of magnitude of the kinetic energy can be estimated from $kX \approx 1$, with X a characteristic size of the mean-field wave function in the confining trapping potential. This predicts typical kinetic energies on the order of 1 nK in a condensate. Figure 3(a) clearly shows that $K(\text{mol})$ remains large even for E/k_B as small as 1 nK. The threshold law requires that $K(\text{mol})$ approach a constant value as $T \rightarrow 0$. Figure 3(b) shows that $K(\text{loss})$ can be strongly reduced by the existence of the resonance, remaining up to two orders of magnitude less than $K(\text{mol})$ in this extreme low-temperature limit.

Taking $10^{-10} \text{ cm}^3/\text{s}$ as a conservative estimate for $K(\text{mol})$ in a condensate, the time scale for converting $1/e$ of the atoms in a condensate is $[K(\text{mol})n]^{-1} = 100 \mu\text{s}$ at the $n = 10^{14} \text{ cm}^3/\text{s}$ density characteristic of magnetic traps, and only $3 \mu\text{s}$ at the $n = 3 \times 10^{15} \text{ cm}^3/\text{s}$ density possible in an optical trap [15]. If $K(\text{loss})/K(\text{mol}) = 0.01$, a laser pulse on the order of $1 \mu\text{s}$ would convert a large fraction of the atoms in the condensate into the target molecular level, with about half of the atoms being lost via the uncontrolled spontaneous decay channel. A shorter pulse could convert a smaller fraction of atoms, but with greatly reduced losses.

These calculations suggest that stimulated Raman pulses can be used to form a large number of molecules from the atom pairs in a Bose-Einstein condensate on a time scale

short compared to the oscillation period in the harmonic trap. This process is analogous to the use of an rf pulse to rapidly flip the internal spin state of condensed atoms, causing a coherent matter wave in the outcoupled (i.e., untrapped) spin state to be produced [4]. We have assumed that the molecules produced by the coherent Raman process, which operates on the internal relative motion coordinate of atom pairs, are produced in an untrapped state so that the center of mass of the associated pair (i.e., the molecule) moves out of the trap. This will result in a “pulsed molecule laser” analogous to the “pulsed atom laser” of Ref. [4], with similar coherence properties expected for the molecule wave. Molecules produced in the $X^1\Sigma_g^+$ state will not be trapped by a magnetic field, whereas $^3\Sigma_u^+$ molecules may be trapped or not, depending on the specific molecular Zeeman levels produced. If the molecules remained trapped, and did not exit by the γ_2 process, a different theoretical approach appropriate to initial and final bound states would be necessary. However, our calculations suggest that coherent conversion of atom pairs in the atomic condensate to trapped molecules may be feasible.

An important experimental issue for making our molecule production scheme viable is the ability to situate the Raman resonance within the kinetic-energy spread of the cold atomic gas or the condensate. With the parameters we have used, assuming a residence time broadening on the order of 1

kHz, this requires locking the frequencies of the two lasers to a difference defined to within a kHz. This is relatively straightforward to do if the frequencies do not differ by more than a few GHz. Thus, it may be best to try to produce molecules in very high-lying, weakly bound levels of the lowest two potentials. Such levels have been measured for the Rb_2 molecule using two-color photoassociation spectroscopy [16], and presumably such measurements could be done for the Na_2 molecule as well.

Finally, we note that if the excitation laser ω_1 is tuned *between* molecular photoassociation resonances, the binary pair light scattering rate will be strongly reduced, and in fact will be small compared to the free atom light scattering rate [10]. In this case, it may be feasible to use Raman pulses in lieu of rf pulses to induce the spin flip of the atomic Zeeman sublevels, say, from the trapped $F=1, M=-1$ level to the untrapped $F=1, M=0$ level. Thus, by tuning the two lasers, it may be possible to switch between outcoupled atoms and outcoupled molecules. The prospects of using a stimulated Raman adiabatic passage (STIRAP) process [6] in this context should be investigated.

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