

Creating Ground State Molecules with Optical Feshbach Resonances in Tight Traps

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We propose to create ultracold ground state molecules in an atomic Bose-Einstein condensate by adiabatic crossing of an optical Feshbach resonance. We envision a scheme where the laser intensity and possibly also frequency are linearly ramped over the resonance. Our calculations for ⁸⁷Rb show that for sufficiently tight traps it is possible to avoid spontaneous emission while retaining adiabaticity, and conversion efficiencies of up to 50% can be expected.

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The formation of ultracold molecules and the creation of molecular Bose-Einstein condensates (BEC) [1] open the way to study new collective phenomena and a new, ultracold chemistry [2,3]. Since no direct cooling method for molecules can reach the transition temperature for BEC, most experiments start from ultracold atoms. Molecules are created by applying an external field, either magnetic [4] or optical [5], to two colliding atoms. This process is described in terms of a Feshbach resonance (FR) [6] where the energies of two colliding atoms and of a bound molecular level coincide. Magnetic FRs have been particularly successful in creating alkali dimer molecules [4,7,8]. In contrast, optical FRs involve electronically excited states, where spontaneous emission may lead to loss of coherence [2,5]. Apart from this obstacle, optical FRs have the advantage that optical transitions are almost always available, whereas magnetic FRs require the presence of a hyperfine manifold of the atom and may occur at magnetic field strengths which are difficult to obtain in experiments. Furthermore, optical FRs offer more flexibility since two parameters (laser intensity and frequency) instead of just one (magnetic field strength) can be tuned. While optical FRs have been employed to create molecules in cold gases via photoassociation (PA) [9] and to tune the scattering length [10], they have not yet been used to coherently create molecules except for the recent work of Ref. [2].

In this Letter, we propose to employ optical FR to create weakly bound ground state molecules (in singlet and triplet ground state potentials both labeled “ground state” in the following). In analogy to magnetic FR, we envisage a scheme of adiabatically ramping over the resonance (cf. Fig. 1). The resulting wave function has components on both electronic ground and excited states with the latter being subject to spontaneous emission losses. In a second step, the laser field therefore needs to be switched off. This corresponds to projecting the wave function onto the field-free eigenstates.

The goal is to sweep intensities and frequencies such that this projection is predominantly onto the last bound level of the ground state, i.e., onto stable molecules. Our scheme is different from one-color PA [11] which popu-

lates *excited* state levels. It is rather similar in spirit to stimulated Raman adiabatic passage [12] in that population of the excited state is minimized using a two-photon transition. It differs from two-color PA [2] since the sudden switch-off breaks the symmetry of the coupling between the bound molecular level and the trap (or continuum) state. We show that for sufficiently tight traps, intensity and frequency of the field can be tuned such that spontaneous emission losses are avoided while adiabaticity is retained. Such ramps can be realized experimentally employing acousto-optical modulators or diode lasers. Sufficiently tight confinement can be reached in microscopic dipole traps [13] or deep optical lattices [14].

Our calculations are performed for ⁸⁷Rb. The generality of the scheme is emphasized by employing both singlet and triplet ground state potentials. We consider two ⁸⁷Rb atoms which collide in an isotropic harmonic trap and interact with a continuous wave (cw) laser field. The center of mass motion is decoupled, and the dynamics in the internuclear distance R is governed by the Hamiltonian

$$\hat{H} = \begin{pmatrix} \hat{H}_g & \hbar\Omega \\ \hbar\Omega & \hat{H}_e - \hbar(\omega_0 - \Delta_L) - \frac{i\hbar}{2}\Gamma \end{pmatrix}. \quad (1)$$

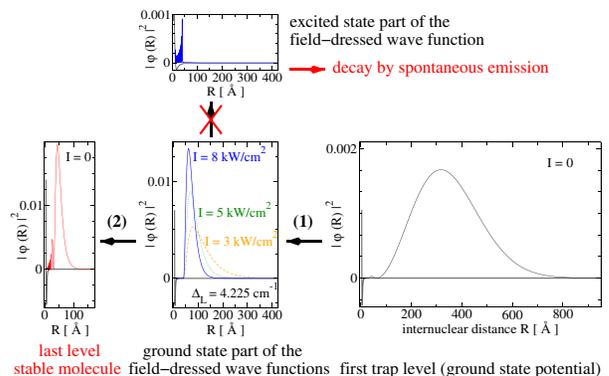


FIG. 1 (color online). Our proposed scheme for the creation of molecules: step (1) is an adiabatic ramp of the laser intensity (and possibly frequency), while step (2) is a sudden switch-off of the laser (wave functions shown for $\nu_{tr} = 50$ kHz).

$\hat{H}_{g(e)} = \hat{T} + V_{g(e)}(\hat{\mathbf{R}}) + (-)V_{\text{tr}}(\hat{\mathbf{R}})$ is the single channel Hamiltonian with \hat{T} the kinetic energy operator and $V_{g(e)}(\hat{\mathbf{R}})$ the ground (excited) state interaction potential. $V_{\text{tr}}(\hat{\mathbf{R}}) = \frac{1}{2}m\omega_{\text{tr}}^2\hat{\mathbf{R}}^2$ is the potential of the dipole trap, and Γ the decay rate modeling spontaneous emission. m denotes the reduced mass and ω_{tr} the frequency of the trap ($\omega_{\text{tr}} = 2\pi \times \nu_{\text{tr}}$). The frequency of the laser, $\omega_L = \omega_0 - \Delta_L$, is red detuned by Δ_L relative to the atomic resonance at ω_0 . In Eq. (1), we invoke the dipole and rotating wave approximations (RWA). The Rabi frequency Ω is then given by $\Omega = E_0\vec{D}(\hat{\mathbf{R}}) \cdot \vec{\epsilon} \approx E_0\vec{D} \cdot \vec{\epsilon}$, where E_0 is the amplitude of the laser field, $\vec{D}(\hat{\mathbf{R}})$ the dipole moment, and $\vec{\epsilon}$ the polarization vector of the laser field. $\vec{D}(\hat{\mathbf{R}}) \cdot \vec{\epsilon}$ is approximated by its asymptotic value deduced from standard long-range calculations [15]. In Eq. (1), we neglect the hyperfine structure. This is justified for sufficiently detuning the laser from the atomic resonances (about 4 cm^{-1} or 120 GHz ; the largest energy difference between hyperfine levels is 7 GHz between $F = 1$ and $F = 2$ for $5^2S_{1/2}$). The potentials $V_{g(e)}(\hat{\mathbf{R}})$ have been obtained by matching the results of *ab initio* calculations [16] to the long-range dispersion potentials $V_{\text{asy}}(\hat{\mathbf{R}}) = (C_3/\hat{\mathbf{R}}^3 +)C_6/\hat{\mathbf{R}}^6 + C_8/\hat{\mathbf{R}}^8$. The coefficients for the $5S + 5S$ and $5S + 5P$ asymptote are found in Refs. [17,18]. The repulsive barrier of the ground state potential has been adjusted to give a triplet (singlet) scattering length of $100 a_0$ ($90 a_0$). The Hamiltonian, Eq. (1), is represented on a grid, employing a mapping procedure [19] which reduces the number of required grid points by a factor of 5 to 30.

We proceed in two steps. First, we diagonalize the Hamiltonian, Eq. (1), and obtain the field-dressed eigenstates and eigenenergies as a function of laser intensity and frequency. The term $-i\hbar\Gamma/2$ causes the Hamiltonian to be non-Hermitian with complex eigenvalues. Γ is assumed to be independent of $\hat{\mathbf{R}}$ which is consistent with the approximation $\vec{D}(\hat{\mathbf{R}}) \approx \vec{D}$. Therefore, the imaginary part of the eigenvalues becomes $\Gamma/2$ times the projection of the eigenfunction onto the excited state [20]. In a second step, we solve the time-dependent Schrödinger equation to illustrate the creation of molecules. Γ is then set equal to its asymptotic value, $\sqrt{2}\Gamma_{\text{at}}$, with $\Gamma_{\text{at}} = \hbar/\tau_{\text{at}}$ and $\tau_{\text{at}}(5S + 5P_{3/2}) = 26.24 \text{ ns}$, $\tau_{\text{at}}(5S + 5P_{1/2}) = 27.70 \text{ ns}$.

The following calculations are performed for transitions between the triplet ground state $a^3\Sigma_u^+(5S + 5S)$ and the $0_g^-(5S + 5P_{3/2})$ excited state. Figure 2(a) shows the binding energy of the last bound level below the $(5S + 5S)$ asymptote as a function of laser intensity and detuning. The range of detunings is chosen around 4 cm^{-1} , large enough to avoid hyperfine coupling and small enough such that the resonances occur with excited state levels ν' which have a good Franck-Condon overlap with the last bound ground state level. Two resonances are found within this range ($\nu' = 40$ at 4.225 cm^{-1} and $\nu' = 41$ at 3.98 cm^{-1}).

In Fig. 2(b), the energies of the four last bound levels below the $(5S + 5S)$ asymptote are plotted versus laser intensity for a specific detuning. Resonances at about 2.5 kW/cm^2 , 16.5 kW/cm^2 , and 36.5 kW/cm^2 are observed. At each resonance, the number of bound states is increased by one. Usually, only the detuning is varied in optical FR. The increase in the number of bound states can then be understood as follows: in the RWA, ground and excited state potentials cross, and the excited state asymptote is at $\hbar\Delta_L$ above the ground state dissociation limit. Decreasing the detuning therefore pushes one more excited state level below this dissociation limit. The same happens as intensity is increased. It corresponds to the light shifts displacing the resonance positions with increasing intensity [cf. Fig. 2(a)]. To further illustrate this “creation” of bound levels, Fig. 1 (middle) shows the projection onto the ground state of one field-dressed wave function, $|\langle g|\varphi_{n=81}^\Omega\rangle|^2$ for different intensities, i.e., different Ω (n counts all eigenstates). At $I = 0$, $|\varphi_{n=81}^{\Omega=0}\rangle$ coincides with the lowest trap state (Fig. 1, right). As the intensity is increased, the wave function is deformed and pushed toward shorter internuclear distances such that it eventually resembles the wave function of the last bound level (Fig. 1, left) [21]. The first step in our scheme is therefore a slow ramp in intensity (and possibly frequency) such that the wave function adiabatically follows the field-dressed eigenfunctions, $|\varphi_{n=81}^\Omega\rangle$. In a second step, the field should be suddenly switched off projecting the field-dressed onto the field-free eigenfunctions. The probability to form a ground state molecule is then given by the projection of the field-dressed eigenfunction onto the last bound ground state level, $P_{\text{mol}} = |\langle \varphi_{\text{last}}^g|\varphi_{n=81}^\Omega\rangle|^2$ (also lower bound levels can contribute to molecule formation, but this is much

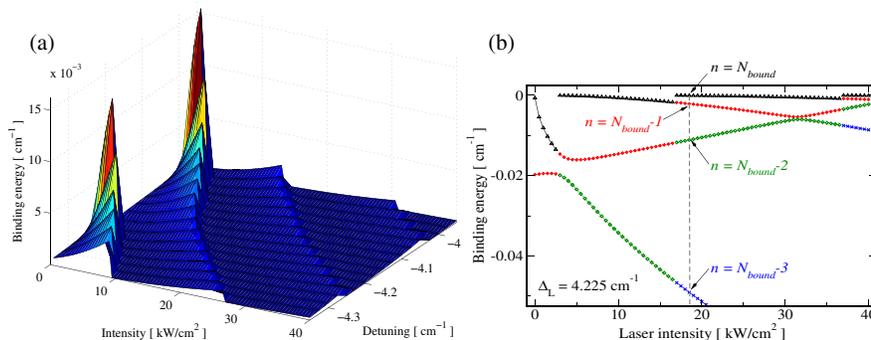


FIG. 2 (color online). (a) Binding energy of the last bound level below the $(5S + 5S)$ asymptote as a function of laser intensity and detuning. (b) Binding energy of the last bound levels n as a function of intensity. For $I = 19 \text{ kW/cm}^2$ (vertical line) there are N_{bound} levels below the $(5S + 5S)$ asymptote.

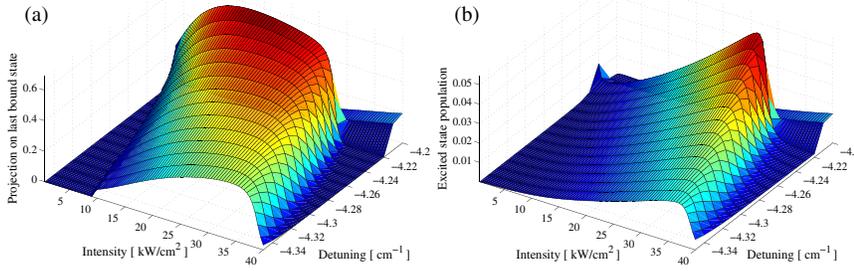


FIG. 3 (color online). Probability of molecule formation P_{mol} (a) and excited state component p_{exc} (b) as function of laser intensity and detuning ($\nu_{\text{tr}} = 250$ kHz).

less likely). The lifetime of the field-dressed eigenfunction, $\tau_v = \tau_{\text{at}}/(\sqrt{2}p_{\text{exc}})$, is determined by its excited state component, $p_{\text{exc}} = |\langle e | \varphi_{n=81}^\Omega \rangle|^2$. That is, P_{mol} corresponds to a gain while p_{exc} might lead to a loss. Both are shown in Fig. 3 as a function of laser intensity and detuning.

Close to resonance with an excited state level ($\Delta_L = -4.225$ cm $^{-1}$), at moderate intensities (5 kW/cm $^2 \leq I \leq 10$ kW/cm 2) the projection onto the last bound level is 50% and higher, while the excited state population does not exceed 0.01; i.e., the lifetime of the field-dressed eigenstate is ≥ 2 μs . This lifetime defines an upper limit for the time window within which the ramp across the resonance should be completed. The lower limit, T_{ad} , is due to the requirement of adiabaticity. It is determined by the vibrational period of the lowest trap state, T_{vib} , which depends on the trap frequency and the interaction potential (cf. Table I).

We can now estimate the time scales for our scheme: assuming the ramp should be performed in a time $T_{\text{ad}} = 5 \times T_{\text{vib}}$ to be adiabatic, spontaneous emission losses should be minimal for a trap frequency of $\nu_{\text{tr}} \geq 250$ kHz ($T_{\text{ad}} \approx 210$ ns). For $\nu_{\text{tr}} \approx 50$ kHz, T_{ad} (≈ 1.4 μs) and τ_v are on the same order of magnitude, and spontaneous emission losses will play a role.

To verify our conclusions from the time-independent picture, we have explicitly studied the creation of molecules solving the time-dependent Schrödinger equation,

$$i\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = \hat{\mathbf{H}}(t) |\Psi(t)\rangle, \quad (2)$$

with a Chebychev propagator. The time dependence in $\hat{\mathbf{H}}(t)$ is due to the linear ramp in Ω (i.e., E_0 or \sqrt{I}) and ω_L (i.e., Δ_L), respectively. Figure 4 shows the projection of the wave function $\Psi(R;t)$ onto the last bound level and onto the lowest trap levels of the field-free Hamiltonian versus time. Also plotted is the overall loss due to sponta-

neous emission (we assume that any population undergoing spontaneous emission is lost from the trap).

We combined a ramp in intensity with a ramp in frequency; both ramps are performed within 85 ns and 540 ns, respectively, i.e., $2 \times T_{\text{vib}}$. This turned out to be sufficient to retain adiabaticity. While this result might be surprising at first glance, it reflects that the main source of nonadiabaticity is Rabi cycling which in turn is suppressed by spontaneous emission. Simulations *without* spontaneous emission showed that each ramp time had to be at least $5 \times T_{\text{vib}}$ for the ramp to be adiabatic. Figure 4(c) shows that for $\nu_{\text{tr}} = 250$ kHz, almost 50% of the population can be converted into ground state molecules, while about 10% are lost. The remaining population is distributed over the lowest trap states. For $\nu_{\text{tr}} = 50$ kHz [Fig. 4(d)] the losses are somewhat higher at 24%, but the conversion probability still reaches almost 30%. Conversion probabilities higher than 50% could be obtained for even tighter traps allowing for faster ramps.

We found that combining a ramp in intensity with a ramp in frequency is the most efficient way to create molecules. The initial ramp in intensity is performed with the laser frequency tuned in between two resonances. Such a ramp deforms the wave function already consider-

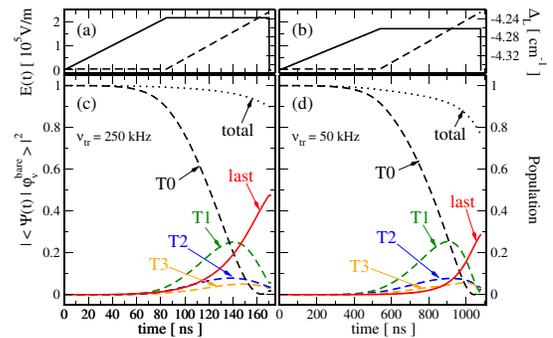


FIG. 4 (color online). Calculations for a trap frequency $\nu_{\text{tr}} = 250$ kHz (left) and $\nu_{\text{tr}} = 50$ kHz (right). (c)–(d) Projection of the time-dependent wave function onto the last bound ground state level (solid red line) and the first trap states T0–T3 (dashed lines) of the bare Hamiltonian. Also shown is the total population $|\langle \Psi(t) | \Psi(t) \rangle|^2$ (dotted line). (a)–(b) Variation of the laser field strength $E(t)$ (solid line) and detuning Δ_L (dashed line). The maximum field intensity is respectively $I = 8$ kW/cm 2 and $I = 5$ kW/cm 2 .

TABLE I. Vibrational periods of the lowest trap state (calculated from its eigenenergy) for the triplet and singlet ground state potentials.

ν_{tr}	1 kHz	50 kHz	100 kHz	250 kHz	500 kHz
$T_{\text{vib}}(d^3\Sigma_u^+)$	16.6 μs	270 ns	124 ns	42.4 ns	18.5 ns
$T_{\text{vib}}(X^1\Sigma_g^+)$	16.6 μs	292 ns	137 ns	48.7 ns	21.4 ns

TABLE II. Final probability of ground state molecule formation, $P = |\langle \Psi(t_{\text{final}}) | \varphi_g^{\text{bare}} \rangle|^2$, for different switch-off times of the laser.

T_{switch}	0	10 ns	5 ns	1 ns
P with $\nu_{\text{tr}} = 250$ kHz	0.475	0.269	0.389	0.466
P with $\nu_{\text{tr}} = 50$ kHz	0.288	0.203	0.246	0.284

ably while keeping p_{exc} [cf. Fig. 3(b)] and hence the spontaneous emission loss extremely small. In a second step the frequency is ramped toward the resonance. This ensures a maximum overlap with the bound state wave function [cf. Fig. 3(a)]. Ramping up the intensity with the laser tuned in between two resonances is not advantageous to create molecules. The overlap with the trap levels exceeds in this case by far the overlap with the bound state, i.e., one mainly excites higher trap states. If the intensity is ramped up with the laser tuned close to resonance, due to the stronger coupling with the excited state, the ramp needs to be slower to be adiabatic, and spontaneous emission losses are larger (cf. Fig. 3).

Finally, since in the experiment the laser cannot be switched off instantly, we address the question of a finite switch-off time after the ramp. As can be seen in Table II, the field should ideally be switched off within 1 ns. The probability of molecule formation is then reduced by less than 1%. Switching off the field within 10 ns causes about one third of the created molecules to be lost, i.e., to be transferred back into a pair of trapped atoms. Because of the shorter time scales, this problem is more severe in tighter traps.

We have performed the same set of calculations for a transition between the $X^1\Sigma_g^+(5S+5S)$ and $0_u^+(5S+5P_{1/2})$ potentials, which differ markedly from the previous ones. The common point is the asymptotic $1/R^3$ behavior in the excited state potential, providing extremely long-range levels with a large Franck-Condon overlap with the lowest trap state(s). We therefore find exactly the same pattern of binding energies, probability of molecule formation, and excited state population as shown in Figs. 2–4.

To summarize, we have shown that in tight traps, loosely bound ground state molecules can be created efficiently and without loss of coherence by adiabatic ramping over an optical FR. Both detuning and intensity are varied within an asymmetric scheme which involves first adiabatic following and then a fast switch-off. Three-body effects are neglected in our model: it is therefore applicable to low densities or ideally to a Mott-Insulator state [14]. We assumed the trap to be isotropic. However, we expect our scheme to work also for anisotropic traps as long as no new time scale is introduced, i.e., as long as the lowest trap frequency is ≥ 50 kHz. We point out that in shallow traps the creation of molecules by optical FR does not seem to be feasible. In that case nonadiabatic processes or adiabatic schemes employing short laser pulses [22] should be considered. For tight traps by contrast, calculations performed

for rubidium atoms demonstrated conversion probabilities up to 50%. The presented scheme should work for any optical transition which is also efficient in photoassociation, involving long-range wells or resonant coupling in the excited state [23]. The range of possible applications therefore extends well beyond homonuclear systems and alkali atoms.

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