Creating Ground State Molecules with Optical Feshbach Resonances in Tight Traps

Christiane P. Koch,1,2 Françoise Masnou-Seeuws,1 and Ronnie Kosloff2

1Laboratoire Aimé Cotton, CNRS, Bât. 505, Campus d’Orsay, 91405 Orsay Cedex, France
2Department of Physical Chemistry and The Fritz Haber Research Center, The Hebrew University, Jerusalem 91904, Israel

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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) open the way to study new collective phenomena and a new, ultracold chemistry. Since no direct cooling method for molecules can reach the transition temperature for BEC, the formation of molecules from ultracold atoms has been a focus of recent research. Molecules are created by applying an external field, either magnetic or optical, to two colliding atoms. This process is described in terms of a Feshbach resonance (FR), where the collision energy of the two atoms coincides with the energy of a bound molecular level. Magnetic FR have been particularly successful in creating alkali dimer molecules, even heteronuclear. In contrast, optical FR involve electronically excited potentials, where spontaneous emission may lead to loss of coherence. Apart from this obstacle, optical FR have the advantage that optical transitions are almost always available, whereas magnetic FR 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 FR offer more flexibility since two parameters (laser intensity and frequency) instead of just one (magnetic field strength) can be tuned. While optical FR have been employed to create molecules in cold gases via photoassociation (PA) and to tune the scattering length, 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 labelled ‘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, which populates excited state levels. It is rather similar in spirit to Stimulated Raman Adiabatic Passage in that population of the excited state is minimized using a two-photon transition. It differs from two-color PA 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 or deep optical lattices.

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

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\text{d} \phi(R) / \text{d}R = -i \left[ H_{\text{field}}, \phi(R) \right] + \text{Raman term}
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the Hamiltonian

\[ \hat{\mathcal{H}} = \left( \hat{\mathcal{H}}_g + \hat{\mathcal{H}}_e - \hbar \omega - \Delta_L - i \frac{\Gamma}{2} \right). \]  

(1)

\[ \hat{\mathcal{H}}_g(e) = \hat{T} + V_g(e)\hat{\mathbf{R}} + (-)V_{tr}(\hat{\mathbf{R}}) \]

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. \( V_{tr}(\hat{\mathbf{R}}) = \frac{1}{2}m\omega_L^2\hat{\mathbf{R}}^2 \) is the potential of the dipole trap, and \( \Gamma \) the decay rate modelling spontaneous emission. \( m \) denotes the reduced mass and \( \omega_L \) the frequency of the trap \( (\omega_L = 2\pi \times v_L) \). The frequency of the laser, \( \omega_L = \omega_0 - \Delta_L \) is red-detuned by \( \Delta_L \) relative to the atomic resonance at \( \omega_0 \). In Eq. (1), we invoke the dipole and rotating wave approximations (RWA). The Rabi frequency \( \Omega \) is then given by \( \Omega = E_0\hat{D}(\hat{\mathbf{R}}) \cdot \hat{\epsilon} \approx E_0\hat{D} \cdot \hat{\epsilon}, \)

where \( E_0 \) is the amplitude of the laser field, \( \hat{D}(\hat{\mathbf{R}}) \) the dipole moment and \( \hat{\epsilon} \) the polarization vector of the laser field. \( \hat{D}(\hat{\mathbf{R}}) \cdot \hat{\epsilon} \) is approximated by its asymptotic value deduced from standard long range calculations [19]. 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 \( ^5S_{1/2} \)). The potentials \( V_g(e)\hat{\mathbf{R}} \) have been obtained by matching the results of \( ab \) initio calculations [17] to the long-range dispersion potentials \( V_{any}(\hat{\mathbf{R}}) = (C_6/\hat{\mathbf{R}}^6) + C_8/\hat{\mathbf{R}}^8 \). The coefficients for the \( 5S + 5S \) and \( 5S + 5P \) asymptote are respectively found in Ref. [18] and Ref. [19]. The repulsive barrier of the ground state potentials 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 [20] 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. \( \Gamma \) is assumed to be independent of \( \hat{\mathbf{R}} \) which is consistent with the approximation \( \hat{D}(\hat{\mathbf{R}}) \approx \hat{D} \). Therefore the imaginary part of the eigenvalues becomes \( \Gamma/2 \) times the projection of the eigenfunction onto the excited state [22]. In a second step, we solve the time-dependent Schrödinger equation to illustrate the creation of molecules. \( \Gamma \) is then set equal to its asymptotic value, \( \sqrt{2}\Gamma_{at} \) with \( \Gamma_{at} = h/\tau_{at} \) and \( \tau_{at}(5S + 5P_{3/2}) = 26.24 \text{ ns}, \tau_{at}(5S + 5P_{1/2}) = 27.70 \text{ ns}. \)

The following calculations are performed for transitions between the triplet ground state \( a^3Σ_u^+(5S+5S) \) and the \( 0^+_g \) \( (5S+5P_{1/2}) \) excited state. Fig. 2 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 \( \nu' \) which have a good Franck-Condon overlap with the last bound ground state level. Two resonances are found within this range \( (\nu' = 40 \text{ at } 4.225 \text{ cm}^{-1}, \nu' = 41 \text{ at } 3.98 \text{ cm}^{-1}) \). In Fig. 2a, the energies of the four last bound levels below the \( (5S+5S) \) asymptote are plotted vs. 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 potential cross and the excited state asymptote is at \( h\Delta_L \) 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. 2a). To further illustrate this "creation" of bound levels, Fig. 2b (middle) shows the projection onto the ground state of one field dressed wave function, \( |\langle g|\Psi_{n=81}^\Omega \rangle|^2 \) for different intensities, i.e., different \( \Omega \) (\( n \) counts all eigenstates). At \( I = 0 \), \( |\langle g|\Psi_{n=81}^\Omega \rangle|^2 \) coincides with the lowest trap state (Fig. 2 right). As the intensity is increased, the wave function is deformed and pushed toward shorter inter-nuclear distances such that it eventually resembles the wave function of the last bound level (Fig. 2 left) [22].

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, \( |\Psi_{n=81}^\Omega \rangle \). 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, \( P_{mol} = |\langle \Psi_{n=81}^\Omega |\Psi_{n=81}^\Omega \rangle|^2 \) (also lower bound levels can contribute to molecule formation, but this is much less likely). The lifetime of the field-dressed eigenfunction, \( \tau_n = \tau_{at}/(\sqrt{2}p_{exc}) \), is determined by its excited state component, \( p_{exc} = |\langle e|\Psi_{n=81}^\Omega \rangle|^2 \). That is,
$P_{\text{mol}}$ corresponds to a gain while $p_{\text{exc}}$ might lead to a loss. Both are shown in Fig. 4 as a function of laser intensity and detuning. Close to resonance with an excited state level ($\Delta_L = -4.225 \text{ cm}^{-1}$), at moderate intensities ($5 \text{ kW/cm}^2 \leq I \leq 10 \text{ kW/cm}^2$) 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 $\geq 2 \mu$s. This lifetime defines an upper limit for the time window within which the ramp across the resonance should be completed. The lower limit, $T_{\text{ad}}$, is due to the requirement of adiabaticity. It is determined by the vibrational period of the lowest trap state, $T_{\text{vib}}$, which depends on the trap frequency and the interaction potential (cf. Table I). We can now estimate the timescales 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 $\nu_T \geq 250 \text{ kHz}$ ($T_{\text{ad}} \approx 210 \text{ ns}$). For $\nu_T \approx 50 \text{ kHz}$, $T_{\text{ad}} \approx 1.4 \mu$s and $\tau_p$ 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,

$$ih\frac{\partial}{\partial t}|\Psi(t)\rangle = \hat{H}(t)|\Psi(t)\rangle,$$

with a Chebychev propagator. The time-dependence in $\hat{H}(t)$ is due to the linear ramp in $\Omega$ (i.e. $E_0$ or $\sqrt{T}$) and $\omega_L$ (i.e. $\Delta_L$), respectively. Fig. 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 vs. time. Also plotted is the overall loss due to spontaneous emission (we assume that any population undergoing spontaneous emission is lost from the coherence of the scheme, and possibly 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. Fig. 4 shows that for $\nu_T = 250 \text{ kHz}$, 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 $\nu_T = 50 \text{ kHz}$ (Fig. 4b) 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 considerably while keeping the excited state population (cf. Fig. 4a) 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. 4b). 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

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

| $\nu_T$     | 1 kHz | 50 kHz | 100 kHz | 250 kHz | 500 kHz |
|-------------|-------|--------|---------|---------|---------|
| $T_{\text{vib}}(a^3\Sigma^+_g)$ | 16.6 $\mu$s | 270 ns | 124 ns | 42.4 ns | 18.5 ns |
| $T_{\text{vib}}(X^1\Sigma^+_g)$ | 16.6 $\mu$s | 292 ns | 137 ns | 48.7 ns | 21.4 ns |

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

**FIG. 4:** (Color online) (c)+(d): Projection of the time-dependent wave function onto the last bound ground state level (solid red line) and the first trap states $T$ (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 $\Delta_L$ (dashed line). The maximum field intensity is $I = 8 \text{ kW/cm}^2$ for $\nu = 250 \text{ kHz}$ and $I = 5 \text{ kW/cm}^2$ for $\nu = 50 \text{ kHz}$. 

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

| \( T_{\text{switch}} \) (ns) | 0 | 10 ns | 5 ns | 1 ns |
|-----------------------------|---|------|------|------|
| \( P \) with \( \nu = 250 \) kHz | 0.475 | 0.269 | 0.389 | 0.466 |
| \( P \) with \( \nu = 50 \) kHz | 0.288 | 0.203 | 0.246 | 0.284 |

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 adiabatically following and then a fast switch-off. Three-body effects are neglected in our model: it is therefore adiabatic following and then a fast switch-off. Three-body effects are neglected in our model: it is therefore adiabatic following and then a fast switch-off. Three-body effects are neglected in our model: it is therefore adiabatic following and then a fast switch-off. Three-body effects are neglected in our model: it is therefore adiabatic following and then a fast switch-off.

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