Coherent Optical Transfer of Feshbach Molecules to a Lower Vibrational State

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Using the technique of stimulated Raman adiabatic passage (STIRAP) we have coherently transferred ultracold ^{87}Rb$_2$ Feshbach molecules into a more deeply bound vibrational quantum level. Our measurements indicate a high transfer efficiency of up to 87%. Because the molecules are held in an optical lattice with not more than a single molecule per lattice site, inelastic collisions between the molecules are suppressed and we observe long molecular lifetimes of about 1 s. Using STIRAP we have created quantum superpositions of the two molecular states and tested their coherence interferometrically. These results represent an important step towards Bose-Einstein condensation of molecules in the vibrational ground state.

Recently, there has been a rapidly growing interest in ultracold molecules since they lend themselves to a large number of interesting studies in, for instance, few body collision physics [1–3], chemistry in the ultracold regime, high resolution spectroscopy, as well as quantum computation [4]. Furthermore, molecules in their vibrational ground state are of special interest, because they allow for the formation of an intrinsically stable molecular Bose-Einstein condensate (BEC). Current pathways towards the production of ultracold molecules in well-defined quantum states are either based on sympathetic cooling [5] or association of ultracold neutral atoms using photoassociation [6] or Feshbach resonances [7]. The method of Feshbach ramping has proved especially successful and efficient, but it only produces molecules in the last bound vibrational level. In order to selectively convert molecules into more deeply bound states, it has been proposed [8] to use a sequence of stimulated optical Raman transitions to step molecules down the vibrational ladder. This process takes place while the molecules are held in an optical lattice isolating them from each other and thus shielding them from detrimental collisions. Recently, optical transfer of molecules into their vibrational ground state was demonstrated experimentally using a "pump-dump" method without a lattice at a moderate efficiency and selectivity [9].

Here we report the realization of an efficient and highly selective transfer scheme, where an ensemble of ^{87}Rb$_2$ Feshbach molecules in an optical lattice is coherently converted to a deeper bound molecular state via stimulated Raman adiabatic passage (STIRAP). STIRAP is known as a fast, efficient, and robust process for population transfer based on a Raman transition [10]. During transfer it keeps the molecules in a dark superposition state, which decouples from the light and thus suppresses losses due to spontaneous light scattering. In our proof-of-principle ex-

periment we transfer the Feshbach molecules with a STIRAP pulse from their last bound vibrational level (binding energy 24 MHz $\times$ h), which we denote $|a\rangle$, to the second-to-last bound vibrational level, $|g\rangle$ [see Fig. 1(a) and 1(b)]. Both levels have a rotational quantum number $l = 0$ and a total spin $F = 2$, $m_F = 2$. The level $|g\rangle$ is known from previous experiments [11–14]. It has a binding energy of 637 MHz $\times$ h at 973 G and can be conveniently reached via Raman beams generated with an acousto-optic modulator (AOM). In order to detect the more deeply bound molecules, a second STIRAP pulse converts the
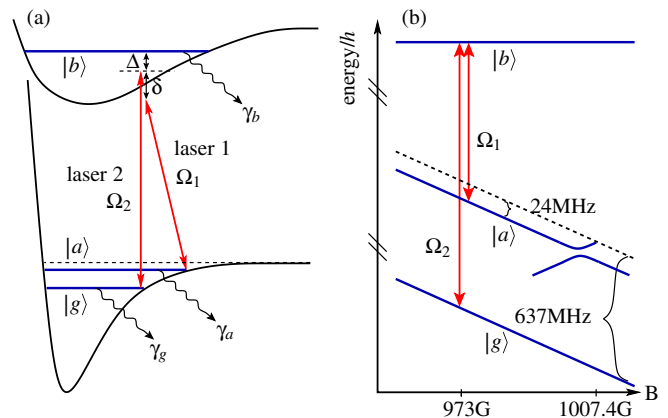

FIG. 1 (color online). (a) Level scheme for STIRAP. Lasers 1, 2 couple the ground state molecular levels $|a\rangle$, $|g\rangle$ to the excited level $|b\rangle$ with Rabi frequencies Ω_1, Ω_2, respectively. Δ and δ denote detunings. γ_a, γ_b, γ_g give effective decay rates of the levels. (b) Zeeman diagram of relevant energy levels. At 1007.4 G a molecular state crosses the threshold of the unbound two atom continuum (dashed line) giving rise to a Feshbach resonance. From there this molecular state adiabatically connects to the last bound vibrational level $|a\rangle$, the state of the Feshbach molecules.

molecules back to the last bound vibrational level, where they are detected as atoms after dissociation via Feshbach ramping. The complete cycle has an efficiency of 75%, indicating a single STIRAP efficiency of 87%.

We use essentially the same setup as in Ref. [15]. Starting point for the experiments is a pure ensemble of 2×10^4 ultracold $^{87}Rb_2$ Feshbach molecules which are held in the lowest Bloch band of a cubic 3D optical lattice. There is no more than a single molecule per site and the whole molecular ensemble occupies a volume of about $20 \times 20 \times 20 \ \mu m^3$. The lattice is 50 E_r deep for molecules ($E_r = 2\pi^2\hbar^2/m\lambda^2$, where m is the mass of the atoms and $\lambda = 830.44$ nm the wavelength of the lattice laser), suppressing tunneling between sites. The molecular ensemble is initially produced from an atomic ^{87}Rb BEC after loading it into the lattice, subsequent Feshbach ramping at 1007.40 G [16] and a final purification step [15] which removes all chemically unbound atoms. Lowering the magnetic field to 973 G transfers the atoms to the adiabatically connected state $|a\rangle$, which has nearly the same magnetic moment as $|g\rangle$ (see Fig. 1). This results in an almost magnetic field insensitive Raman transition [17].

In order to efficiently carry out STIRAP, a suitable excited molecular level, $|b\rangle$, has to be identified (see Fig. 1). We chose the electronically excited molecular state $|0_g^-, \nu = 31, J = 0\rangle$ located 6.87 cm^{-1} below the $S_{1/2} + P_{3/2}$ dissociation asymptote [18]. The corresponding line is strong and solitary; i.e., within a 2 GHz vicinity no other strong molecular lines are found which could interfere with STIRAP. Coupling to other excited molecular states leads to loss of the molecules, since these levels typically decay spontaneously into a variety of undetected vibrational levels in the ground state. Furthermore, it is advantageous that the chosen level $|b\rangle$ has a similar Franck-Condon overlap with states $|a\rangle$ and $|g\rangle$. It can be shown that this also helps to minimize losses through off-resonant coupling channels.

With this choice of states $|a\rangle, |b\rangle, |g\rangle$, we observe a clear molecular dark resonance when coupling the states with resonant Raman laser light (see Fig. 2). The corresponding molecular dark superposition state shows a long lifetime. This is a necessary precondition for our STIRAP experiments, because the molecules have to be kept in a dark state during the whole STIRAP process which in our case typically takes hundreds of μs. The Raman laser beams are both derived from a single Ti:sapphire laser with a short term linewidth of less than 1 MHz. The Ti:sapphire laser is offset locked relative to the D_2 line of atomic rubidium with the help of a scanning optical cavity, which yields an absolute frequency stability of better than 5 MHz. The frequency difference between the two beams is created with an acousto-optical modulator (AOM) with a frequency of about 307 MHz in a double-pass configuration. This allows precise control of the relative frequency difference between the beams over several tens of MHz and ensures phase locking. Both beams propagate collinearly and have a waist of about 290 μm at the location of the
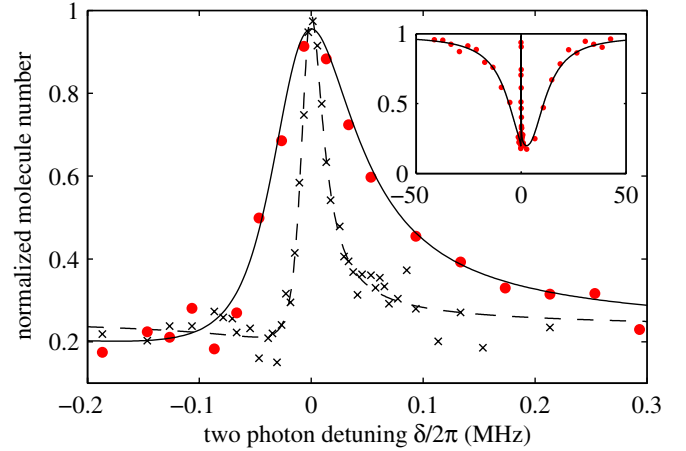

FIG. 2 (color online). Dark resonance. The data show the remaining fraction of Feshbach molecules in state $|a\rangle$, after subjecting them to a 200 μs square pulse of Raman laser light in a narrow range around 0 of the two-photon detuning δ. The inset shows the scan over the whole line of state $|b\rangle$. The strong suppression of loss at $\delta = 0$ is due to the appearance of a dark state. The laser intensities are $I_1 = 2.6$ mW/cm^2, $I_2 = 13$ mW/cm^2 ($\times$), $I_2 = 51$ mW/cm^2 ($\bullet$). Δ is in general tuned close to zero and for the shown measurements happens to be $\Delta/2\pi \approx 2.5$ MHz, which gives rise to the slightly asymmetric line shape of the dark states. The solid and dashed lines are model calculations (see text).

molecular ensemble. The polarization of the beams is parallel to the direction of the magnetic bias field of 973 G.

In order to transfer the molecules from state $|a\rangle$ to state $|g\rangle$, we carry out a STIRAP pulse which consists of a so-called counterintuitive succession of two laser pulses [see Fig. 3(a)]. We first switch on laser 2 and then ramp its intensity to zero within the pulse time $\tau_p = 200 \ \mu$s. Simultaneously we ramp up the intensity of laser 1 from zero to its final value. We fix the ratio of the maximal pulse intensities of laser 1 and 2 to $I_2^{max}/I_1^{max} = 1/3.2$ in order to partially compensate for the unequal Franck-Condon factor of the $|a\rangle - |b\rangle$ and $|g\rangle - |b\rangle$ transitions. Ideally, after the first STIRAP pulse all molecules from state $|a\rangle$ should end up in state $|g\rangle$. In order to determine the population in state $|g\rangle$, we apply, after a holding time of $\tau_h = 5$ ms, a second STIRAP pulse which is the mirror image in time of pulse 1. This transfers the molecules back into state $|a\rangle$. We then ramp the magnetic field over the Feshbach resonance at 1007.4 G which dissociates the molecules with unit efficiency [15] into pairs of atoms. These are subsequently detected with standard absorption imaging. Figure 3(b) shows in a time resolved way how molecules in state $|a\rangle$ first disappear and then reappear during the course of the STIRAP sequence. After applying the first STIRAP pulse, no molecules can be observed in state $|a\rangle$. This is to be expected, since any molecule which is left over in state $|a\rangle$ at the end of the first STIRAP pulse is in a bright state and will be quickly removed by resonantly scattering photons from laser 1. This confirms, that after completion of the
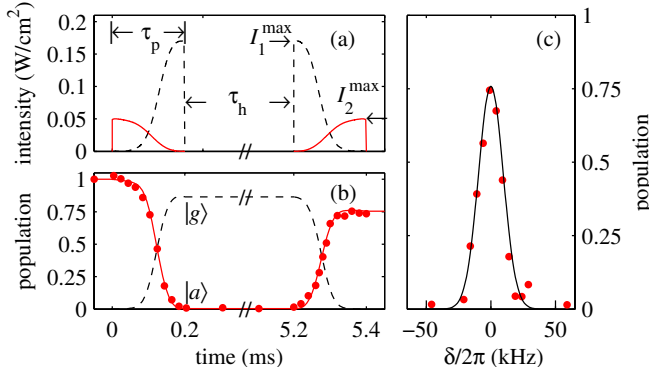
FIG. 3 (color online). STIRAP. (a) Counterintuitive pulse scheme. Shown are laser intensities as a function of time (laser 1: dashed line, laser 2: solid line). The first STIRAP pulse with length $\tau_p = 200$ μs transfers the molecule from state $|a\rangle$ to state $|g\rangle$. After a holding time $\tau_h = 5$ ms, the second pulse (identical, but reversed) transfers the molecules back to $|a\rangle$. $I_{1,2}^{max}$ indicates the maximal intensity of laser 1 (2) in the pulse, respectively. (b) Corresponding population in state $|a\rangle$ (data points, solid line) and state $|g\rangle$ (dashed line). The data points are measurements where at a given point in time the STIRAP lasers are abruptly switched off and the molecule population in state $|a\rangle$ is determined. For these measurements $\Delta \approx 0 \approx \delta$. The lines are model calculations (see text). (c) Efficiency for population transfer from state $|a\rangle$ to state $|g\rangle$ and back via STIRAP as a function of the two-photon detuning δ. The line is a model calculation, showing a Gaussian line shape with a FWHM width of ≈ 22 kHz.

second STIRAP pulse we only detect molecules that were previously in state $|g\rangle$. We observe an efficiency of 75% for the full cycle of conversion into state $|g\rangle$ and back. Figure 3(c) shows how this efficiency depends critically on the two-photon detuning δ.

In Fig. 4 we investigate further the complete STIRAP cycle efficiency as a function of the laser intensity and pulse length. In these measurements we use pulses with the same shape as in Fig. 3(a), which are rescaled to adjust pulse time τ_p and laser intensity. Again, for the best settings we reach an efficiency of about 75% for the two pulses, which corresponds to a transfer efficiency to state $|g\rangle$ of about 87%. The dependence of the efficiency on intensity and pulse length can be qualitatively understood as follows. For short pulse lengths or low intensities, the dark state cannot adiabatically follow the STIRAP pulse, resulting in a low transfer efficiency. For very long pulse lengths and high intensities the losses due to an imperfect dark state become dominant, also resulting in a low transfer efficiency. Thus in order to find an optimum value for the transfer efficiency there is a trade off between adiabaticity and inelastic photon scattering.

We are also able to quantitatively understand our data by using a three level model. It describes the evolution of the quantum mechanical probability amplitudes a, b, and g for a molecule in the respective states $|a\rangle$, $|b\rangle$, and $|g\rangle$ in terms of the following set of differential equations:

$$i\dot{a} = (-i\gamma_a/2)a - \tfrac{1}{2}\Omega_1 b,$$
$$i\dot{b} = [(\Delta + \delta) - i\gamma_b/2]b - \tfrac{1}{2}(\Omega_1 a + \Omega_2 g), \quad (1)$$
$$i\dot{g} = (\delta - i\gamma_g/2)g - \tfrac{1}{2}\Omega_2 b.$$

Here, the Rabi frequencies Ω_1, Ω_2, the detunings Δ and δ, and the decay rates γ_a, γ_b, γ_g are defined as shown in Fig. 1. After experimentally determining Ω_1, Ω_2 and γ_a, γ_g and using $\gamma_b = 2\pi \times 12$ MHz, we are able to consistently describe all data in Figs. 2–4 with a single set of parameters. From one-photon and two-photon scans (as, e.g., in Fig. 2) we obtain $\Omega_1 = 2\pi \times 2.9$ MHz$[I_1/(\mathrm{W\,cm}^{-2})]^{1/2}$ and $\Omega_2 = 2\pi \times 6.0$ MHz$[I_2/(\mathrm{W\,cm}^{-2})]^{1/2}$. The effective decay rates γ_a, γ_g are intensity dependent and are mainly due to the off-resonant coupling of $|a\rangle$ with laser 2 and $|g\rangle$ with laser 1. We determine γ_a (γ_g) by shining laser 2 (laser 1) on the molecules in state $|a\rangle$ ($|g\rangle$) and measuring the off-resonant losses. We find that $\gamma_a/I_2 = 2\pi \times 0.72$ kHz/(W cm^{-2}) and $\gamma_g/I_1 = 2\pi \times 0.40$ kHz/(W cm^{-2}). From these independent measurements, all parameters of Eqs. (1) are determined without further adjustable parameters. In the calculations shown in Fig. 3(b), 3(c), and 4 the time dependent pulse shapes [see Fig. 3(a)] are included. The agreement between theory and experiment is very good.
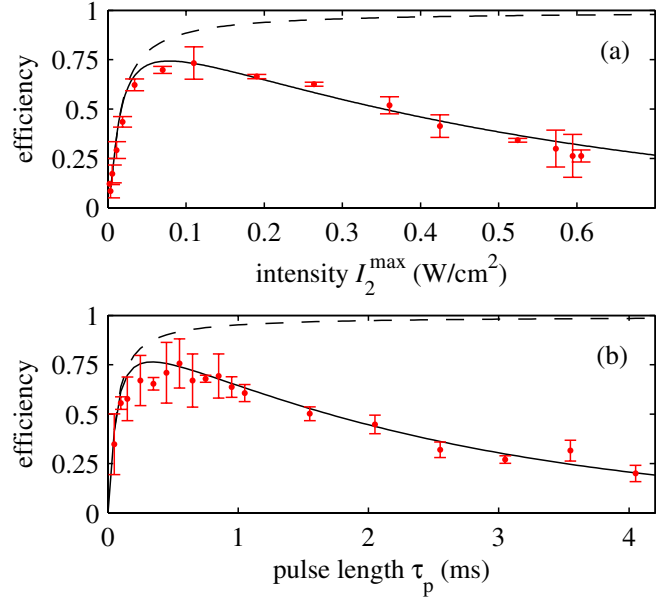

FIG. 4 (color online). Efficiency for population transfer from state $|a\rangle$ to state $|g\rangle$ and back with two STIRAP pulses. (a) Efficiency vs the laser intensity I_2^{max} (fixed pulse length of $\tau_p = 200$ μs). (b) Efficiency vs the pulse length τ_p (fixed laser intensity $I_2^{max} = 44$ mW/cm^2). For (a) and (b) the intensity ratio $I_2^{max}/I_1^{max} = 1/3.2$. The lines are from calculations without free parameters using Eqs. (1). Setting $\gamma_a = \gamma_g = 0$, the efficiency would reach unity for a fully adiabatic transfer (dashed lines). Using for γ_a, γ_a the experimentally determined values, the calculations (solid lines) are in good agreement with the data. The error bars represent a 1σ statistical error.
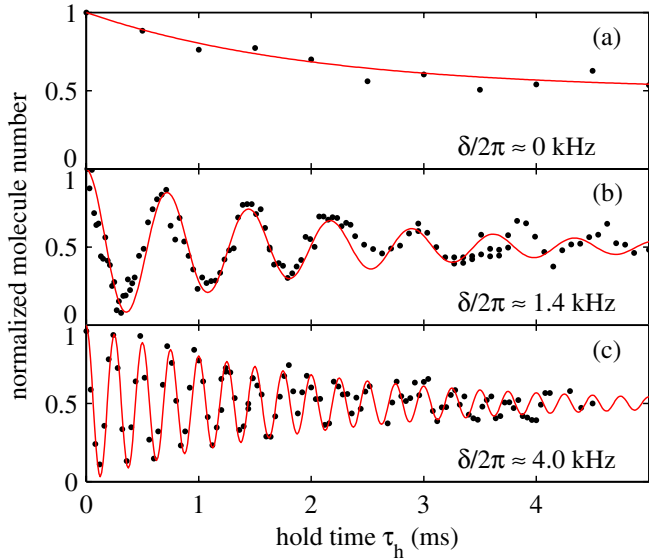
FIG. 5 (color online).   Coherence of the $(|a\rangle - |g\rangle)$ superposition state. Shown is the molecule number in state $|a\rangle$ as a function of holding time τ_h for different detunings δ as indicated. The oscillations indicate coherent flopping of the molecular superposition state between the dark and a bright state. The lines are given by $0.5 \exp(-\tau_h/\tau) \cos(\delta\tau_h) + 0.5$, with a damping time $\tau = 2$ ms.

During STIRAP the molecules are in a quantum superposition, $\Omega_2|a\rangle - \Omega_1|g\rangle$. In order to probe the coherence of this superposition, we perform a Ramsey-type experiment. First we create a dark superposition state with equal population in the two states, $|a\rangle - |g\rangle$, by going halfway into the first STIRAP pulse of Fig. 3(a). We then simultaneously switch off both STIRAP lasers for a variable holding time τ_h, after which we retrace in time the same STIRAP half pulse. As a result we observe oscillations in the number of molecules in level $|a\rangle$ as a function of the holding time τ_h (see Fig. 5). During the holding time, the superposition state freely evolves, coherently flopping between the dark and a bright state with a frequency equal to the two-photon detuning δ. At the end of the holding time, when we switch on again the STIRAP lasers, the dark state is transferred back to state $|a\rangle$ whereas the bright state will be immediately destroyed by the light and leads to complete loss of the corresponding molecules. The observed oscillations are exponentially damped on a time scale of about 2 ms. This damping can be explained by a magnetic field inhomogeneity of about 20 mG over the molecular cloud, which leads to a spatial variation of $2\pi \times 250$ Hz in the two-photon detuning δ. Additionally we observe a slight shift of the oscillation frequency with time on the order of $2\pi \times 50$ Hz/ms. This can be attributed to a small drift of the magnetic field.

Furthermore, we have performed lifetime measurements of the molecules in state $|g\rangle$ by varying the holding time τ_h between the two STIRAP pulses [see Fig. 3(a)]. At a lattice depth of 60 E_r for molecules, we observe a long lifetime of 0.8 s (assuming exponential decay), which is longer than the lifetime of 0.4 s for Feshbach molecules in state $|a\rangle$. At these deep lattices molecular decay is exclusively due to inelastic scattering of lattice photons.

To conclude, using STIRAP we have demonstrated a coherent transfer of a molecular quantum gas from a weakly bound molecular level to a more deeply bound molecular level with a high transfer efficiency of 87%. The method can be extended in a straightforward manner to create arbitrarily deeply bound molecules. With a *single* STIRAP pulse all vibrational levels down to level $X^1\Sigma_g^+(v = 116)/a^3\Sigma_u^+(v = 32)$ should be easily reached since the Franck-Condon factors to state $|b\rangle$ are of similar order as for level $|g\rangle$ [19]. This includes the level $X^1\Sigma_g^+(v = 119)/a^3\Sigma_u^+(v = 35)$ with its binding energy of 30 GHz $\times h$, from which the vibrational ground state $X^1\Sigma_g^+(v = 0)$ of the singlet potential can be reached with two additional Raman (or STIRAP) transitions [8]. Thus STIRAP is a promising tool for the creation of a molecular BEC in the molecular ground state.

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