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 \(^{87}\text{Rb}_2\) 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, which holds them from detrimental collisions, 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.

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Recently, there has been a rapidly growing interest in ultracold molecules due to their 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 \(^{87}\text{Rb}_2\) 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 experiment we transfer the Feshbach molecules with a STIRAP pulse from their last bound vibrational level (binding energy \(24\text{ MHz} \times \hbar\)) to the second-to-last bound vibrational level, \(|g\rangle\) [see Fig. 1(a) and 1(b)]. Both levels have a rotational 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 \(637\text{ MHz} \times \hbar\) at \(973\text{ G}\) 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 \(\Omega_1, \Omega_2\), respectively. \(\Delta\) and \(\delta\) denote detunings. \(\gamma_a, \gamma_b, \gamma_f\) 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 87Rb 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 × 20 × 20 μm^3. The lattice is 50 E_r deep for molecules (E_r = 2π^2ℏ^2/mλ^2, where m is the mass of the atoms and λ = 830.44 nm the wavelength of the lattice laser), suppressing tunneling between sites. The molecular ensemble is initially produced from an atomic 87Rb 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⟩, which has nearly the same magnetic moment as |g⟩ (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⟩, has to be identified (see Fig. 1). We chose the electronically excited molecular state \( |0g⟩, ν = 31, J = 0 \) 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⟩ has a similar Franck-Condon overlap with states |a⟩ and |g⟩. It can be shown that this also helps to minimize losses through off-resonant coupling channels.

With this choice of states |a⟩, |b⟩, |g⟩, 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 \( D_1 \) 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 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⟩ to state |g⟩, 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 \( τ_p = 200 \mu s \). 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 \( I_{2\text{max}}/I_{1\text{max}} = 1/3.2 \) in order to partially compensate for the unequal Franck-Condon factor of the |a⟩ - |b⟩ and |g⟩ - |b⟩ transitions. Ideally, after the first STIRAP pulse all molecules from state |a⟩ should end up in state |g⟩. In order to determine the population in state |g⟩, we apply, after a holding time of \( τ_h = 5 \text{ ms} \), a second STIRAP pulse which is the mirror image in time of pulse 1. This transfers the molecules back into state |a⟩. 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⟩ 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⟩. This is to be expected, since any molecule which is left over in state |a⟩ 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
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 $\delta$.

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 $\tau_p$ 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:

$$i\dot{a} = \left(-\frac{i}{2} \gamma_a / 2\right) a - \frac{i}{2} \Omega_1 b,$$

$$i\dot{b} = \left[(\Delta + \delta) - i\gamma_b / 2\right] b - \frac{i}{2}(\Omega_1 a + \Omega_2 g),$$

$$i\dot{g} = \left(\delta - i\gamma_g / 2\right) g - \frac{i}{2} \Omega_2 b.$$

Here, the Rabi frequencies $\Omega_1$, $\Omega_2$, the detunings $\Delta$ and $\delta$, and the decay rates $\gamma_a$, $\gamma_b$, $\gamma_g$ are defined as shown in Fig. 1. After experimentally determining $\Omega_1$, $\Omega_2$ and $\gamma_a$, $\gamma_g$ 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/$I_1/(W\text{cm}^{-2})^{1/2}$ and $\Omega_2 = 2\pi \times 6.0$ MHz/$I_2/(W\text{cm}^{-2})^{1/2}$. The effective decay rates $\gamma_a$, $\gamma_g$ 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 $\gamma_a$ ($\gamma_g$) 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 $\gamma_g/I_2 = 2\pi \times 0.40$ 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.
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 \(\tau_h\) for different detunings \(\delta\) 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.

![Figure 5](image)

**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 \(\tau_h\) for different detunings \(\delta\) 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.

[17] At 973 G the dependence of the two-photon resonance on the magnetic field is about 12 kHz/G.