Long-Lived Feshbach Molecules in a Three-Dimensional Optical Lattice

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We have created and trapped a pure sample of $^{87}Rb_2$ Feshbach molecules in a three-dimensional optical lattice. Compared to previous experiments without a lattice, we find dramatic improvements such as long lifetimes of up to 700 ms and a near unit efficiency for converting tightly confined atom pairs into molecules. The lattice shields the trapped molecules from collisions and, thus, overcomes the problem of inelastic decay by vibrational quenching. Furthermore, we have developed an advanced purification scheme that removes residual atoms, resulting in a lattice in which individual sites are either empty or filled with a single molecule in the vibrational ground state of the lattice.

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Using magnetic Feshbach resonances [1] to create ultracold diatomic molecules in their highest rovibrational state has become a key to exciting developments and breakthroughs. Feshbach molecules made of bosonic atoms behave in a strikingly different way from Feshbach molecules made of fermionic atoms. For weakly bound dimers of fermionic atoms, vibrational quenching and inelastic decay are strongly suppressed by a Pauli blocking effect in a close encounter of two molecules [2]. This has been vital to the experimental creation of molecular Bose-Einstein condensates (BEC) and investigations of the crossover to a strongly interacting fermionic superfluid [3]. For dimers of bosonic atoms [4–7], however, progress has been hampered by strong inelastic decay due to atom-molecule and molecule-molecule collisions. Therefore, the experiments have been focused on the transient regime, studying, e.g., the collision and dissociation dynamics [6–11].

A three-dimensional optical lattice offers many interesting opportunities for research on ultracold molecules. Lattice sites occupied with exactly two atoms represent a perfectly controlled quantum system which can be rigorously treated theoretically. Matrix elements for atom-molecule coupling are strongly enhanced with the prospect of efficient atom-molecule conversion. Moreover, it is expected that the lattice can isolate molecules from each other and shield them from detrimental collisions so that a long-lived sample can be created also with dimers of bosonic atoms. Recently, first experiments with molecules in a lattice have studied photoassociation [12,13] or demonstrated modifications of the binding energy of tightly confined Feshbach molecules [14].

In this Letter, we report on the creation of a pure sample of ultracold Rb_2 Feshbach molecules trapped in a 3D optical lattice. The observed long lifetimes of up to 700 ms greatly exceed previous values reported for dimers of bosonic atoms [6,7,9,10]. Further, we experimentally investigate association and dissociation of the Feshbach molecules and reach efficiencies of 95% for converting pairs of atoms into molecules. In brief, we adiabatically load a ^{87}Rb BEC into the vibrational ground state of the lattice. For our experimental conditions, about 20% of the condensate atoms are grouped in pairs of two into the lattice sites. By ramping adiabatically over a magnetic Feshbach resonance at 1007.4 G, we convert these pairs into molecules. Another 20% of atoms are located in triply and more highly occupied lattice sites. After the Feshbach ramp, however, inelastic collisions between the created molecules and atoms within the high occupancy sites quickly remove these particles from the lattice. Finally, the remaining 60% of the condensate atoms are found in singly occupied sites and are unaffected by the Feshbach ramp. Using a novel resonant purification scheme, we can remove these atoms from the lattice, which results in a pure molecular sample with each molecule being shielded from the others by the lattice potential.

The starting point for our experiments is an almost pure BEC of about 6×10^5 ^{87}Rb atoms in the spin state $|F = 1, m_F = -1\rangle$ [15]. It is transferred from a quadrupole Ioffe configuration trap (QUIC) into another Ioffe-type magnetic trap with trap frequencies $\omega_{x,y,z} = 2\pi \times (7, 19, 20)$ Hz, leading to a peak density of the BEC of about 4×10^{13} cm^{-3}. Our 3D lattice is cubic and consists of three retroreflected intensity-stabilized laser beams which propagate orthogonally to each other. They are derived from a frequency-stable single-mode Ti:sapphire laser ($\approx$500 kHz linewidth) with a wavelength of $\lambda =$ 830.44 nm. For this wavelength, the laser is detuned by about 100 GHz from the closest transition to an excited molecular level, minimizing light induced losses as a precondition for long molecular lifetimes. The laser beams are polarized perpendicularly to each other, and their frequencies differ by several tens of megahertz to avoid disturbing interference effects. The waists of all three beams are about 160 μm, and the maximum obtainable power is about 110 mW per beam, which results in calculated lattice depths of up to 40 recoil energies ($E_r = h^2/2m\lambda^2$, where m is the atomic mass of ^{87}Rb and h is Planck's constant). We have verified the lattice depths by measuring the energy

gap between bands of the lattice [16]. The relative uncertainty of our lattice depth is $\pm 15\%$.

After the BEC is adiabatically loaded into a $35E_r$ deep 3D optical lattice within 100 ms, we turn off the magnetic trap. By suddenly reversing the bias magnetic field of a few gauss, we flip the spins of our atoms to the high field seeking state $|F = 1, m_F = +1\rangle$ with an efficiency higher than 99% (see also [17]). This state features the Feshbach resonance at 1007.4 G. Afterwards, we ramp up a homogeneous magnetic field in 3 ms to about 1015 G using the QUIC quadrupole coils in Helmholtz configuration. The current through the coils is actively stabilized to a relative accuracy of about 10^{-4}. The fast diabatic crossing of the Feshbach resonance has basically no effect on the atoms in the lattice. If we slowly ramp in 5 ms from 1015 to 1000 G (crossing the Feshbach resonance at 1007 G), molecules are adiabatically produced in the multiply occupied lattice sites. If, however, we cross the Feshbach resonance very quickly, e.g., by simply switching off the magnetic field, less than 10% of the atoms are converted into molecules. Note that, after the first Feshbach ramp, we observe an immediate irretrievable loss of 20% of the atoms. We attribute this loss to inelastic collisions involving molecules for sites initially occupied by 3 or more atoms. The remaining occupied sites each contain either a single atom or a single molecule.

Atom numbers are measured with absorption imaging at low magnetic fields (≈ 2 G) after release from the optical lattice and 11 ms of ballistic expansion. In order to determine molecule numbers, they are first dissociated into atoms by slowly ramping back across the Feshbach resonance and then quickly switching off the magnetic field. We also use absorption imaging to map out the band occupation of the lattice. For this, the lattice is ramped down in 2 ms, and we typically observe a momentum distribution which is fully contained in a cube of width $2\hbar k$ corresponding to the first Brillouin zone of the lattice [18]. This demonstrates that atoms and molecules are in the vibrational ground state of the lattice sites.

In order to create a pure molecular sample, we have developed an advanced purification scheme to remove all atoms which combines the great selectivity of microwave excitation with the high efficiency of atom removal through resonant light pressure [7]. We apply a combined microwave and light pulse at a magnetic field of 1000 G for 3 ms. The microwave drives the transition at a frequency of 9113 MHz between levels which correlate with $|F = 1, m_F = +1\rangle$ and $|F = 2, m_F = +2\rangle$. The light pulse drives the closed transition $|F = 2, m_F = +2\rangle \rightarrow |F = 3, m_F = +3\rangle$. The optical transition frequency is 1402 MHz blue detuned compared to the transition at zero magnetic field. After this pulse, which heats the atoms out of the lattice and an additional hold time (~ 20 ms), no more atoms can be detected. The direct effect of the microwave and light field pulse on the molecules is negligible because the radiation is off resonance. As an indirect effect, however, we find that during the first purification pulse we still lose about 40% of the molecules, probably due to inelastic collisions with the blown away atoms. Further losses are not observed in subsequent purification pulses. We end up with a pure molecular sample formed from about 10% of the initial atoms, which corresponds to 3×10^4 molecules.

We have investigated the lifetimes of the Feshbach molecules in the lattice under various conditions (see Figs. 1 and 2). Figure 1 shows the decay of molecules at a lattice depth of $10E_r$. The pure molecular sample exhibits a remarkably long lifetime of 700 ms. For the case of an unpurified sample, where the atoms at singly occupied sites have not been removed, the lifetime of the molecules is considerably reduced to ≈ 35 ms. This observation suggests that the molecular decay is based on a process where an atom tunnels to a site occupied by a molecule and inelastically collides with it. These inelastic collisions can, in principle, also happen between two molecules. However, compared to an atom, a molecule has a much lower tunneling rate, since it experiences twice the dipole potential and has twice the mass of a single atom. Using simple scaling arguments, for a molecule to have the same tunneling rate as an atom, the lattice light intensity needs to be more than 4 times smaller. This explains the comparatively long lifetime of the purified molecular samples. We note that, if molecular decay is based on inelastic collisions, its time dependence is intrinsically nonexponential. However, exact modeling of the decay would be quite involved and requires precise knowledge of atom or molecule distributions in the lattice. Since these distributions are not known to us, we simply base our estimates for the molecular lifetimes on an exponential decay law.

Figure 2 shows the measured lifetimes of the molecules for various lattice depths. For sufficiently high lattice

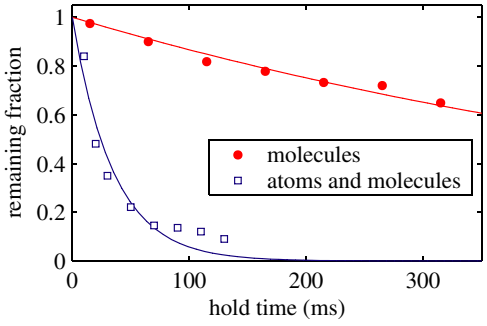

FIG. 1 (color online). Decay of molecules in a 3D optical lattice with a potential depth of $(10 \pm 2)E_r$. Shown is the remaining fraction of molecules in purified (solid circles) and unpurified (squares) samples as a function of hold time. The continuous lines are exponential fits to the data indicating a lifetime of 700 and 35 ms for purified and unpurified molecular samples, respectively. In order to determine molecule numbers in the unpurified sample, purification was performed at the end of the hold time. The hold time was limited to below 400 ms due to the heating up of the coils.

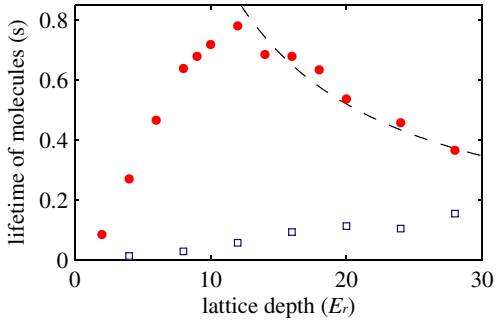

FIG. 2 (color online). Molecular lifetimes for purified molecular samples (circles) and for unpurified samples (squares) as a function of the lattice depth. For this measurement, the lattice depth was reduced from $35E_r$ to the given value after the creation of the molecules. The dashed line is inversely proportional to the lattice depth.

depths, we observe a lifetime for the purified molecular sample inversely proportional to the lattice depth (see dashed line in Fig. 2). From this, we conclude that above a lattice depth of about $12E_r$ the tunneling of the molecules is strongly suppressed, and the lifetime is limited by light induced losses due to off-resonant transitions to an excited molecular state which subsequently decays. Below this value, decay is dominated by tunneling [19] and following inelastic collisions. Thus, the molecular lifetime is maximized in a tradeoff between tunneling and light induced losses. As already shown in Fig. 1, the presence of atoms considerably reduces the lifetime of the molecules, even at larger lattice depths. In the limit of vanishing lattice depths, our experimental lifetimes decrease to values similar to those observed in Refs. [6–10]. Figure 2 clearly demonstrates that shielding of the molecules against inelastic collisions grows with increasing lattice depth.

We now investigate the dynamics for both association and dissociation of a single Feshbach molecule in a lattice site during Feshbach ramping. This fundamental system is of special interest since it can be theoretically treated exactly and solved analytically [21]. We prepare a purified sample of molecules at 1000 G in a lattice of $35E_r$ depth. We then ramp the magnetic field in a symmetric way across the Feshbach resonance up to 1015 G and back (see Fig. 3). Afterwards, purification is applied to remove atoms which have not recombined to form molecules. In a last step, the molecule number is measured. If dissociation and association are not fully adiabatic in a conversion cycle, a loss of molecules will result; e.g., during the association ramp, a pair of atoms might not be converted into a molecule or during dissociation the molecule might break up into two atoms which, after tunneling, are located in separate sites. For slow ramps, we observe small loss signals indicating high adiabaticity for the dissociation-association cycle. In order to increase the loss signal, to improve its accuracy, and to check for consistency, we repeated this experiment with a higher number of cycles (see Fig. 3). The two data

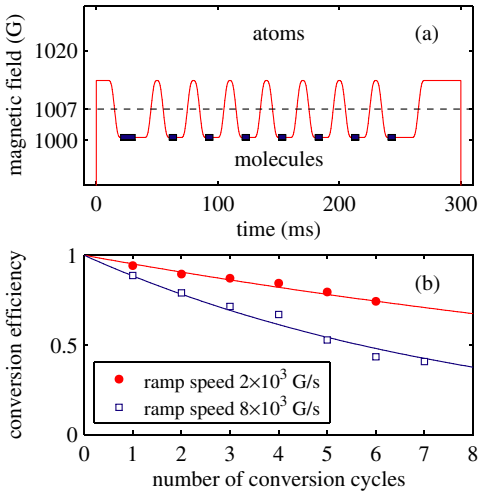

FIG. 3 (color online). (a) Scheme for measurement of conversion efficiency, shown for 7 dissociation-association cycles. The shaded areas indicate the application of our purification procedure to remove atoms. The dashed line at 1007 G shows the position of the Feshbach resonance. (b) Conversion efficiency for a given number of complete dissociation-association cycles for two different ramp speeds of the magnetic field. We measure a conversion efficiency of 95% per cycle for the slow ramp and 89% per cycle for the fast ramp. The solid lines are described by exponential fit curves as described in the text. The lattice depth is $35E_r$.

sets in Fig. 3(b) correspond to two different ramp speeds (2×10^3 G/s, 8×10^3 G/s) and can be described by the exponential functions 0.95^n and 0.89^n, respectively, where n is the number of cycles. Thus, for a slow Feshbach ramp (2×10^3 G/s), we observe an unprecedented high efficiency of up to 95% for the whole dissociation-association cycle. For a faster ramp (8×10^3 G/s), the efficiency drops to 89%. We have taken care that light induced losses have been corrected for in the data [Fig. 3(b)]. Our high conversion efficiencies in the optical lattice are in strong contrast to the low values of $\sim$10% observed previously in a ^{87}Rb BEC [10] which were presumably limited by strong inelastic collisions. In our deep lattice, however, inelastic collisions are suppressed.

After having determined the efficiency for the full dissociation-association cycle, we now study dissociation and association individually. Figure 4(a) shows the measured conversion efficiency of atom pairs to molecules for different ramp speeds. The atom pairs were prepared by creating a pure molecular sample and then dissociating the molecules by slowly (2×10^3 G/s) ramping backward over the Feshbach resonance. Then, again, the magnetic field was swept across the Feshbach resonance at various speeds, and, finally, after switching off completely the magnetic field, the remaining nonconverted atoms are detected. The dashed line in Fig. 4(a) is based on a Landau-Zener expression without adjustable parameters [21] and is given by

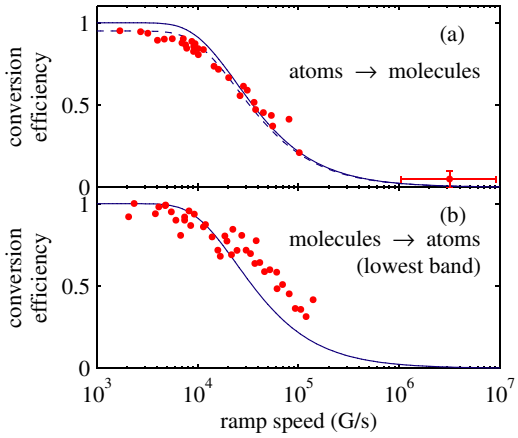

FIG. 4 (color online). (a) Conversion efficiency of atoms (mostly pairs) into molecules as a function of the ramp speed. (b) A purified sample of molecules is dissociated into atom pairs at different ramp speeds. We measure the number of atoms which are observed in the first Brillouin zone of the lattice after release, i.e., atoms that populate the lowest energy band of the lattice. The data is normalized to the atom number at the lowest ramp speeds. The continuous lines in (a) and (b) are calculations as described in the text.

$$p = 1 - \exp\left(-\frac{2\sqrt{6}\hbar}{m a_{\mathrm{ho}}^3}\left|\frac{a_{\mathrm{bg}}\Delta B}{\dot{B}}\right|\right), \qquad (1)$$

where p is the probability of creating a molecule, $a_{\mathrm{bg}} = 100.5a_0$ the background scattering length, $\Delta B = 0.21$ G [10] the width of the Feshbach resonance, $\dot{B}$ the ramp speed at the Feshbach resonance, and $a_{\mathrm{ho}} = \sqrt{\hbar/m\omega}$ the harmonic oscillator length. Using the best estimate for our trapping frequency of $\omega = 2\pi \times (39 \pm 3)$ kHz (corresponding to a lattice depth of $35 \pm 5E_r$), we get good agreement with our data. We note that, even for the slowest ramp speeds, the measured conversion efficiency never reaches unity but levels off at 95%, in agreement with the results in Fig. 3. This, however, does not exclude a true unit conversion efficiency for atom pairs into molecules, because it is possible that 5% of the atoms are not grouped in pairs, e.g., due to nonadiabaticity in dissociation and tunneling. In order to facilitate the comparison of the data distribution and theory, we have scaled the Landau-Zener curve by a factor of 0.95 (dashed line). The maximum controllable ramp speed ($\sim 10^5$ G/s; see Fig. 4) is limited by the performance of our current supply for the magnetic field coils. The data point at 3×10^6 G/s was obtained by simply switching off the coil currents with an external switch. The abrupt switching induces eddy currents, which results in a less controlled ramp with a large error margin. For fast switching, we measured a conversion efficiency of $5 \pm 5\%$.

In Fig. 4(b), we study the dissociation of a purified sample of molecules. We measure the number of atoms which populate the lowest band of the lattice after dissociation. At low ramp speeds, Feshbach molecules get adiabatically converted to pairs of atoms in the lattice ground state. At higher speeds, molecules are energetically lifted above the molecule threshold and can decay into higher lattice bands or into the continuum. Assuming the reversibility of the Landau-Zener transition, we use the same theory curve as in Fig. 4(a). For higher ramp speeds, we measure larger atom numbers than expected. This is probably due to imperfections of our data analysis which can overestimate the atom number in the lowest band by adding in some atoms from higher bands.

To summarize, we have demonstrated that ultracold Feshbach molecules can be created with high conversion efficiency in a 3D optical lattice. After purification, we observe long molecular lifetimes up to 700 ms. These strong improvements over previous experiments open promising perspectives for applications, e.g., in high resolution molecular spectroscopy and quantum information processing in optical lattices. They may also represent an important step in the creation of a stable BEC of molecules in their vibrational ground state.

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