

## State Selective Production of Molecules in Optical Lattices

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We demonstrate quantum control over both internal and external quantum degrees of freedom in a high number of identical “chemical reactions,” carried out in an array of microtraps in a 3D optical lattice. Starting from a Mott insulating phase of an ultracold atomic quantum gas, we use two-photon Raman transitions to create molecules on lattice sites occupied by two atoms. In the atom-molecule conversion process, we can control both the internal rovibronic and external center of mass quantum state of the molecules. The lattice isolates the microscopic chemical reactions from each other, thereby allowing photoassociation spectra without collisional broadening even at high densities of up to  $2 \times 10^{15} \text{ cm}^{-3}$ .

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The formation of ultracold molecules is currently one of the most actively pursued topics in atomic and molecular physics. Recently, spectacular progress in this field has been achieved by using adiabatic sweeps over Feshbach resonances to produce quantum gases of a variety of alkali dimers [1–6] culminating in the observation of Bose-Einstein condensation of molecules [7–9] and resonance superfluidity [10,11]. A complementary approach to the creation of molecules is based upon photoassociation (PA) [12–19]. Here a two-photon Raman process can access molecular states beyond the scope of magnetic Feshbach resonances. It has been pointed out that the combination of photoassociation with 3D optical lattices can considerably enhance the efficiency of the molecule formation process [20] due to the tight confinement of the atoms at lattice sites. Here we report on the production of  $^{87}\text{Rb}_2$  molecules in a 3D optical lattice via one- and two-photon photoassociation. We show that, in addition to the internal quantum state of the molecule, the quantum state of the external center of mass (c.m.) motion can be controlled. Moreover, we demonstrate that two-body loss processes induced by photoassociation can be a sensitive tool in determining the number of atoms on the lattice sites.

Starting from an atomic Bose-Einstein condensate (BEC) in the optical lattice, a Mott insulator state with a central region with two atoms per site can be formed [21]. Thereby, a high number of identical microscopic two-particle systems is created. Through a coherent Raman process, one can then couple exactly two atoms on a single lattice site to a bound molecule in a well-defined rovibronic quantum state via an intermediate excited molecular state, while avoiding perturbing mean field shifts during the photoassociation process. Furthermore, if both atoms initially occupy the lowest internal atomic energy state, the density of the atoms can

be raised to values not achievable in the homogeneous case, due to the absence of any two- and three-body loss processes.

Similar to our previous experiments, we start with the production of an almost pure BEC of approximately  $3 \times 10^5$   $^{87}\text{Rb}$  atoms in the  $|F = 1, m_F = -1\rangle$  state. This condensate can then be transferred into a 3D optical lattice by superimposing far detuned optical standing waves along three orthogonal axes. These standing waves are formed by laser light with a wavelength of  $\lambda = 840 \text{ nm}$  and beam waists ( $1/e^2$ ) of  $w = 150 \mu\text{m}$  at the position of the BEC. We convert the BEC into a Mott insulator by gradually increasing the potential depth of the 3D lattice to a value of up to  $27E_r$  over a time of 80 ms. Here  $E_r = \hbar^2 k^2 / 2m$  denotes the recoil energy,  $m$  the mass of a rubidium atom, and  $k = 2\pi/\lambda$  is the wave vector of the lattice laser light. This results in trapping frequencies at a lattice site of up to  $\omega = 2\pi \times 35 \text{ kHz}$ , with peak densities as high as  $2 \times 10^{15} \text{ cm}^{-3}$  for doubly occupied sites. While for our experimental parameters the outer regions of the Mott insulator should mainly consist of singly occupied sites, the core should be mainly composed of doubly occupied sites.

In order to obtain an accurate detuning relative to the intermediate excited molecular state, we have first performed one-photon photoassociation. This process occurs in an ultracold atomic gas when two colliding atoms absorb a laser photon of energy  $\hbar\omega_a$  corresponding to the energy difference between the asymptotic free atom state and a bound excited molecular level. The coupling strength on this transition  $\Omega_a$  is determined by the electronic Rabi frequency, multiplied by an overlap integral between the free collisional and bound molecular wave function according to the Franck-Condon principle. A deep 3D optical lattice will lead not only to a quasiharmonic confinement for the c.m. motion of the atom pair,

but will also significantly affect the long-range part of the interatomic potential [see Fig. 1(a)]. As a result, the initial collisional wave function is localized in space and thereby the Franck-Condon factor of the photoassociation transition can be considerably enhanced. In fact, the confinement of atoms on lattice sites turns the free-bound transition into a bound-bound transition.

In our experiment, molecule formation is detected by monitoring atom losses. This is done by measuring the remaining atom number with standard absorption imaging after the atoms have been illuminated for a fixed period of time with a PA laser beam. From the reduction in atom number, we can deduce the number of created molecules, under the assumption that secondary loss processes can be neglected. In order to stabilize the frequency of the photoassociation laser, the laser is offset locked relative to the  $D1$  line of atomic rubidium. It is tuned with an accuracy of 1 MHz to the vicinity of a particular excited molecular level of the  $0_g^-$  potential, approximately 0.69 THz below the atomic resonance frequency [18]. This laser beam exhibits a total power of 3 mW and is focused to a  $(1/e^2)$  beam waist of 200  $\mu\text{m}$  at the position of the quantum gas in the optical lattice. After locating the position of the one-photon molecular resonance, we have measured the time dependence of the molecule formation process by placing the PA laser on resonance with the molecular transition and recording the atom losses vs time. Assuming only two-body losses, the atomic loss rates can be described by

$$\frac{dN}{dt} = -K_2 \int n^2(\vec{r}) d^3r, \quad (1)$$

where  $N$  is the total atom number after a time  $t$ ,  $n(\vec{r})$  is the atomic density, and  $K_2$  is a characteristic loss constant.

This equation displays an interesting dimensionality effect. For a 1D optical lattice, most lattice sites contain  $N \gg 2$  atoms. Therefore, many molecules can succes-

sively be formed on one site. As more and more atoms are converted into molecules, the remaining atom number decreases, causing a change in the atomic density  $n(\vec{r})$ . This leads to a nonexponential decay in the atom number, which we observe in the experiment (see Fig. 2). If we impose a deep 3D optical lattice, such that we reach the Mott insulator regime, the lattice sites are occupied by single atoms or atom pairs for our experimental parameters. As the lattice isolates atoms on different sites from each other, only atoms on doubly occupied lattice sites can be converted into molecules. Figure 2 shows a decay of the remaining atom number asymptotically tending to a constant offset. The decay is related to photoassociation on doubly occupied lattice sites and its form is purely exponential since the rate is independent of the total atom number. The remaining atom number decreases as

$$N(t) = N_1 + 2N_2 \exp(-t/\tau), \quad (2)$$

where  $N_{1,2}$  is the initial number of lattice sites occupied by one or two atoms, respectively. From  $N_1$  and  $N(0)$  we can conclude that for our lattice parameters the ratio of single to doubly occupied lattice sites is 1.6(1), which is consistent with a shell structure of Mott insulator regions with single and double occupancy.

Having determined the resonance frequency corresponding to the excited molecular level, we can now drive a resonantly enhanced two-photon Raman transition (see Fig. 1). In this process, two colliding atoms absorb one photon from the first laser field  $\omega_a$  and emit another photon into an additional stimulating laser field  $\omega_b$ . The second laser is phase locked with respect to the first in order to enable the coherent two-photon transition. Under the action of the two-photon coupling, both atoms will be transferred to a molecule in a specific molecular state. The rovibronic state is selected by choosing a Raman-laser difference frequency  $\delta = \omega_a - \omega_b$ , which

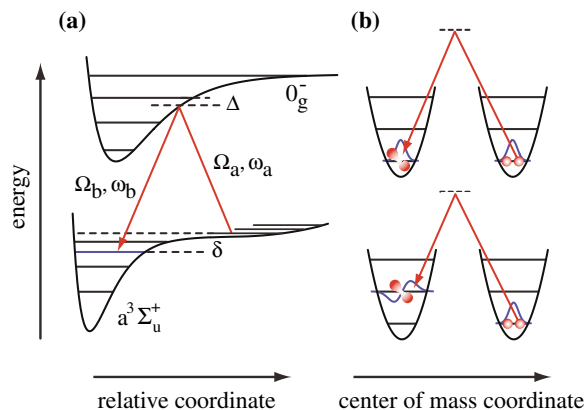


FIG. 1 (color online). Two-color photoassociation process of two atoms being placed in the external ground state at a single site of an optical lattice potential. Both the rovibrational quantum state in the relative motion of the two atoms forming the molecule (a) and the c.m. motional quantum state of the molecule (b) can be controlled.

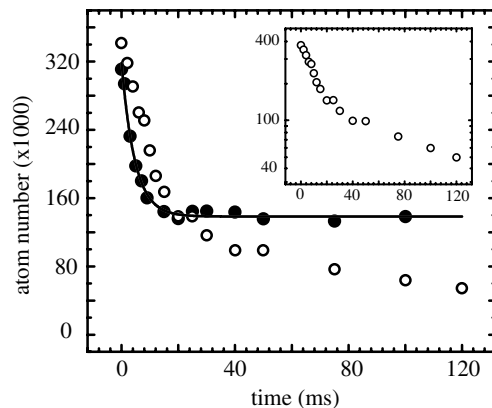


FIG. 2. Time-resolved one-color photoassociation; in 1D (empty circles) and 3D lattice (filled circles) configurations. The photoassociation laser is tuned to resonance with an intensity of  $I = 5 \text{ W/cm}^2$  and illuminates the sample for a varying time, after which the total atom number is measured. The inset shows the 1D lattice loss data in a log-lin plot.

matches the binding energy of the molecular state. In our case, a parallel polarization ensures  $\Delta J = 0$  transitions, where  $J$  is the rotational quantum number. As the molecules are formed in  $s$ -wave collisions, we can therefore selectively produce molecules in the rotational ground state. The final level is chosen to lie in the  $a^3\Sigma_u^+$  electronic state potential. If the virtual intermediate level of energy  $\hbar\omega_a$  is relatively close to a real molecular level, as in the case of our experiment, the effective two-photon Rabi frequency of the process is given by  $\Omega_{\text{eff}} = \Omega_a\Omega_b/(2\Delta)$ , where  $\Omega_{a,b}$  are the respective one-photon Rabi frequencies and  $\Delta$  is the detuning from the intermediate level. The one-photon Rabi frequencies include the Franck-Condon factors for the corresponding transitions. Therefore, the external confinement by the optical lattice also increases the efficiency of the Raman process through  $\Omega_a$ .

A major obstacle lies in the possibility of spontaneous Raman scattering. A molecule that has just been formed can absorb a single photon from either laser beam and be excited to an intermediate molecular state, from where it can decay via spontaneous emission to other bound or unbound states, leading to trap losses. In order to ameliorate these problems, we choose a rather large detuning  $|\Delta|$ , which decreases the spontaneous scattering rate. Thus far, however, we have not been able to enter a regime, where the coherent coupling exceeds the damping rate, such that, e.g., coherent Rabi oscillations between atoms and molecules could be observed [1,22].

We present measurements at difference frequencies of  $\delta \approx 2\pi \times 24$  MHz and  $\delta \approx 2\pi \times 636$  MHz corresponding to the uppermost vibrational levels in the ground state potential [18]. Both lasers illuminate the atomic ensemble for a fixed time in a counterpropagating beam configuration parallel to one axis of the optical lattice. Thereafter, the remaining number of trapped atoms is again recorded (see Fig. 3). On all spectra, we observe a progression of resonances spaced by roughly 30 kHz. This series of resonances corresponds to a resolved quantized c.m. motion of the molecules formed at single lattice sites. In fact, when the lifetime of the molecules is longer than the c.m. oscillation period at each lattice site, it should be possible to resolve the different c.m. motional states, which is the case for our experimental parameters. The highest resonance frequency recorded denotes a transition in which a molecule in the ground state of a single lattice site has been formed. The other resonances originate from transitions to higher lying motional quantum states [see Fig. 1(b)]. The appearance of these resonances confirms that the emerging molecules are trapped on the sites of the optical lattice. The frequency separation of the resonances is directly given by the vibrational splitting of the c.m. motional quantum states. It is interesting to note that the observed trapping frequencies almost coincide with the atomic trapping frequencies. This can be understood as the polarizability of molecules formed in high

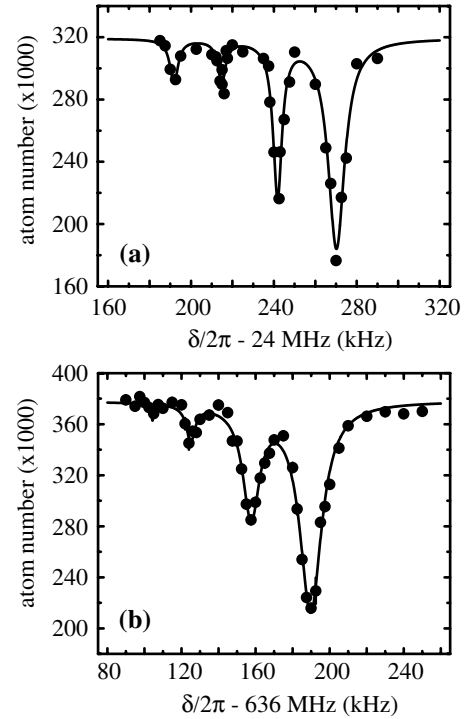


FIG. 3. Two-photon photoassociation of the uppermost vibrational level in the electronic ground state  $^3\Sigma_u^+$  at  $\delta/2\pi \approx 24$  MHz and at  $\delta/2\pi \approx 636$  MHz, with the quantized c.m. motion resolved. The remaining atom number after a photoassociation time of about 120 ms is measured for varying difference frequencies  $\delta$  at a constant detuning of (a)  $\Delta = 2\pi \times 700$  MHz and laser intensities of  $I_a = 12$  W/cm<sup>2</sup> and  $I_b = 6$  W/cm<sup>2</sup> and (b) a detuning of  $\Delta = -2\pi \times 990$  MHz and  $I_a = 16$  W/cm<sup>2</sup> and  $I_b = 12$  W/cm<sup>2</sup>. The solid curve is a fit assuming four independent Lorentzian profiles and a constant background. The difference in the vibrational level spacings between (a) and (b) are mainly due to different lattice depths of (a)  $18E_r$  and (b)  $27E_r$  along the optical lattice axis of the photoassociation lasers.

lying rovibrational states should be close to twice the polarizability of a single atom [23]. Since the molecules have twice the mass of a single atom, this leads to identical trapping frequencies as in the case of free atoms. Note, however, that the external potential for the molecules is twice as deep as for the atoms and supports more bound states. Furthermore, we observe that the spacing between higher lying vibrational c.m. quantum states is slightly reduced due to the anharmonicity of the trapping potential at a lattice site.

The reduced strength of the transitions to higher lying c.m. motional quantum states is related to an additional Franck-Condon factor in the c.m. motion, which depends on the initial and final motional quantum states  $|\tilde{v}\rangle$  and  $|\tilde{v}'\rangle$  and is given by  $\langle \tilde{v}' | \exp(2ikz) | \tilde{v} \rangle|^2$ . The total transition rate to form molecules is then directly proportional to the internal rovibronic and external c.m. Franck-Condon factors. In Fig. 3, we observe a good quantitative agreement with the expected weaker transitions to higher lying motional quantum states to within 10% accuracy.

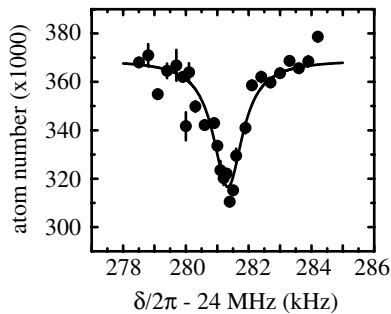


FIG. 4. High resolution two-photon photoassociation of the transition to the most weakly bound internal vibrational state in the  $^3\Sigma_u^+$  potential and the ground state of the c.m. motion. The scan was taken at an intermediate detuning of  $\Delta = -2\pi \times 900$  MHz. The solid line is a Lorentzian fit to the data with a width of  $\gamma = 1.1(2)$  kHz.

Spontaneous Raman scattering mostly limits the observed linewidths and, by reducing the intensities of the corresponding laser, we have been able to observe narrow two-photon resonance linewidths as low as 1 kHz (see Fig. 4). Note that this high resolution spectroscopy is carried out at high atomic on-site densities of  $2 \times 10^{15} \text{ cm}^{-3}$ . This proves that mean field shifts are absent in our system and do not broaden the lines, as has been observed in [18] even for an order of magnitude lower densities. Limits on the linewidth observed here could be caused by a differential light shift between the atoms and the molecules, a resonant spectral background on our diode lasers, or mechanical vibrations. From the observed lowest linewidths, we can deduce the lifetime of the molecules, which should be on the order of 1 ms.

In conclusion, we have demonstrated two-photon Raman photoassociation of  $^{87}\text{Rb}$  atoms from a Mott insulator phase in an 3D optical lattice. By choosing appropriate parameters for the photoassociation laser fields, we have achieved control over both internal and external degrees of freedom of the molecules. A combination of Feshbach techniques and two-color Raman transitions could further increase the efficiency of molecule formation while allowing one to address deeply bound molecular states in future experiments [24]. A counterintuitive photoassociation pulse sequence leading to stimulated Raman adiabatic passage (STIRAP, [25]) could result in a very fast and robust molecule production scheme. In combination with a 3D optical lattice, one would then end up with a molecular Mott insulator. The melting of this Mott insulator [20] could then be used to create a Bose-Einstein condensate of molecules in almost arbitrary internal quantum states.

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