Dynamically probing ultracold lattice gases via Rydberg molecules

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Abstract

We show that the excitation of long-range Rydberg molecules in a three-dimensional optical lattice can be used as a position- and time-sensitive probe for doubly occupied sites in the system. To this end, we detect the ions which are continuously generated by the decay of the formed Rydberg molecules. While a superfluid gas shows molecule formation for all parameters, a Mott insulator with \( n = 1 \) filling reveals a strong suppression of the number of formed molecules. In the limit of weak probing, the technique can be used to probe the superfluid to Mott-insulator transition in real-time. Our method can be extended to higher fillings and has various applications for the real-time diagnosis and manipulation of ultracold lattice gases.

The quantum phase transition from a superfluid to a Mott insulator in ultracold atomic lattice gases [1–3] highlights the large potential of atomic quantum gases for the study of many-body quantum systems. One of the most prominent signatures of the Mott insulating phase is the suppression of atom number fluctuations. Measuring the site occupancy has been a key instrument to diagnose the properties of these systems. The employed techniques include spin changing collisions [4, 5], imaging the Mott shells via microwave clock states [6] or direct in situ fluorescence imaging [7, 8]. In all these approaches, the measurement requires the destruction of the whole atomic sample. Here, we introduce a new technique based on the formation of long-range Rydberg molecules in an optical lattice. In the limit of weak probing, the technique allows for a real-time detection of the double occupancy.

Rydberg atoms feature prominent properties such as a giant polarizability, which allows them to strongly interact with other excited atoms over distances of several micrometers [9]. This leads to the so-called Rydberg blockade [10], which prevents the excitation of more than one atom in a micrometer-sized volume [11–13]. Another peculiar feature is that the electron in a Rydberg state has a contact interaction with ground state atoms immersed in the electron’s wave function. The interaction can be described by a Fermi-type pseudo-potential [14]. For an attractive interaction, this leads to the formation of long-range Rydberg molecules with binding energies in the order of MHz [15, 16]. They have first been demonstrated with Rydberg S-states [15, 17] and later on for Rydberg D-states [18]. The molecular binding mechanism is not restricted to one single ground state atom. Also two or more atoms can be caught by the electron. Even a whole atomic sample can be located inside the Rydberg wave function for high principal quantum numbers \( n > 100 \) [19], leading to a mean field shift of the atomic transition. For an attractive interaction the transition frequency is lowered compared to the single atom transition. Thus, for small values of \( n \) well defined molecular states can be excited, while for higher values of \( n \) the molecular binding energy gets smaller and eventually results in a many body broadening and red shift of the single atom transition [20]. In order to create a long-range Rydberg molecule, at least two ground state atoms have to be closer to each other than the extension of the electronic wave function of the Rydberg electron, which is in the order of 100–1000 nm for main principal quantum numbers between \( n = 30 \) to \( n = 100 \). In an optical lattice configuration in the tight binding limit, the parameters can be chosen such that only two atoms at the same lattice site can form a molecule. The formation of molecules is then proportional to the probability to find two atoms at the same site. This can be used as a diagnostic tool to check for doubly occupied lattice sites.

In the experiment, we use an ultracold gas of rubidium 87 atoms in a crossed optical dipole trap with equal trapping frequencies \( \omega = 2 \pi \times 65 \text{ s}^{-1} \) in all three directions [21]. The atoms can additionally be loaded in a...
superimposed three-dimensional optical lattice with a wavelength of $\lambda = 742$ nm. Two axes are realized in a retro reflected configuration, while in the third axis—due to geometrical contraints—two orthogonal laser beams are used. This leads to lattice constants of $d_1 = d_2 = 371$ nm and $d_3 = 524$ nm. We excite the atoms to the $30P_{3/2}$ state using a UV laser at 297 nm. The UV light is produced by a frequency doubled dye laser locked to the $3^2P_1$ state of rubidium. The measurement was done with a Bose–Einstein condensate in the $|F = 1, m_F = 1\rangle$ state at a residual magnetic field of approximately 1 G and with a 20 ms UV pulse of a power of 3 $\mu$W. The signal is normalized to the ion yield on resonance. Throughout this work, the measurements were taken on the molecular peak around $-55$ MHz, indicated by the dashed line. The red solid line is a guide to the eye.

Figure 1. Spectrum of Rydberg molecules for the excitation to the $30P_{3/2}$ state in $^{87}$Rb. The measurement was done with a Bose–Einstein condensate in the $|F = 1, m_F = 1\rangle$ state at a residual magnetic field of approximately 1 G and with a 20 ms UV pulse of a power of 3 $\mu$W. The signal is normalized to the ion yield on resonance. Throughout this work, the measurements were taken on the molecular peak around $-55$ MHz, indicated by the dashed line. The red solid line is a guide to the eye.

The sensitivity to doubly occupied sites can be directly proven by comparing the molecular spectrum of a BEC with that of a Mott insulator with a filling of $n = 1$ in the trap center. The latter is realized by adiabatically ramping up the three-dimensional optical lattice with a lattice depth of $20E_{\text{rec}}$ in all three directions. In figure 2 we compare the two spectra for a small system, where the $n = 2$ Mott shell does not build up. To distinguish the produced atomic ions from molecular ions we have measured the spectrum in a time of $20$ ms. The spectrum shows several molecular peaks red-detuned from the main resonance. The peak at a detuning of about $-55$ MHz from resonance is chosen for the experiments presented in this work. A scaling analysis of this peak with varying atom number reveals that this peak corresponds to a diatomic Rydberg molecule. The large detuning from resonance ensures that the off-resonant excitation of single atoms is highly suppressed. The extension of a Rydberg molecule is given by the diameter of the electronic wave function. For $n = 30$ this amounts to $d_{\text{Ry}} = 146$ nm, which is significantly smaller than the lattice spacing. As the molecule formation requires a contact interaction between the Rydberg electron and a ground state atom, molecule formation can only take place on the same lattice site. The Wannier function in the tight binding limit is of similar magnitude: the standard deviations $\sigma$ of the Gaussian onsite density distribution for a lattice depth of $20E_{\text{rec}}$ are $\sigma_1 = 60$ nm and $\sigma_2 = 85$ nm for the different lattice axis.

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The method can now be used to measure the critical atom number $N_{\text{crit}}$ for the transition to the $n = 2$ Mott shell. To this end, we vary the atom number of the sample before we ramp into the Mott insulator and excite the Rydberg molecules. In figure 3 we compare the number of excited Rydberg molecules for a BEC and a Mott insulator (lattice depth $20E_{\text{rec}}$) in dependency of the atom number $N$. Since the excitation of a Rydberg molecule is a two-body process, the ion production rate in a BEC, $\Gamma_{\text{BEC}}$, scales with the volume integrated squared density, $\int n(r)^2d^3r \propto n(0)^2R_{\text{TF}}^3$. Here, $R_{\text{TF}}$ is the Thomas–Fermi radius of the condensate. The central density and the
Thomas–Fermi radius of a BEC with parabolic confinement scale as $n(0) \propto N^{5/7}$ and $R_{TF} \propto N^{1/7}$ such that we find $\Gamma_{BEC} \propto N^{1/5}$. From a power law fit to the data we extract an exponent $b = 1.58 \pm 0.24$, compatible with the predicted value of 1.4. In the case of a Mott insulator the ion production rate is proportional to the number of doubly occupied lattice sites. For an atom number which does not allow for an $n = 2$ shell to build up, no molecules should be formed (compare figure 2(b)). Once the atom number exceeds a critical atom number $N_{crit}$, the $n = 2$ shell starts to grow. From an analysis of the shell sizes of a Mott insulator in the atomic limit for zero temperature, one can show that the number of doubly occupied sites $Z_2$ is connected to the total number of atoms as $N = (N_{crit}^{2/3} + Z_2^{1/3})^{3/2} + Z_2$. The number of molecules is then proportional to $Z_2$. A fit to the data (see figure 3) with this function shows good agreement and reveals a critical atom number of $(14 \pm 3) \times 10^3$, above which the $n = 2$ shell grows. An estimate of the critical atom number in the atomic limit for $T = 0$ yields $N_{crit} = (27 \pm 5) \times 10^3$. We attribute the difference to the finite temperature and the finite tunneling coupling, which both lead to the formation of doubly occupied sites for smaller total atom numbers [25]. Below the critical atom number, we still find a small number of excited molecules, which scale linear with the atom number (fitted exponent $b = 1.0 \pm 0.1$). The linear dependence can be understood as a consequence of the linearly growing

Figure 2. Time of flight spectra for a BEC and a Mott insulator for varying detuning in a logarithmic scaling (the dark blue color represents rates of 500 Hz and below). The atom number was adjusted such that the $n = 2$ shell in the Mott insulator is suppressed. The generated ions are detected after a time of flight of $t = 70 \mu s$ in the case of atomic ions (left dashed line), while molecular ions have a $\sqrt{2}$ longer time of flight (right dashed line). The data was averaged over 1000 pulses of $1 \mu s$ length each.

Figure 3. Log–log plot of the number of excited molecules in dependence of the total atom number $N$ for a BEC (black points) and a Mott insulator (red points). The sample was excited with 250 consecutive pulses of $1 \mu s$ duration and a laser power of $1 \text{ mW}$. The lines are fits as explained in the text. The dashed red line indicates the critical atom number $N_{crit}$ obtained from the fit. The number of excited molecules is deduced from the number of detected ions with help of a conversion factor $f = 16 \pm 4$. This factor is obtained from atom loss experiments (compare figure 4).
volume of the \( n = 1 \) Mott insulator. The small number of doubly occupied sites originates most likely from thermal excitations of the Mott insulator.\(^{[25]}\) The data show that the technique is capable to detect very small numbers of molecules.

When a molecule is excited in the lattice, several inelastic processes occur. Besides the decay into a molecular ion, photoionization and spontaneous decay are also possible. In most cases, it is unlikely that after the decay both atoms are again in the ground state of the lattice site. The excitation of a molecule is therefore connected with a loss of at least one of the atoms. This gives a handle to the spatially resolved measurement of doubly occupied sites in the lattice. With a scanning electron microscope\(^{[26, 27]}\) that is implemented in our setup, it is possible to image this change of density. This is done by making an image of the Mott insulator with and without a UV excitation. Figures 4\((a)\) and \((b)\) show both density profiles and the difference of both signals is shown in Figure 4\((c)\). The density in the center is clearly reduced compared to the wings. As losses are only expected to appear in the \( n = 2 \) shell, the radius of the loss area should indicate the transition to the \( n = 1 \) shell. In the experiment we find a radius of \( 6 \pm 1 \mu \text{m} \). The estimated radius of \( 7.5 \mu \text{m} \) is indeed close to this value.

An important feature of the excitation to molecular Rydberg states in an optical lattice compared to other techniques, is that the ion signal is continuously generated. If the power of the excitation laser is reduced, the sample is only slightly disturbed and a continuous measurement of the double occupancy becomes possible, even when the lattice parameters are changed. To demonstrate this, we ramp the optical lattice to \( 20E_{\text{rec}} \) within 150 ms, while continuously exciting Rydberg molecules at a very small laser power of \( P = 1.5 \mu \text{W} \).

The time-resolved signal is shown in figure 5, where the time axis has been converted to the actual lattice depth. We compare the dynamic measurement with a static measurement where the system was adiabatically...
prepared for a given lattice depth and probed for $\tau = 20$ ms. The measurement was performed for an atom number below and above $N_{\text{crit}}$. In both cases, the shape of the signal is the same (note that we have to normalize the two signals as the limited longtime stability of the UV laser (about 1–2 MHz) is larger than the linewidth of the molecular resonance and the excitation efficiency varies between the measurements). The measurements show that for weak enough probing, the dynamics of doublon formation can be probed in real time. The temporal resolution of the technique is only limited by the lifetime of the excited molecules which is in the order of 20 $\mu$s. This is much faster than any intrinsic timescale of a typical lattice gas and does not impose any restrictions. The sensitivity of the technique is given by the conversion factor $f = 16 \pm 4$ of excited (and subsequently lost) molecules to produced ions. In the experiments presented in figure 5, less than 10% of the atoms have been removed during the dynamic measurement.

We have demonstrated that the excitation of Rydberg molecules in an optical lattice is a valuable tool to study the double occupancy in static and dynamic configurations. Due to the imprinted losses, the approach is position sensitive, provided it is combined with a spatially resolved in situ imaging method. Various intriguing applications of this approach are feasible. First, the technique is applicable to any bosonic or fermionic species, where long-range Rydberg molecules can be excited. Also mixtures of different hyperfine states or even different atomic species can be probed. Second, for high principal quantum numbers, where the electronic wave function reaches to the neighboring lattice site, nearest neighbor density correlations could be probed. Eventually, the existence of Rydberg molecules including more than one ground state atom allows the probing of multiply occupied lattice sites.

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