## **Probing Number Squeezing of Ultracold Atoms across the Superfluid-Mott Insulator Transition**

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The evolution of on-site number fluctuations of ultracold atoms in optical lattices is experimentally investigated by monitoring the suppression of spin-changing collisions across the superfluid-Mott insulator transition. For low atom numbers, corresponding to an average filling factor close to unity, large on-site number fluctuations are necessary for spin-changing collisions to occur. The continuous suppression of spin-changing collisions is thus direct evidence for the emergence of number-squeezed states. In the Mott insulator regime, we find that spin-changing collisions are suppressed until a threshold atom number, consistent with the number where a Mott plateau with doubly occupied sites is expected to form.

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One of the most fundamental signatures of the Mott insulator (MI) transition undergone by ultracold atomic gases in optical lattices [1–13] is a drastic change in atom number statistics. In a very shallow lattice, ultracold bosons tend to form a Bose-Einstein condensate. In this case, a measurement of the probability for finding *n* atoms at a given lattice site would reveal a characteristic Poisson distribution with large on-site fluctuations. However, for deeper lattices, the influence of repulsive interactions, which disfavor such fluctuations, becomes increasingly dominant and results in the emergence of numbersqueezed states with suppressed number fluctuations. Above a critical lattice depth, the ultracold gas enters the MI regime, where the number fluctuations almost vanish. In experiments so far, interaction-induced numbersqueezed states were detected through the observation of increased phase fluctuations, the canonically conjugate variable to number fluctuations  $[1-3]$ , or through an increased time scale for phase diffusion [4].

In this Letter, we directly observe the continuous suppression of number fluctuations when the ultracold sample evolves from the superfluid (SF) regime to deep in the MI regime. The idea behind our measurement is illustrated in Fig. 1. After producing an ultracold gas in an optical lattice, we suddenly increase the lattice intensity, suppressing tunneling and freezing the number distribution. A probe sensitive only to the presence of *atom pairs* at a given lattice site is finally applied. Close to unity filling, a nonzero probe signal is obtained only if initially large onsite fluctuations produce a nonzero fraction of sites with two atoms. While we observe this behavior for a gas initially in the SF regime, the probe signal is progressively suppressed when approaching the Mott transition, indicating increasingly number-squeezed states.

The specific two-particle probe used in this work are spin-changing collisions (see [14], and references therein), which convert at each lattice site pairs of spin  $f = 1$  atoms in the  $m = 0$  Zeeman sublevel to pairs with one atom in  $m = +1$  and the other in  $m = -1$ . In principle, other

schemes, e.g., measuring the interaction energy [11] or monitoring atom losses due to Raman photoassociation [15,16] or Feshbach resonances [17], could be suitable for this measurement. Spin-changing collisions have advantages when used as a probe for atom pairs: they are nondestructive (see also [18]), and they can be resonantly controlled using the differential shift between Zeeman sublevels induced by an off-resonant microwave field [19–21]. We show that this technique allows one to measure selectively doubly occupied sites in the optical lattice.

Our experimental setup has been described in detail in [14]. We first load a degenerate gas of  $87Rb$  atoms in the  $|F = 1, m = -1$  Zeeman sublevel into a combined magnetic trap plus optical lattice potential at an initial lattice depth  $V_0$ . The intensities are then rapidly increased from  $V_0$  to  $V_f = 40E_r$  within  $t_{up} = 1$  ms (see Fig. 2). Here  $E_r =$  $h^2/2M\lambda^2$  is the single photon recoil energy, and  $\lambda =$ 842 nm the lattice laser wavelength. Immediately after this ramp, the magnetic potential is switched off, and the cloud is held for 60 ms in order to let the magnetic bias



FIG. 1 (color). Illustration of the number statistics measurement. Spin-changing collisions turn atom pairs initially in the Zeeman substate  $m = 0$  (no arrow) to pairs in  $m = \pm 1$  states (up and down arrows). This process happens for sites with  $n = 2$  (a) or  $n = 3$  (b) atoms. For one atom per site on average, whether this occurs depends drastically on the many-body correlations. For a Bose-Einstein condensate (c), large on-site fluctuations create a finite number of sites with 2 or 3 atoms, where  $\pm 1$  pairs can be created. On the contrary, for a MI state (d), only isolated atoms are found and no  $m = \pm 1$  pairs are created.





FIG. 2. Time sequence of the experiment.

field stabilize to its final value  $B \approx 1.2$  G. The atoms are then prepared in the  $m = 0$  state using microwave transfer pulses, and held for a variable time *t*osc, during which a collisional spin oscillation takes place. This coherent evolution is detected experimentally as a reversible exchange between the populations in the  $m = 0$  and  $m = \pm 1$ Zeeman sublevels, measured by absorption imaging after 12 ms of free expansion.

Spin-changing interactions in deep optical lattices have been described in detail in [14]. We consider spin  $f = 1$ atoms and assume that tunneling can be neglected, so that the lattice sites are isolated from each other. At a single lattice site, the spin-changing collisions are critically sensitive on the filling *n* of the well. For sites with filling  $n =$ 0 or 1, spin-changing collisions cannot occur. The first nontrivial case corresponds to doubly occupied wells. In this case, only two spin states (one with both atoms in  $m =$ 0 and the other with a single  $m = \pm 1$  pair) are accessible [see Fig. 1(a)]. Therefore, the atom pair undergoes Rabilike oscillations at the effective Rabi frequency  $\frac{1}{2}$  $\delta_2^2+\Omega_2^2$  $\tilde{\epsilon}$ . The energy mismatch (''detuning'') between the two states is  $\hbar \delta_2 = \Delta \epsilon + U_s$ , where  $\Delta \epsilon = \epsilon_{+1} + \epsilon_{-1} - 2\epsilon_0$  corresponds to the difference in Zeeman energies  $\epsilon_m$ . The spindependent interaction energy *Us* depends on atomic and lattice parameters [14], and also determines the coupling ratuce parameters [14], and also determines the coupling<br>strength as  $\Omega_2 = 2\sqrt{2}U_s$ . Sites with  $n = 3$  atoms behave in a similar way [Fig. 1(a)], however, with an energy difference  $\hbar \delta_3 = \Delta \epsilon - U_s$  and a coupling strength  $\Omega_3 =$  $2\sqrt{6}U_s$ .

In principle, site occupancies  $n > 3$ , whose spin dynamics involve more than one  $m = \pm 1$  pair, are also possible. However, during the hold time  $t_{hold}$  indicated in Fig. 2, those sites can be emptied by three-body (3B) recombination events at an event rate  $\gamma_n = \gamma_{3B}n(n-1)(n-2)$  [22], with  $\gamma_{3B} \approx 0.5 \text{ s}^{-1}$  for our parameters. Therefore, sites with  $n \geq 4$  are efficiently removed after the wait time.

In our experiment, we produce large ensembles of atoms in the optical lattice with spatially inhomogeneous atom number distribution. The inhomogeneity results from an additional trapping potential *V*ext present on top of the optical lattice [23]. In the MI regime, this potential leads to the formation of flat Mott plateaus with a well-defined atom number per site [5,7,24]. Also, the local fluctuations have an inhomogeneous distribution. Experimentally, we measure the ''spin-oscillation amplitude'' for the entire atomic cloud, i.e., the global population  $A_{\text{osc}} = (N_{+1} +$  $N_{-1}/N$  of the  $m = \pm 1$  states after an evolution time  $t_{\text{osc}}$ , normalized to the total atom number *N*. This amplitude is related to the probability  $\overline{P}_n$  of finding *n* atoms per lattice site, averaged over the cloud spatial profile.

Let us suppose that we are able to tune the singleparticle detuning to  $\Delta \epsilon = -U_s$ , such that doubly occupied sites are exactly on resonance. Then, neglecting sites with  $n \geq 4$ , the oscillation amplitude is obtained by summing the contribution from sites with  $n = 2$  and  $n = 3$ ,

$$
\mathcal{A}_{\text{osc}} \approx \bar{\mathcal{P}}_2 \sin^2 \left(\frac{\Omega_2 t_{\text{osc}}}{2}\right) + \frac{6}{7} \bar{\mathcal{P}}_3 \sin^2 \left(\sqrt{\frac{7}{8}} \Omega_2 t_{\text{osc}}\right). \tag{1}
$$

From Eq. (1), we conclude that atom pairs and triplets oscillate essentially out of phase. By choosing  $\Delta \epsilon$  =  $-U_s$  and  $t_\pi = \pi/\Omega_2$ , all doubly occupied sites are converted to  $m = \pm 1$  pairs, whereas the conversion efficiency for triplets is around 3%. Recording the amplitude of the spin oscillations thus allow one to probe the distribution of atom pairs alone. This is reminiscent of cavity quantum electrodynamics [25,26], where Fock states of the cavity field could be discriminated due to different coupling strengths to an atomic transition. In particular, choosing  $\Delta \epsilon = U_s$  would allow one to measure the fraction of triply occupied sites remaining after three-body decay.

To achieve full conversion of doubly occupied sites, it is necessary to tune the spin oscillations for doubly occupied sites into resonance, i.e., set  $\Delta \epsilon = -U_s$ . In a magnetic field *B*, the quadratic Zeeman shift contributes a *positive* amount to  $\Delta \epsilon$ . Hence, if  $U_s > 0$  (which is the case for  $87Rb$ ), the interaction energy  $U_s$  leads to a residual detuning in zero magnetic field that prevents reaching the resonance. For this reason, we introduce a different technique using the differential level shift induced on the individual Zeeman sublevels by a far off-resonant microwave field (''ac-Zeeman shift''). With a suitable choice of polarization, detuning, and power, the detuning  $\Delta \epsilon$  can be tuned at will in the range of interest, and allows one to compensate the magnetic field contribution to  $\Delta \epsilon$  plus the interaction term  $U_s$ . In this work, the microwave field is detuned by several hundred MHz to the red of any hyperfine resonance to suppress population transfer to  $f = 2$ . Indeed, no such transfer is observed within our experimental sensitivity.

In Fig. 3, the fraction of atoms found in  $m = \pm 1$  is plotted as a function of the microwave power for a fixed  $t_{\rm osc} = 15.5 \text{ ms} \approx t_{\pi}$ , corresponding to maximum conversion. These data were taken for constant initial lattice depth  $(V_0 = V_f = 40E_r)$  and atom number ( $N \approx 2.6 \times 10^5$ ). For very low microwave powers, the spin dynamics is suppressed by the quadratic Zeeman detuning ( $\Delta \epsilon \approx 2\pi \times$ 207 Hz), much larger than the spin-dependent interaction  $U_s \approx 2\pi \times 10.7$  Hz. The ac-Zeeman shift can compensate



FIG. 3. Resonance curve of the spin-oscillation amplitude in the far-detuned microwave field, measured at a magnetic field  $B \approx 1.2$  G and at a fixed hold time  $t_{hold} = 15.5$  ms. Solid (open) circles denote the population in the Zeeman substate  $m = 0$  $(m = \pm 1).$ 

for this detuning and for the interaction part, inducing a resonance in the number of  $m = \pm 1$  pairs shown in Fig. 3. The oscillation amplitude, close to the expected  $\mathcal{A}_{\text{osc}} \approx$ 0*:*5, indicates that nearly all atom pairs are converted into  $\pm$ 1 pairs, in agreement with further experiments discussed in a companion paper [21].

We now turn to the measurement of number statistics. We choose  $t_{\rm osc} \approx 15.5$  ms and a dressing field tuned to resonance, as in the previous paragraph. At a given lattice depth  $V_0$ , we have recorded the oscillation amplitude in a broad range of atom numbers, from about  $10^4$  to a few  $10^5$ [27]. The experiment is then repeated for various lattice depths, from the SF regime ( $V_0 = 4E_r$ ) to deep in the MI regime ( $V_0 = 20E_r$  and  $40E_r$ ). As shown in Fig. 4, at low lattice depths, the spin oscillations occur for any atom number *N*, with an amplitude slowly increasing with *N*. For small atom number, the oscillation amplitude is increasingly suppressed with increasing lattice depth, and completely vanishes for large lattice depths. This qualitative behavior is consistent with the behavior expected from the Bose-Hubbard model [5–13]. On approaching the Mott transition, the ground state adapts to an increased interaction energy by reducing its number fluctuations, eventually producing an array of one-atom Fock states at each site where spin-changing collisions cannot occur.

Within the MI regime [Figs.  $4(d) - 4(f)$ ], we observe that the suppression of spin oscillations persists up to some threshold atom number  $[6.0(3) \times 10^4$  for the data in Fig. 4(f)]. This is consistent with the expected formation of Mott plateaus with increasing atom number, as the cloud expands in the trapping potential. A Mott plateau with *n* atoms per site forms when the cloud radius reaches the size  $R_n$  where the potential energy  $V_{ext}(R_n)$  matches the on-site interaction energy  $U(n - 1)$ . For a harmonic potential with trapping frequency  $\omega_{\text{ext}}$ , this happens at a threshold number [29]  $N_n \approx N_2 \sum_{k=1}^n k^{3/2}$ . Above  $N_2 \approx$  $4\pi/3$  $\left(\frac{m\omega_{\text{ext}}^2d^2}{2U}\right)^{-3/2}$ , a core with two atoms per site starts to grow, thus enabling the spin oscillations. For the parameters that correspond to Fig.  $4(f)$  ( $\omega_{ext}$ )  $2\pi \times 80$  Hz and  $V_0 = 40E_r$ ), we calculate  $N_{\text{th}} \sim 6.8 \times$ 104, close to the measured value. For even higher atom number (corresponding to  $N_3 \sim 3 \times 10^5$ ), a shell of triply occupied sites start to form, reducing the fraction of atoms in the  $n = 2$  shell. This can be seen in Fig. 4(f), where we indeed observe a decrease of the spin amplitude above this number.

In order to compare our experimental results with the prediction of the Bose-Hubbard model [5], we solve this



FIG. 4 (color). Amplitude of the spin oscillation vs atom number and different lattice depths:  $V_0 = 4, 8, 11, 13, 20, 40E_r$  (a)–(f). The thin dashed lines show the prediction of a theoretical model, where the atom number distribution is deduced from a mean-field approach at  $T = 0$  (see text). The thin solid lines show only the fraction of atom pairs calculated from the same model. The thick solid

model numerically within a mean-field approximation at zero temperature [30,31]. Accounting for losses during the hold time *t*hold (wait time plus oscillation time), we obtain the distribution  $P_n$ . For each filling *n* and a given  $t_{\text{osc}}$ , we calculate the conversion efficiency  $\eta_{\pm 1}(n)$  to  $m = \pm 1$  pairs, and obtain the total spin amplitude from  $A_{\text{osc}} = \sum_{n} n n_{\pm1}(n) \bar{P}_n$ . The results of this calculation, indicated by the thin dashed lines in Fig. 4, lie very close to the fraction of pairs  $\bar{P}_2$  predicted by the same model (thin solid lines), in agreement with the arguments leading to Eq.  $(1)$ . Deep in the MI regime [Figs.  $4(e)$  and  $4(f)$ ], the calculations agree well with the measurements. For lower lattice depths, although the qualitative trend is still reproduced, we find discrepancies. Near the Mott transition [Fig. 4(d)], the mean-field calculations predict an amplitude lower than observed, a behavior consistent with the study of number correlations beyond mean-field reported in [12]. Below the transition point [Figs.  $4(a)-4(c)$ ], the model predicts an oscillation amplitude higher than observed. Deviations from the initial distribution may arise in this low lattice depth regime if excitations are generated during the preparation phase and result in an increased populations in the low-density regions of the cloud, which barely participate to the spin oscillations. Such ''finite temperature'' effects have possibly less influence in the MI regime, where the many-body system is protected by an interaction gap.

In conclusion, we have shown how spin oscillations can be used to probe number squeezing in optical lattices via the detection of the fraction of atom pairs. Our observations confirm the expected scenario: near-Poissonian fluctuations for shallow lattices, strongly suppressed fluctuations for deep lattices, and a smooth interpolation in between. Moreover, the observed behavior is consistent with the expected formation of Mott plateaus, a signature of the incompressibility of this system. Our results indicate that number squeezing is robust with respect to experimental manipulations, such as transfer to the purely optical trap. In this sense, they are promising to employ those number-squeezed states, e.g., in Heisenberg-limited atom interferometry [32].

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