Quantum dynamics of a mobile spin impurity

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One of the elementary processes in quantum magnetism is the propagation of spin excitations. Here we study the quantum dynamics of a deterministically created spin-impurity atom, as it propagates in a one-dimensional lattice system. We probe the spatial probability distribution of the impurity at different times using single-site-resolved imaging of bosonic atoms in an optical lattice. In the Mott-insulating regime, the quantum-coherent propagation of a magnetic excitation in the Heisenberg model can be observed using a post-selection technique. Extending the study to the superfluid regime of the bath, we quantitatively determine how the bath affects the motion of the impurity, showing evidence of polaronic behaviour. The experimental data agree with theoretical predictions, allowing us to determine the effect of temperature on the impurity motion. Our results provide a new approach to studying quantum magnetism, mobile impurities in quantum fluids and polarons in lattice systems.

Deepening our knowledge of quantum magnetism1 is one of the main goals in quantum simulation. In particular, one highly desired aim is a better understanding of high-\(T_c\) cuprate superconductors, which are believed to be described by Heisenberg-type effective spin models in the limit of low doping2. More generally, the physics characterized by the Heisenberg model governs the properties of many strongly correlated materials,6, and allows the realization of various types of spin order ranging from spin solids to spin liquids1,4. In low-dimensional quantum magnets, rich physics emerges owing to the dynamics of spin excitations5. A basic mechanism of quantum magnetism in strongly correlated electronic systems is superexchange, in which, for example, opposite spins on adjacent lattice sites coherently exchange their positions. Ultracold atoms in optical lattices offer an ideal testbed to explore these phenomena in a controlled experimental environment6, as shown by the observation of superexchange in double-well systems8 or plaquettes9. Furthermore, the recently demonstrated single-atom-resolved detection10,11 opens entirely new prospects for the quantum simulation of strongly correlated spin systems. For example, this technique has enabled the simulation of one-dimensional anti-ferromagnetic Ising spin chains through mapping of the site occupation onto a pseudo-spin12. In ultracold-atom experiments, however, observation of superexchange-based quantum magnetism in many-body systems has so far been hindered by the fact that typical temperatures are much larger than the superexchange coupling energy.

In this work, we report on space- and time-resolved observation of a coherently propagating spin wave, by tracking the motion of a deterministically created single-spin impurity in a one-dimensional (1D) spin chain (Fig.1). Specifically, we studied the quantum dynamics of a mobile boson of type \(\uparrow\) on a 1D lattice, surrounded by a bath of bosons of type \(\downarrow\). Such a system can be described within a two-species single-band Bose–Hubbard model, parametrized by the spin-independent single-particle tunnelling rate \(J\) and on-site interaction energy \(U\) (Supplementary Information). Deep in the Mott-insulator regime (\(U \gg J\)) with unity filling, this model can be mapped to the isotropic Heisenberg model1–14:

\[
\hat{H} = -J_{\text{ex}} \sum_{\langle j,k \rangle} \hat{S}_j \cdot \hat{S}_k \quad (1a)
\]

\[
= -\frac{J_{\text{ex}}}{2} \sum_{\langle j,k \rangle} (\hat{S}_j^+ \hat{S}_k^- + \hat{S}_j^- \hat{S}_k^+) - J_{\text{ex}} \sum_{\langle j,k \rangle} \hat{S}_j^z \hat{S}_k^z \quad (1b)
\]

in which the effective superexchange coupling \(J_{\text{ex}} = 4J^2/U\) arises from a second-order tunnelling process. The bosons \(\uparrow\) and \(\downarrow\) are identified with spin states \(|\uparrow\rangle\) and \(|\downarrow\rangle\) and the corresponding operators of the effective spin system are \(\hat{S}_j = (\hat{S}_j^x, \hat{S}_j^y, \hat{S}_j^z), \hat{S}_j^+ = \hat{S}_j^x + i\hat{S}_j^y, \hat{S}_j^- = \hat{S}_j^x - i\hat{S}_j^y, \hat{S}_j^z = |\uparrow\rangle\langle\uparrow| + |\downarrow\rangle\langle\downarrow|\) and \(\hat{S}_j^z = (\hat{n}_j + \hat{n}_j^+)/2\), with the number operators \(\hat{n}_j\) for bosons of type \(\sigma = \uparrow, \downarrow\) on lattice site \(j\). In the case of a single flipped spin in a ferromagnetic domain, the second term of equation (1b), which describes the longitudinal spin coupling, gives rise only to an energy offset and can therefore be neglected. The remaining first term of equation (1b) is structurally equivalent to the single-species single-particle tunnelling Hamiltonian within the tight-binding model of a 1D lattice system15,17, with the tunnelling rate \(J\) being replaced by \(J_{\text{ex}}/2\), and the atomic creation and annihilation operators being replaced by the spin-flip operators \(\hat{S}_j^\uparrow\) and \(\hat{S}_j^\downarrow\).

For larger \(J/U\), the mapping to the Heisenberg model breaks down owing to the fluctuations of the site occupancies and owing to higher-order processes, and no analytic solution describing the time evolution of the impurity exists. In this regime, the motion of the impurity is modified compared with both a free particle and an isolated magnetic excitation. This effect can be interpreted as a result of interactions between the impurity and low-lying density excitations, which resemble phonons, leading to polaron-like effects.

Experimental procedure

To create 1D spin chains in the optical lattice, we followed the experimental procedure of our previous work18. We started

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by preparing a 2D degenerate gas of about 170 $^{87}$Rb atoms in a single antinode of a vertical optical lattice (lattice spacing $a_{\text{lat}} = 532$ nm) along the $z$ direction. We then switched on two horizontal optical lattice beams within 120 ms with potential depths $V_x = 10.0(3)E_r$ and $V_y = 30(2)E_r$ (the number in parentheses marks the uncertainty of the last digit), where $E_r = \hbar^2/(8ma_{\text{lat}}^2)$ denotes the recoil energy and $m$ is the atomic mass of $^{87}$Rb. This creates an array of parallel 1D Mott insulators along the $x$ direction, each containing 8 to 16 atoms. The atoms were initially prepared in the hyperfine state $|F = 1, m_F = -1\rangle$ ($\uparrow\uparrow$). We introduced the spin impurity by changing the hyperfine state of an atom at the centre of the chain to state $|F = 2, m_F = -2\rangle$ ($\downarrow\downarrow$) using single-site addressing\textsuperscript{19}. In this scheme, a $\sigma^+$-polarized, off-resonant laser beam at 787.65 nm wavelength focused onto the selected lattice site results in a negative energy shift (attractive potential) only for the state $\downarrow$ while leaving the initial spin state $\uparrow$ almost unaffected. A microwave pulse, resonant with the shifted atomic transition, then produced the spin-flip from $\uparrow\uparrow$ to $\downarrow\downarrow$. In contrast to our previous work\textsuperscript{19}, a spatial light modulator generated an addressing beam profile in the form of a line instead of a radially symmetric Gaussian beam, to create the impurity in all 1D chains simultaneously (Methods). This multiple-site addressing technique offers the advantage to prepare an arbitrary spin pattern in the lattice more rapidly, which minimizes heating effects during the preparation. In addition, the line-shaped beam allowed us to hold the impurities in all 1D systems, and to release them simultaneously by switching off the beam within 1 ms. We then allowed the impurity in each chain to propagate during a variable hold time, and finally froze the dynamics by rapidly increasing the lattice depth of all axes to $>80 E_r$ within 300 ms.

The resulting spin distribution was detected by single-site-resolved fluorescence imaging using a high-resolution microscope objective\textsuperscript{11}. We used two alternative detection methods to determine the position of the impurity: the direct imaging of the impurity spin component $\uparrow\uparrow$ after removing all atoms in state $\uparrow\uparrow$ with a resonant laser pulse (positive image) and detecting it as an empty site in the bath of $\uparrow$ atoms after removing the spin impurity (negative image). The latter has the advantage to provide information about the thermal excitations (holons and doublons) of the system as well, which are also detected as empty sites. Although we cannot distinguish between spin impurity and thermal excitations, post-selecting samples with only one empty site in the chain enables us to filter out a lower-temperature subset of the data. If the only empty site arises from a spin-flipped atom, the position of the impurity can be determined exactly and the selected samples contain no other excitations.

**Evolution of the impurity in the Mott-insulator regime**

We first studied the time evolution of a spin impurity deep in the Mott-insulator regime using negative images. The probability distribution of its position was obtained by averaging data from different chains, post-selected to contain only one empty site within the central 10–14 sites. The distributions (see Fig. 2 for $J/U = 0.053(7)$) show clear maxima and minima, resulting from the quantum interference due to the coherent evolution of the spin impurity. Owing to the weaker superexchange coupling, the observed dynamics occurs on timescales much longer than the tunnelling time ($\hbar/4 = 4$ ms) that would characterize the motion of non-interacting atoms. We compared our data with the time evolution of the spin impurity in the exactly solvable homogeneous Heisenberg model at zero temperature. In this model, the probability of finding the impurity at time $t$ on site $j$ after starting its evolution at $t = 0$ from the centre of the chain ($j = 0$) is

$$P_j(t) = \left[ J_j \left( \frac{\hbar}{2} \right) \right]^2$$

where $J_j$ is the Bessel function of the first kind\textsuperscript{20}. A single fit to all distributions observed at different hold times (red curves in Fig. 2) with the superexchange coupling as a free parameter yields good agreement with the data for $J_{\text{he}}/\hbar = 65(1)$ Hz, which is close to $J_{\text{he}}/\hbar = 51(4)$ Hz obtained from an $ab$ initio band-structure calculation using the independently measured lattice depths (see also Fig. 4). Owing to the finite addressing fidelity it is possible that no spin impurity was created in a single run of the experiment, but a single thermal excitation is wrongly identified as a spin impurity. This results in small differences between the experimental data and the model (see Supplementary Information for more details). We note that the effect of the external potential on the spin dynamics can be neglected in the Mott-insulator limit, as long as the impurity has not yet reached the edge of the system (Supplementary Information).

**Temperature effects**

To examine the effects of temperature, we measured the same distribution using positive imaging (Fig. 3a), in which we directly observed the impurity position in the finite-temperature bath.
Figure 2 | Dynamics of a mobile spin impurity. a,b. Top: fluorescence images of the atoms (left) taken with the negative imaging technique (see main text) together with the reconstructed atom distribution in the central region (right). The 1D systems are oriented horizontally. The red vertical stripe denotes the initial position of the spin impurity (detected as an empty lattice site). For the generation of an effective low-temperature subset, only the samples containing exactly one empty site were kept (green tick marks), whereas those containing more than one empty site were discarded (red crosses). a–d. The histograms show the position distribution of the spin impurity after different hold times for $J/U = 0.053$. Each histogram is obtained from an average over 200–250 1D systems. The error bars denote the 1σ statistical uncertainty. The red line is a simultaneous fit to all distributions with the analytic solution of the Heisenberg model of equation (2), yielding $\Delta_0 = 65(1)$ Hz.

Compared with the distribution from the post-selected negative images with one empty site (Fig. 3b), it has almost the same width, but less contrast. This indicates that the propagation velocity of the spin impurity is not much affected by temperature, although its coherent evolution is hindered. Our experimental observation is quantitatively confirmed by numerical simulations (Fig. 3c,d) using the time-dependent density-matrix renormalization group (t-DMRG) algorithm, including temperature (Methods). Our intuitive interpretation of this effect is that thermal excitations can move back and forth across the spin impurity, introducing phase slips that wash out the coherence of the distribution. However, a single excitation alters the position of the spin impurity at most by one site in a 1D system and as a consequence the width of the distribution is less affected. For a quantitative analysis, we evaluated the contrast and the propagation speed from the simulated distributions for different temperatures of $T = 0 - 0.2 \, U/k_B$ (Fig. 3d). We defined the contrast as $C = (P_{\text{max}} - P_{\text{min}})/(P_{\text{max}} + P_{\text{min}})$, where $P_{\text{max}}$ is the peak value at positions other than the centre, and $P_{\text{min}}$ denotes the minimum in the centre region between the peaks (that is, $C = 0$ if there is no other maximum except for the central peak). The contrast obtained from the positive image equals that of the simulated distribution at $T = 0.15 \, U/k_B$ (blue point in Fig. 3d). It is consistent with the system’s temperature of $T = 0.14(3) \, U/k_B$, determined independently by comparing the distribution of thermal excitations to an analytical model in the atomic limit11. The contrast from negative images corresponds to $T = 0.11 \, U/k_B$ (green point in Fig. 3d), demonstrating that the post-selection enables us to extract a lower-temperature subset of the data. The actual temperature of this subset is, in fact, even lower, because our simulation does not take into account the finite spin-flip fidelity of 88(5)% and the fact that the addressed site could be empty owing to a thermal excitation (see Supplementary Information). We also determined the propagation speed of the spin impurity as a function of temperature by fitting the simulated distribution with the function of equation (2), rewritten as $P_j(t) = [U_j/\langle a^\dagger a \rangle] F_j$, where we introduced $\tau$ as the speed of the impurity. We found that the change in velocity within the simulated temperature range is only 4% (Fig. 3d). This result is consistent with our observation of similar propagation speeds from positive and negative images (see also Fig. 4d) and the intuitive argument mentioned above.

Propagation velocity of the impurity

Deep in the Mott-insulator regime, the impurity propagates in an environment that is defect-free except for thermal excitations, leading to the coherent quantum motion described in the previous section. The situation is different in the superfluid regime, in which the bath of $\uparrow$ spins has quantum fluctuations of the on-site density and contains many low-energy excitations. In one dimension, those excitations are known to be phonon-like21, which lead to a Fröhlich-type Hamiltonian now describing the modification of the impurity motion in the bath by the propagation of a polaron17,22.
To investigate this regime, we measured the local impurity density at different times for increasing values of $J/U$. As the impurity is always prepared inside a Mott-insulating state at $J/U = 0.053$, we decreased the lattice depth $V_z$ along the chain within 50 ms to reach the desired final value of $J/U$. While preparing the bath in this way, we kept the impurity spin pinned by the addressing beam before releasing it. For large final $J/U$, close to the superfluid–Mott-insulator critical point, post-selecting samples with only one empty site is no longer effective for tracking the spin because of the proliferation of particle–hole pairs induced by quantum fluctuations\cite{footnote23}. We therefore recorded the dynamics at large $J/U$ using only positive images, in which we directly detected the impurity spins (Fig. 4a–c).

As there is no full analytical solution describing the dynamics of the spin impurity for intermediate values of $J/U$, we chose to analyse the experimental data by fitting the width of the position distribution with the function $P_1(t)$ with the velocity as an adjustable parameter. This function is indeed the analytic solution both for the Heisenberg (Mott insulator) regime ($J/U \ll 1$) with $v = I_{lat}a_{lat}/\hbar = 4(J/U) \cdot (I_{lat}/\hbar)$ and for a non-interacting impurity ($J/U = \infty$) with $v = 2I_{lat}/\hbar$. We found that a fit with $P_1(t)$ captures well the edges of the position distribution, allowing us to determine the maximum propagation velocity for all $J/U$ values used in this study. We employed the same fit to determine the propagation velocity from the numerical simulations. These took into account the statistical atom number fluctuations from the experiment as well as the trapping potential, which are both important in the superfluid regime.

Over the whole range of $J/U$ accessed in the experiments, the measurement shows a good agreement with the simulation, both for the position distribution (Fig. 4a–c) and for the impurity velocity (Fig. 4d). For small $J/U$, the experimentally determined spreading velocity normalized to the tunnelling rate $f$ increases as $v/f = 4J/U$, as expected from the superexchange coupling $I_{lat}/\hbar = 4J^2/U$. The spreading velocity becomes smaller than the velocity $v/f = 4J/U$ close to the point where the superfluid–Mott-insulator transition would occur in a homogeneous system in the thermodynamic limit.

We interpret this change of velocity as a result from the interaction of the impurity with the increasingly large number of low-energy excitations of the bath, which now affect not only the contrast but the velocity as well. In the superfluid regime, experiments and numerical simulations show that the normalized velocity is only $v/f \approx a_{lat}/\hbar$; that is, it is only about half the velocity expected for free-particle tunnelling. This indicates that the system’s strong interactions in the superfluid regime still slow down the motion of the impurity significantly. Such a behaviour is consistent with analytical calculations\cite{footnote23} and can be interpreted as a mass increase of the impurity expected from polaronic physics\cite{footnote24,footnote25,footnote26,footnote27,footnote28,footnote29,footnote30}.

**Indications of polaronic physics**

In a simple picture, a polaron can be seen as an impurity propagating coherently together with a dip in the bath density created by the repulsive interaction between the impurity and bath atoms\cite{footnote24} (Fig. 5a,b, inset). To confirm this interpretation of our experimental results, we investigated the time evolution of the bath. We first measured the spatial distribution of the bath at $J/U = 0.47(5)$ before releasing the impurity from the addressing beam. The measured distribution was the parity distribution $P_{lat} = \langle \mod_{2}(\hat{N}_i) \rangle$ because our imaging process records the parity projection of the on-site atom number\cite{footnote31}. The distribution shows a clear dip at the centre site where the impurity is located (Fig. 5a). As a reference, we observed the distribution of the bath without the microwave pulse that introduces the impurity spin, with the sequence being otherwise identical. The comparison shows that the dip is due to the localized impurity and its interaction with the bath and that it is independent of the presence of the pinning beam on the central site. The difference between the bath distribution can be seen only at the centre site. This indicates that the healing length of the bath at the centre of our system is of the order of or smaller than the lattice spacing.

The same experiment was performed after switching off the addressing beam within 1 ms and after a subsequent evolution time.
of 0.5 ms (Fig. 5b). The difference in the bath distributions with and without the impurity is still visible. It matches the probability distribution of the impurity after the same evolution time (Fig. 5c), which shows that the impurity leaves an imprint in the bath density, thus supporting the picture of polaronic dynamics. Moreover, both impurity and bath distributions agree with our DMRG simulations (Fig. 5a–c). Measurements for different evolution times yielded similar results (see Supplementary Information).

To illustrate how correlations between the impurity and bath atoms develop around the impurity as it starts to propagate, we calculated the time evolution of the correlator $C_{j,k}(j) = \sum_k \mathcal{N}_{j,k}(k, k+j)$ from the simulations (Fig. 5d). Here $\mathcal{N}_{j,k}(j,k) = \langle \hat{n}_{j,k} \hat{n}_{j,k} \rangle - \langle \hat{n}_{j,k} \rangle \langle \hat{n}_{j,k} \rangle$ is the density–density correlator between the impurity and the bath, and $j,k$ denote the lattice sites. As the impurity and the bath deformation propagate together, we expect an anti-correlation of $\uparrow$ and $\downarrow$ atoms ($\mathcal{N}_{j,k}(j,k) < 0$ around the impurity position). Before releasing the impurity, no correlation exists ($t = -1$ ms in Fig. 5d), because the impurity is pinned at the centre site and its distribution is independent of the bath distribution. As the impurity starts moving, it continues to displace bath atoms owing to the repulsive interaction, resulting in the build-up of an anti-correlation around the impurity position. An experimental observation of such correlations requires simultaneous imaging of both impurity and bath atoms with single-site resolution, which is a major challenge for future work. However, our observed opposite imprint of the average impurity density on the average bath density strongly supports the picture of a single moving polaron. Further discussion on the time evolution of the correlator $\mathcal{N}_{j,k}(j,k)$ and the correlation hole can be found in the Supplementary Information.

**Conclusion**

We have observed the dynamics of a deterministically created spin impurity in a 1D lattice across the superfluid–Mott-
insulator transition. The good agreement with numerical results demonstrates that our experiment is suitable for the quantitative study of mobile impurities in Bose–Hubbard models. In particular, a predicted and new universality class of single-particle excitations in 1D systems could be tested in the future by measurement of spin-flip response and Green’s functions. Other natural extensions of this work would be the investigation of the correlated propagation of several flipped spins due to the ferromagnetic spin–spin coupling in the Heisenberg model or the quantum simulation of the impurity dynamics in 2D, where numerical simulations and analytical studies are much more difficult. Our multiple-site addressing technique could also be used to prepare non-equilibrium states of quantum many-body systems such as domain walls and study their evolution. Further experiments could probe the fractionalization of excitations that naturally occurs in one dimension, either when magnons decay into two spinons, or when spin and charge excitations separate.

Methods

Simultaneous multiple-site addressing. We prepared the initial state with a single circularly shaped Gaussian laser beam focused onto a single lattice site together with a microwave field for addressing an individual spin in the lattice. Instead of a circular beam, we now used arbitrary light-intensity patterns created with a spatial light modulator of the digital-micromirror-device (DMD) type to address several atoms at the same time. We generated a line-shaped light pattern by reflecting a Gaussian laser beam off the DMD (Texas Instruments, DLP Discovery 4100, 1024 × 768 pixel, 13.7 μm pixel size), and coupled it into the high-resolution imaging setup with a dichroic mirror. The magnification was such that one pixel of the DMD corresponds to approx. 70 nm in the object plane. Compared with the diffraction limited spot size of ~600 nm, this oversampling allowed us to implement an error-diffusion algorithm to generate beam profiles with about only 10% root-mean-square variations of the peak intensity. As in our previous work, we applied a feedback that shifted the pattern on the DMD to compensate for slow phase drifts of the optical lattice.

The addressing laser had a wavelength of 787.65 nm and was σ− polarized to minimize the light shift for atoms in the initial state \( |F = 1, m_F = -1\rangle \), while creating a differential light shift between the initial state and \( |F = 2, m_F = -2\rangle \) of ~35 kHz. To flip the spin, we used an HSI pulse of 20 ms duration, 40 kHz sweep width and ~30 kHz frequency offset of the sweep centre from the bare resonance. The addressing beam was switched to full intensity for the spin-flip, and then back to 10% of the intensity for pinning the spin impurity (see main text) with s-shaped ramps of 10 ms duration. In relatively deep optical lattices, we generally used 100 randomly drawn initial states, which are as time-dependent matrix product states (also known as t-DMRG) and simulated the two-species Bose–Hubbard model of Supplementary Equation (1). For our parameters, considering matrix product states of dimension 200 is sufficient to describe the impurity dynamics for \( J/U \leq 0.25 \), whereas for \( J/U > 0.25 \) a dimension of 400 is more appropriate, as the subsystem entanglement grows significantly close to and inside the superfluid regime. We incorporated the experimental conditions and preparation sequences as follows: the finite temperature of the initial Mott-insulator state (experimentally prepared with an effective \( J/U = 0.053 \)), at a lattice depth \( V_x = 10 E_r, V_y = 30 E_r \) is considered by randomly sampling product states of localized atoms from the grand-canonical Gibbs ensemble for the given initial state, with \( J = 0\); zero-field Mott-insulator, this is a very good approximation. Chemical potential \( \mu \) and external confinement \( V_{\text{ext}} \) were determined from the experiment. We generally used 100 randomly drawn initial states, which are then subjected to time evolution in accordance with the parameter changes in the experiment. To obtain the thermal average of the impurity dynamics, the time-dependent local densities \( \rho_{ij}(t) \) for each initial state are used to form an unweighted average.

When we simulated the system in the Heisenberg limit, deep in the Mott-insulator regime, \( J/U \) does not change during the state preparation and the time evolution. The atom on the central site is spin-flipped and then evolved in time together with the background. When simulating the system at larger \( J/U \) than the initial one, we follow the experimental sequence exactly. Correctly modelling the release of the spin impurity is essential, as the timescales for switching the addressing laser are similar to the tunnelling times. Specifically, we simulated...
the adiabatic state preparation as it is done in the experiment using an initial ramp-down of the lattice depth \( V_0 \), within 50 ms according to

\[
V_i(t) = V_0 - \frac{V_i - V_0}{2} \left[ 1 + \coth \left( \frac{T}{\tau} \right) \tanh \left( \frac{2t - T}{\tau} \right) \right]
\]

with \( V_i = 10 E_\text{r}, T = 50 \text{ ms}, \tau = 15 \text{ ms} \) and \( V_0 \) being the desired final lattice depth. We also incorporated into the time-dependent Hamiltonian parameters the switching off of the addressing laser within 1 ms.

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Author contributions

All authors contributed extensively to the work presented in this paper.

Additional information

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Competing financial interests

The authors declare no competing financial interests.