Spin Dynamics Dominated by Resonant Tunneling into Molecular States

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Optical lattices and Feshbach resonances are two of the most ubiquitously used tools in atomic physics, allowing for the precise control, discrete confinement, and broad tunability of interacting atomic systems. Using a quantum simulator of lithium-7 atoms in an optical lattice, we investigate Heisenberg spin dynamics near a Feshbach resonance. We find novel resonance features in spin-spin interactions that can be explained only by lattice-induced resonances, which have never been observed before. We use these resonances to adiabatically convert atoms into molecules in excited bands. Lattice-induced resonances should be of general importance for studying strongly interacting quantum many-body systems in optical lattices.

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Introduction.—The field of cold atoms has enjoyed enormous progress in recent years, largely thanks to two powerful tools: optical lattices, which provide confinement and a way to controllably simulate discrete models, and Feshbach resonances, which allow for tunable interactions between particles. Consequently, there has been keen interest in studying how the lattice modifies atomic scattering properties [1–10]. In particular, several studies have theoretically predicted that the free-space Feshbach resonance vanishes and is replaced by multiple shifted "lattice-induced resonances" [11–14]. Here, we observe multiple resonances in the spin-spin interactions which we associate with near-resonant tunneling into molecular states with excited center-of-mass (c.m.) motion (Fig. 1). Our observations cannot be explained by confinement-induced resonances [1,15–24] but require the theory of lattice-induced resonances which have not been observed before [25]. In contrast to confinement-induced resonances, which rely on anharmonicities, lattice-induced resonances are caused by tunneling.

Since Feshbach resonances in combination with optical lattices have been used extensively in ultracold atom experiments for many years [2,26–33], it is surprising that none of them have observed the dramatic signature of lattice-induced resonances. A possible reason is that many experiments looked only for loss features or studied deep traps where the dynamics is suppressed. Our unique approach of measuring these resonances in the many-body dynamics of Heisenberg spin chains avoids the strong three-body loss, which enabled us to clearly observe and identify these resonances. Lattice-induced resonances should have important consequences for any system in an optical lattice with strong interactions.

In our work, we find that many-body spin dynamics is affected by free-space scattering lengths $|a_s|$ larger than $15\,000a_0$; this exceeds not only the oscillator length $a_{\text{osc}} = \sqrt{\hbar/\mu\omega} \approx 2\,500a_0$ but even the lattice spacing $a_{\text{lat}} \approx 10\,000a_0$. This is possible because the unitarity limit of the oscillator length applies only to two interacting atoms and not to the deeply bound molecular states involved in lattice-induced resonances. Here, a_0 is the Bohr radius, $\mu = m/2$ is the reduced mass, and ω is the trap frequency at the bottom of the sinusoidal lattice potential in the axial direction.

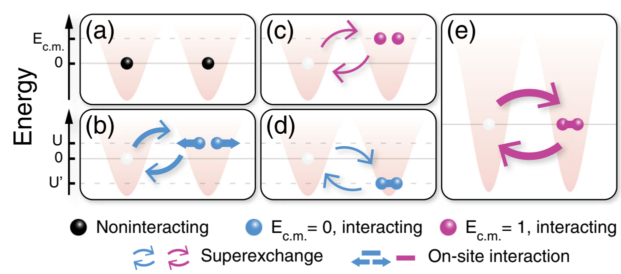

FIG. 1. Resonant tunneling into molecular states. An optical lattice in the Mott insulating regime with one atom per site (a) can host different virtual states, such as two atoms per site with repulsive interaction U (b); two atoms in an excited c.m. mode (c); or a molecule with negative binding energy U' (d). Both states (c) and (d) are typically far detuned from state (a). However, positive c.m. energy (c) can be combined with negative binding energy (d), bringing a molecule in an excited c.m. mode into resonance with the Mott insulator (e). State (a) is connected to (e) via a resonant tunneling process. These are lattice-induced resonances, observed here for a quasi-1D lattice.

Lattice-induced resonances are a phenomenon emerging from the paradigmatic Hamiltonian for particles with short-range interactions in a periodic potential:

$$H = \sum_i \left[\frac{p_i^2}{2m} + V_z \sin^2(kx_i)\right] + g\sum_{i\neq j}\delta(x_i - x_j), \quad (1)$$

where V_z is the lattice depth, k is the wave vector of the lattice light, and g is the interaction strength. This Hamiltonian is a model for many important materials and describes almost exactly the physics of ultracold atoms in optical lattices. We observe the new resonances using bosons in a 1D lattice, but these effects should occur also in higher dimensions and for fermionic systems.

Simple model of lattice-induced resonances.—Lattice-induced resonances can be understood by first analyzing the eigenstates of two interacting atoms on one lattice site. These states can couple to two atoms on adjacent sites via tunneling. When the negative binding energy of a molecule is offset by the positive energy of its c.m. motion, it becomes a zero-energy state [Fig. 1(e)]; consequently, two atoms on isolated sites can resonantly tunnel into this special molecular state. Near such resonances, the tunneling dynamics is greatly enhanced. This coupling is not possible in free space, since the c.m. energy of a bound molecule is determined by the c.m. momentum of the colliding particles. In a lattice with reduced translational symmetry, momentum conservation is replaced by quasimomentum conservation. Two colliding particles near zero quasimomentum can couple to bound states only with zero quasimomentum; however, these bound states can be in any band, which means that there are, in principle, an infinite number of accessible states.

To illustrate a simple case, we plot the spectrum of two particles in an isotropic harmonic trap with contact interactions in Fig. 2(a) [34]. The harmonic approximation captures several salient features of interactions in a deep optical lattice. Because of the separation of the relative and c.m. motion, only the relative motion varies with interaction strength. The total energy spectrum $U = E_{\text{c.m.}} + E_{\text{relative}}$ of both c.m. and relative motion yields infinite copies of the relative motion spectrum (blue curve) upshifted by contributions from the c.m. band structure (magenta, green, and gold curves). In an isotropic trap, the excited branches are degenerate. We note that the first excited molecular branch (magenta curve) crosses zero energy at the free-space Feshbach resonance ($1/a_s = 0$). When tunneling is introduced, the zero crossings of higher c.m. bands yield lattice-induced resonances.

Our experiment realizes one-dimensional chains with strong transverse confinement. Each lattice site, thus, resembles an *anisotropic* harmonic trap with ratio $\omega_x = \omega_y = 1.8\omega_z$ whose spectrum is plotted in Fig. 2(b), where the oscillator length is in units of the axial frequency

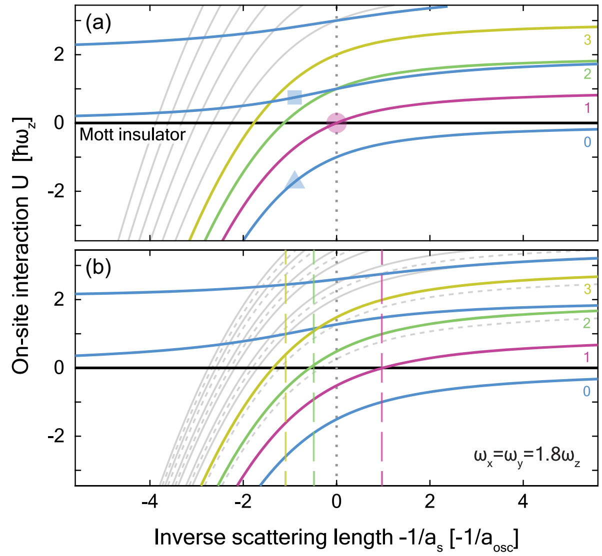

FIG. 2. Energy spectrum for two interacting atoms in a harmonic trap. In an isotropic trap (a), only the relative motion (blue curves) is affected by the scattering length a_s [34]. A copy of this level structure exists for each unit of c.m. excitation $\hbar\omega$. The lowest three c.m. excitations (magenta, green, and gold curves) for the lowest molecular branch become degenerate with the Mott insulator (black line) near a Feshbach resonance ($1/a_s = 0$, dotted line). The square, triangle, and circle correspond to the interactions of Figs. 1(b), 1(d), and 1(e), respectively. Our experimental conditions are more consistent with an anisotropic trap (b) with stronger transverse confinement [35]. In contrast to (a), the first excited branch crosses zero energy at negative finite scattering length. The vertical dashed lines correspond to the observed resonance positions seen in Fig. 3(b) and agree with the harmonic model. Solid gray curves correspond to the next six accessible c.m. excitations for both panels, while the dashed gray curves are branches with odd transverse excitations that have suppressed coupling to the Mott insulator (see Supplemental Material [36]). Colored numbers refer to z c.m. excitations. The first dashed gray curve corresponds to one excitation in either x or y and zero in z.

$a_{\text{osc}} = \sqrt{\hbar/\mu\omega_z}$. The anisotropy partially breaks the degeneracy and leads to a much richer energy spectrum, as all excitations of the z c.m. and even excitations of the x, y c.m. modes are allowed. As the transverse confinement is increased, the interaction energy between two atoms increases and reaches $\hbar\omega_z$ earlier. Hence, the first crossing is shifted to the right of the free-space Feshbach resonance.

Note that, in the harmonic approximation, interactions do not couple the ground and excited bands of the c.m. [34]. Anharmonicities can lead to coupling between them, which has been theoretically studied [24,46] and experimentally observed [47,48]. However, these couplings mix only c.m. states of the same parity, with two atoms *already on the same site*, and cannot couple the ground state to the first excited state. The feature of the lattice that allows lattice-induced resonances is interaction-driven tunneling.

As a particle tunnels onto a neighboring site, its direction breaks the parity symmetry. This allows an atom in the ground band (of an interacting system; see Supplemental Material [36]) to couple to *any* excited band along the direction of tunneling regardless of parity. While we use the harmonic trap model for simplicity, we emphasize that, in lattices, many of the energy level crossings become *avoided* crossings, and each band attains a finite width.

Effects of resonances on spin dynamics.—The spin dynamics in a singly occupied Mott insulator is driven by superexchange, a second-order tunneling process involving virtual intermediate states with two atoms on the same site. The resultant spin-exchange coupling J is

$$J \propto \sum_i -t_i^2/U_i, \tag{2}$$

where U_i is the energy of the intermediate state and t_i the tunnel coupling to the initial state. Because of the energy denominator, superexchange physics is a very sensitive probe for zero-energy states of two atoms per site and, therefore, for lattice-induced resonances. In the regime of small scattering lengths, the dominant intermediate state is two atoms per site without c.m. excitation, as it possesses the smallest energy defect. In this case, the relevant U_i is simply the single-band on-site interaction energy, and t_i the conventional tunneling. The energies of the excited bands are far detuned, and, therefore, their contribution to superexchange is highly suppressed. However, in the strongly interacting regime ($a_s/a_{\rm osc} \gtrsim 1$), lattice-induced resonances can bring the energy of these excited-band states into resonance. Near the zero crossings in Fig. 2(b), the second-order tunneling process Eq. (2) is resonantly enhanced.

Experimental methods.—Our quantum simulator uses ultracold lithium-7 atoms in an optical lattice to realize the two-component Bose-Hubbard model. The atoms are in the lowest two hyperfine states, labeled as a and b, forming a pseudospin. The system is then turned quasi-1D by lowering the lattice depth along the axial (z) direction ($V_{x,y,z} = 35E_R, 35E_R, 10.7E_R$). The bare tunneling t in the lowest band is approximately $0.0164E_R$ along the axial direction, where $E_R = h^2/8ma_{\rm lat}^2 \approx 25$ kHz is the recoil energy.

The SU(2) symmetry of the system implies that it can be mapped onto the spin-1/2 XXZ Heisenberg model

$$H_{XXZ} = \sum_i [J_{xy}(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + J_z(S_i^z S_{i+1}^z)] \tag{3}$$

when prepared as a singly occupied Mott insulator [32,49]. The effective spin-spin interactions J_{xy} and J_z are comprised of superexchange and off-site energies [32,50]. Away from the Feshbach resonance, conventional superexchange involving lowest-band doublons dominates; however, close to the Feshbach resonance, excited-band molecules can form as a result of lattice-induced resonances, and their resonant tunneling can dictate the dynamics of the system.

We have recently observed that spin helix states have an enhanced lifetime at a wave vector directly related to the spin anisotropy $\Delta \coloneqq J_z/J_{xy}$. We use this phenomenon now as a method to determine Δ. In general, Δ depends on interactions of atoms between all three combinations of spin states aa, ab, and bb and can be written (see Supplemental Material [36])

$$\Delta = \frac{J_{bb} + J_0}{J_{xy}}. \tag{4}$$

Here, J_0 and J_{xy} represent spin-exchange interactions between aa and ab atoms, which are not affected by the bb Feshbach resonances [32]. The spin-spin interactions between two b atoms $J_{bb} = J + 2V_{bb}$ incorporates the superexchange terms in Eq. (2) and off-site interactions V_{bb}. Off-site interactions are an extension of the standard Hubbard model, which includes only on-site interactions. Because they are small, they have not been observed for alkali atoms in a lattice before (see Supplemental Material [36]).

Figure 3(b) shows the main result of this Letter: four dispersive features which indicate resonances in the spin-spin interaction J_{bb}. These are signatures of lattice-induced resonances. Near the resonance positions, spin dynamics is dominated by resonant tunneling into molecular states. The strongest lattice-induced resonance we observe is caused by the *first* excited band of the c.m. motion coupled to the Mott insulator, which cannot be observed with rf spectroscopy starting with two atoms per site. Tunneling to excited bands is possible only due to interatomic interactions and can be classified as multiorbital bond-charge hopping [51]. There can be more lattice-induced resonances involving even higher c.m. bands, but the higher resonances have vanishing coupling, as the molecule is more tightly bound and may be broadened by the larger energy spread in higher bands. Therefore, only the lowest three features were observed. The dashed vertical lines in Figs. 2(b) and 3(b) correspond to each other. Note that the observed resonance positions are consistent with only axial c.m. excitations. We also observe the first experimental evidence for contact off-site interactions in lattices (see Supplemental Material [36]).

Sweeping into molecules.—The dispersive features in Fig. 3 confirm the presence of lattice-induced resonances which admix molecules in excited bands [Fig. 2(b)], i.e., create *virtual* molecules. We also observe *real* molecules by adiabatically sweeping the magnetic field across the lattice-induced resonance at 895.537 G in both directions without crossing the free-space resonance. Such sweeps create a high fraction of excited-band molecules as shown in Fig. 4. We note the upsweep could not create molecules

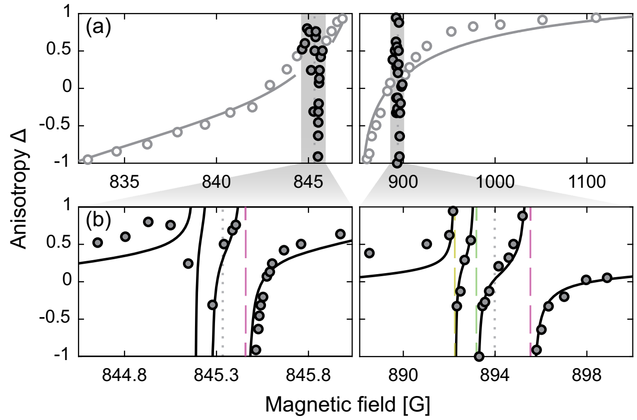

FIG. 3. Spin interaction anisotropy at different magnetic fields. We plot the anisotropy $\Delta = J_z/J_{xy}$ as a function of the magnetic field (a) and enlarged for fields with large bb scattering lengths $|a_{bb}| \gtrsim 500a_0$ (b). Far from the bb Feshbach resonances (dotted lines), the data (open circles) agree well with perturbative calculations (gray curves). Such theory is not applicable in the regions with large scattering lengths (filled circles). The dispersive features at 845.459, 892.249, 893.182, and 895.537 G (dashed vertical lines) are consistent with lattice-induced resonances corresponding to band excitations of $E_{\mathrm{c.m.}}/\hbar\omega_z = 1$, 2, and 3 (magenta, green, and gold curves, respectively). Note that the behavior of the spin anisotropy is smooth across the free-space resonances. Some data are inherited from Ref. [52]. Fits are shown in black curves (see Supplemental Material [36]). The statistical errors of the data and fitted resonances are smaller than the symbols and widths of the dashed lines, respectively.

in a free-space Feshbach resonance, which creates molecules only in one direction of the sweep since the stable molecular branch ends at the resonance position. In contrast, lattice-induced resonances have a stable branch far from the resonance in *either* direction. We repeated these sweep experiments for a Mott insulator of a atoms and found an analogous high molecule conversion around 739.1 G (see Fig. S1).

Our experiment resembles studies that sweep an energy gradient across the value U/a_{lat}, which was shown to map to the Ising model in a transverse field [53–55]. In these works, the sweep drives a quantum phase transition to antiferromagnetic order [56]. However, without the directionality of the gradient, we do not expect antiferromagnetic order in our experiment but a correlated state with an equal number of molecules and holes. This state shares some features with a quantum spin liquid: namely, long-range entanglement between particles and massive degeneracy in the ground state. This could be an interesting test bed for future work to study quantum thermodynamics in many-body systems. While these bound states are in an excited Bloch band which will suffer from collisional loss, the lifetime may be long enough to explore interesting new phases of matter as experimentally shown in Refs. [57,58].

Discussion.—Our work reports the first observation of the long-predicted lattice-induced resonances [13].

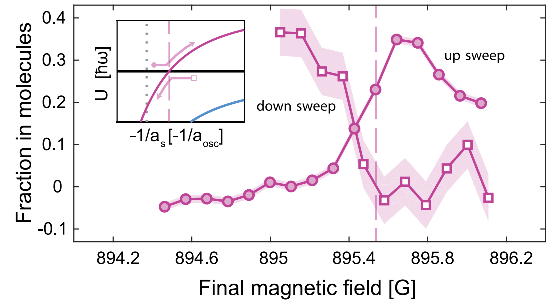

FIG. 4. Creation of molecules with adiabatic sweeps through a lattice-induced resonance. We sweep the magnetic field to different values without crossing the free-space resonance at 894 G (inset, dotted line). In a 3D lattice with shallow axial confinement, molecules are created when sweeping up (filled circles) or down (open squares) past the lattice-induced resonance (dashed line). Statistical errors are given by the shaded area and are larger for the down sweep due to fewer data points. The reduction of molecule fraction beyond 895.7 G can be explained by the finite time of the sweep which was kept constant at 15 ms for all final fields. The small offset of the crossover between the sweeps can be explained by hysteresis of the magnetic field during the ramp.

The resonant tunneling can completely determine the dynamics of a system with strong interactions. The dramatic modification of spin dynamics in our experiment was caused by the three lowest-lying lattice-induced resonances. Higher resonances (with even c.m. excitations) can, in principle, also occur via anharmonicities, and some (if the excitations are transverse) could be classified as confinement-induced resonances. We have not observed any signatures of these higher resonances and expect them to be much weaker than the measured lattice-induced features. Furthermore, because the lowest-lying excitation seems to be the most pronounced, it is very possible that only lattice-induced resonances have a significant impact on the dynamics of a system with unity filling. Further theoretical work is required to understand how the higher-band resonances can affect spin interactions and dynamics in a lattice.

We expect our results to be important for future studies of strongly interacting atoms in optical lattices. Lattice-induced resonances can be used to tune spin-spin interactions and possibly probe new phases of matter. For instance, as shown in Ref. [11], different contributions of superexchange cancel at special points. This could allow higher-order processes to dominate the dynamics and drive the system into exotic phases. It would also be interesting to see how these lattice-induced resonances modify scattering properties of atoms in the superfluid phase, as they should tune elastic interactions and affect the mean field energy of a BEC. Additionally, the adiabatic sweeps to molecules may create novel correlated many-body states. It is an intriguing question whether they can be described as a superfluid state of molecules or some other exotic phase and how such states thermalize.

Note added.—Recently, Ref. [59] reported the observation of confinement-induced resonances in a 3D lattice.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Y. K. L., H. L., and W. K. conceived the experiment. Y. K. L. and H. L. performed the experiment. Y. K. L. and H. L. analyzed the data. All authors discussed the results and contributed to the writing of the manuscript.

The authors declare no competing financial interests.

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