Coherent multi-flavour spin dynamics in a fermionic quantum gas

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Microscopic spin-interaction processes are fundamental for global static and dynamical magnetic properties of many-body systems. Quantum gases as pure and well-isolated systems offer intriguing possibilities to study basic magnetic processes including non-equilibrium dynamics. Here, we report on the realization of a well-controlled fermionic spinor gas in an optical lattice with tunable effective spin ranging from 1/2 to 9/2. We observe long-lived intrinsic spin oscillations and investigate the transition from two-body to many-body dynamics. The latter involves a complex interplay of spin and spatial degrees of freedom and implies an instability of an initially band insulating state. Using an external magnetic field we control the dimensionality of the system and tune the spin oscillations in and out of resonance. Our results open new routes to study quantum magnetism of fermionic particles beyond conventional spin 1/2 systems.

Magnetism plays a key role in the fundamental understanding of materials and in modern technologies. A major focus is the understanding of quantum properties of magnetism, which have their origin in the underlying microscopic processes between elementary spins. Despite much theoretical effort, it remains challenging to derive macroscopic magnetic phenomena such as the fractional quantum Hall effect or the formation of spin liquids directly from first principles. At this point, scalable and controllable model systems could allow this gap to be bridged. Alongside a few tunable magnetic condensed matter systems, atomic physics experiments came into focus in recent years owing to their unrivalled control over all experimental parameters and nearly perfect isolation from environmental influences. Ion chains as well as bosonic quantum gases, either harmonically trapped or confined in optical lattices, have produced striking results towards the simulation of classical and quantum magnetism. Lacking Pauli blocking, however, these experiments did not catch the fermionic character of electronic magnetism. In this direction, the possibility of itinerant ferromagnetism and spin transport in bulk fermionic quantum gases has recently been discussed. To resemble electron spins in real solids even better, it is desirable to have a fully controllable fermionic lattice quantum simulator. In addition, fermionic atoms with spin $s > 1/2$ constitute the building blocks of completely new physical systems that go beyond conventional electronic magnetism. Theoretically proposed phenomena in this direction include unconventional Bardeen–Cooper–Schrieffer superfluid, quantum-chromodynamic-like colour superfluidity, SU(N)-magnetism with alkaline earth atoms and further exotic phases in multi-flavour systems.

Here, we demonstrate the first experimental realization of a well-controlled fermionic spinor gas with interaction-driven long-lived coherent spin oscillations. By properly choosing the initial spin states we can change the effective spin from 1/2 to 9/2. The control over the magnetic field allows us to initialize and stop spin dynamics and to select the number of involved levels. We extract the microscopic interaction parameters and find excellent agreement with a two-particle model including all spin-dependent interactions. By tuning the depth of the optical lattice, we investigated the transition from very well controlled pure on-site spin dynamics to highly complicated many-body systems where spin and spatial degrees of freedom are coupled.

We perform our experiments employing a fermionic spinor gas of $^{40}$K atoms in the $f = 9/2$ manifold. Initially, we prepare an equal mixture of two different spin states with $N = 4 \times 10^5$ atoms in an optical lattice, forming a large-scale band insulator (diameter about 50 lattice sites) as depicted in Fig. 1a (for details see Methods). To shed light on the microscopic collision processes, we first consider two fermionic atoms in different spin states $|m_1 \rangle$ and $|m_2 \rangle$ in the lowest spatial mode on an isolated lattice site, forming a two-particle state $|m_1, m_2 \rangle = 1/\sqrt{2}(|m_1 \rangle |m_2 \rangle - |m_2 \rangle |m_1 \rangle)$. For spin 1/2 particles with solely two hyperfine states, analogous to electrons in solids, there is only one possible two-particle state. For high-spin particles ($s > 1/2$), spin-changing collisions can transfer the atoms into new states $|m'_1, m'_2 \rangle$ at small magnetic field. For increasing spin, the number of involved two-particle states typically increases. Spin-changing collisions of fermionic atoms have to satisfy two physical restrictions: conservation of total magnetization $M = m_1 + m_2$ and Pauli blocking ($m_1 \neq m_2$). For simplicity, consider the case of spin 5/2 as shown in Fig. 1b. Depending on the initially chosen spin states, either a three-level (the involved two-particle states are $| \uparrow, \downarrow \rangle, |\uparrow', \downarrow' \rangle$ and $| \rightarrow, \leftarrow \rangle$), a two-level ($| \uparrow', \rightarrow \rangle$ and $| \uparrow, \leftarrow \rangle$) or a one-level ($| \rightarrow, \rightarrow \rangle$) system can be realized. Employing $^{40}$K in $f = 9/2$ with its ten spin states allows us to experimentally vary the number of involved two-particle states between one and five.

Experimentally, this regime of two-body interaction-driven dynamics in a high-spin system can be investigated in very deep optical lattices, where tunnelling can be neglected and isolated pairs of atoms reside on the individual lattice sites. In a first set of experiments, we recorded the time evolution of exclusively doubly occupied sites (see Methods) in the initial two-particle state.

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Relative population of different spin states

0.4 0.8 0.6

0.1 0.2 0.3 0.4 0.5 0.6 0.7

Figure 1 | Principles of fermionic lattice spin dynamics. a. Sketch of atoms in an optical lattice with harmonic confinement. A band insulator (with two particles per site) is formed in the centre within the lowest spatial state. b. Microscopic collision processes on an isolated lattice site. Spin states for spin 5/2 particles are exemplarily shown (left). The coloured boxes represent possible two-particle states (right). These states are coupled by spin-changing collisions, forming one-level, two-level and three-level systems, where the number of levels is given by the amount of allowed two-particle states. Magnetization conservation and Pauli blocking restrict the number of involved two-particle states. States represented by grey boxes are forbidden owing to Pauli blocking.

\[ |m_1, m_2\rangle = |1/2, 9/2\rangle, \text{ which is coupled only to } |3/2, 7/2\rangle. \text{ As the state } |5/2, 5/2\rangle \text{ is forbidden owing to Pauli blocking, this constitutes a two-level system. As one central result of this work, we observe long-lived coherent spin oscillations of fermionic atoms for the first time. The oscillations are clearly visible for more than 250 ms with an amplitude in excess of 70% as shown in Fig. 2a.} \]

Two energy scales govern the dynamics of the spin-changing collisions as depicted in Fig. 2b: the difference in interaction energy and the difference in Zeeman energy between the individual two-particle states. At large magnetic fields, the atoms are pinned to a fixed spin by the Zeeman energy. For small magnetic fields, the spin interaction becomes relevant and spin dynamics occurs. The new eigenstates become quantum mechanical superpositions of the non-interacting two-particle states. A resonant feature appears in the spin oscillations when Zeeman energy and spin-dependent interaction energy are equal\(^{41-43}\), that is, the involved two-particle states are degenerate (see Supplementary Information S2 for details).

For the experiments, we first prepared the atoms in the Zeeman-dominated regime at large external magnetic field. Quenching the magnetic field to a smaller value \(B_{\text{ex}}\) initializes the observed spin oscillations. We investigated the crossover between the interaction-dominated and the polarized regime by studying spin dynamics at different \(B_{\text{ex}}\). Extracting both frequency and amplitude of the oscillations, a clear Rabi-resonance feature as expected for a two-level system is observed and depicted in Fig. 2c. At resonance, the observed amplitude possesses a maximum and the frequency is minimal. For larger magnetic fields, the system approaches the polarized regime and the spin oscillations vanish. We fit our data to a two-particle model and find excellent agreement. The oscillation frequency and thus the shape of the resonance is related to the difference of the scattering lengths \(\alpha\) in the respective scattering channels with total spin \(F = 8\) and \(F = 6\), which we use as the only free fitting parameter (see Supplementary Information S1, S2 and S6). Our measurements hence also provide a high-precision test for molecular calculations of scattering lengths. We find good agreement between our fitted value of \(\alpha_8 - \alpha_6 = 2.26 \pm 0.07 \alpha_6\) and a theoretical prediction using coupled-channel calculations of \(\alpha_8 - \alpha_6 = 168.53 \alpha_6 - 166.00 \alpha_6 = 2.53 \alpha_6\) (for details see Supplementary Tables ST1 and ST2).

Beyond these pure on-site effects, we observe a slow damping of the coherent oscillations with a rate of \(\Gamma^{-1} = 96.4 \pm 4.6\) ms. In addition, new spin states such as \(|m\rangle = |5/2\rangle\) and \(|-1/2\rangle\) appear, which would be forbidden at zero tunnelling. We have excluded magnetic field noise (\(\Delta B_{\text{ex}} \leq 3\) mG) and gradients (\(\Delta B \leq 0.5\) mG over the sample) to considerably contribute to the dephasing of the coherent
Figure 3 | Coherent multi-flavour spin dynamics of fermionic atoms. 

(a) Coherent spin oscillations with five two-particle states involved. Plotted are the relative populations $n(m)$ of different spin states $|m\rangle$ versus time. The solid lines are fits to the data based on our numerical model (see Supplementary Information S5). The lattice depth is $25 E_r$ and $B_{\text{exp}} = 0.372 \text{ G}$. At these parameters, four levels effectively participate in the spin evolution, leading to pronounced beat notes in the signal. 

(b) Calculated amplitudes of the overlap integral $|\langle n| - 1/2, 1/2 \rangle|^2$ between the initial state and all five two-particle eigenstates $n = 1, \ldots, 5$ versus the magnetic field. At large magnetic fields, the ground state coincides with the initial state. For smaller magnetic fields, an increasing number of eigenstates overlap and thus contribute to spin oscillations. 

(c) Observed frequencies, obtained by discrete Fourier analysis of spin oscillation data such as in (a) for different magnetic fields with corresponding errors. Curves show the numerically calculated frequencies. Their widths are given by uncertainties in lattice depth. The shading of the curves is proportional to the expected strength of the transition as shown in (b). 

(d) Typical discrete Fourier spectra for the populations $n(1/2) + n(-1/2)$ (black lines) and $n(3/2) + n(-3/2)$ (red lines) at magnetic fields $B_{\text{exp}} = 0.372 \text{ G}$ (left) and $B_{\text{exp}} = 1.014 \text{ G}$ (right). 

Spin-changing oscillations. Hence, we attribute the damping as well as the appearance of the new spin states to tunnelling processes at edges and defects, as we outline in more detail below.

In a second set of experiments, we realized a high-spin system by preparing the state $|1/2, -1/2\rangle$ (see Methods), which is coupled to the states $|3/2, -3/2\rangle$, $|5/2, -5/2\rangle$, $|7/2, -7/2\rangle$ and $|9/2, -9/2\rangle$. This constitutes an effective five-level system and is the maximum spin system available employing $^{40}\text{K}$. The high-spin system allows for transitions between two-particle states with $\Delta m = 1, 2, 3$ and 4, which involve up to ten spin oscillation frequencies. This becomes apparent in Fig. 3a, where the corresponding beat-notes of the signal are clearly visible, revealing the coherent nature of the local interaction also in the high-spin system.

We have investigated the spin dynamics of this multi-flavour system at different magnetic field strengths and find that the amount of contributing oscillation frequencies increases with decreasing magnetic field (Fig. 3c). This is in good agreement with our numerical simulation of the complete multi-flavour dynamics, which reveals that the external magnetic field provides full control over the effective dimensionality of the system in spin space (for details see Supplementary Information S2). The number of eigenstates significantly overlapping with the initial state depends crucially on the magnetic field as shown in Fig. 3b. Each observed frequency can be assigned to the superposition of two eigenstates. These frequencies are also very well reproduced by our calculations, explicitly if we include all possible spin-changing collisions, that is $\Delta m = 1, 2, 3$ and 4. Note that this is to our best knowledge the first realization of spin-changing collisions with $\Delta m > 1$. Typical Fourier spectra for the effective four-level system at a magnetic field of $B_{\text{exp}} = 0.372 \text{ G}$ and for the effective two-level system at a magnetic field of $B_{\text{exp}} = 1.014 \text{ G}$ are exemplarily shown in Fig. 3d. In this multi-flavour spin system with its rich dynamics, the observed damping is similar to the two-level case.

The experiments presented so far have been performed in deep optical lattices, in which spin dynamics is strongly dominated by local interactions on the individual lattice sites. Note that in a band insulating system tunnelling is forbidden even for shallow lattices and thus the number of particles per site is fixed. We find however strong evidence, that for shallow lattices, the initially insulating state exhibits an instability that is a direct consequence of the high-spin ($s > 1/2$) system in combination with spin-changing collisions. Even in a perfect band insulator, this instability leads to a finite tunnelling probability. In the following, we present our experimental results on this crossover from deep to shallow lattices. We again started in the $|1/2, 9/2\rangle$ effective two-level band insulator, then decreased the lattice depth along one spatial dimension to a final value $V_L$ and tracked the time evolution of the system in spin space. With decreasing lattice depth, we observe three effects (see Fig. 4a). First, the oscillation frequency decreases, which is consistent with our pure two-particle model because the on-site density determines the oscillation frequency, resulting in slower dynamics for shallower lattices (Fig. 4b). Second, we observe an enhanced damping, leading to a complete disappearance of the spin oscillations for very shallow lattices with the damping constants being of the order of the single-particle tunnelling energy $J$ (see Supplementary Table ST3). Third, the population of the spin states $|5/2\rangle$ and $|1/2\rangle$ increases faster for shallower lattices.

In the following, we show that both the observed damping as well as the appearance of the formerly forbidden spin states can be explained with tunnelling processes that are especially for shallow lattices a result of the spin-changing collisions. This implies that the above-mentioned intrinsic instability of the initial band...
insulator is directly induced by the spin-changing interactions. For simplicity, we limit the explanation to the dominating tunnelling processes shown in Fig. 5. First, tunnelling occurs at edges or defects of the initial band insulator when a singly occupied site is adjacent to a doubly occupied site, which is depicted in Fig. 5a (top). This scenario is immanent for a finite-size and finite-temperature fermionic system as realized in the experiment. After a spin-changing collision on the doubly occupied site (upper middle), a tunnelling process can form another two-particle state on the initially singly occupied site (lower middle). This leads first to a diminished contrast of the spin oscillations and as a second-order effect to the formation of new spin states (bottom). This process is possible at any lattice depth and the probability of such an event is expected to be proportional to $J$. In the band insulating core of the system, a second less obvious tunnelling mechanism occurs in the presence of spin-changing collisions. Consider two coupled lattice sites, as shown in Fig. 5b, that are initially filled with the same two-particle state $| \downarrow, \uparrow \rangle$, as realized in the centre of the band insulator (top). Spin dynamics on each individual lattice site leads to a time-dependent superposition of the two-particle states $| \uparrow, \uparrow \rangle$ and $| \downarrow, \downarrow \rangle$ (upper middle). In this case, Pauli blocking no longer prevents tunnelling (see Supplementary Information S4), because more than two spin states are present in the system, which in principle allows for triple occupation of the individual lattice sites, being equivalent to a finite tunnelling probability. Concerning the global properties of the system, this implies that in the presence of local spin-changing collisions, the initial band insulator becomes unstable. Again, these tunnelling processes allow, either through first-order tunnelling processes or superexchange, for the realization of new spin states (bottom). The corresponding timescales for these processes are proportional to $1/J$ and $U/J^2$, respectively.

For a deeper understanding of the relevant dynamical processes, we performed intensive numerical simulations of a four-well system using exact diagonalization methods (see Supplementary Information S3). We calculated the time evolution at different lattice depths as shown in Fig. 6 and find a very good agreement with the experimental results (see Fig. 4). For deep lattices, the numerically calculated oscillations in a perfect initial band insulator are undamped on the experimental timescale, because tunnelling is strongly suppressed owing to on-site interactions and superexchange is not relevant on these timescales. By taking into account possible defects, that is singly occupied sites, the numerical simulations reproduce the experimentally observed damping as well as the growth of the new spin states in the case of deep lattices (top). In contrast, for shallow lattices, tunnelling in the core alone already leads to a strong decrease of the spin oscillation amplitude. We find that this behaviour is very robust and that a finite number of defects does not change the dynamics significantly (bottom). Independently of the number of defects, the damping occurs on a timescale faster than the spin-changing dynamics, leading to the complete disappearance of the oscillations, in good agreement with our experimental observations (see Fig. 4). The finite size of the calculated system does not allow for quantitative predictions, but the decrease of oscillations is generally even more pronounced for larger systems. The simulations clearly show that for shallow lattices, core tunnelling leads to the observed damping and appearance of new spin states. Note that this instability is only possible in a high-spin fermionic system and is completely absent in conventional spin 1/2 systems.

We have thoroughly studied a high-spin fermionic spinor gas in an optical lattice. Our results reveal the complex dynamics of these systems governed by the interplay between spin-dependent interactions, Zeeman energy, Pauli blocking and tunnelling. For the first time, we have observed long-lived coherent spin oscillations of fermionic atoms and demonstrated the ability to fully control these spin oscillations. The results in deep optical lattices show excellent agreement with a two-body calculation in the zero-tunnelling limit. In particular, we have realized for the first time coherent spin oscillations with $\Delta n > 1$. By investigating the crossover to the tunnelling-dominated regime, we have observed an interaction-induced instability of the initially band insulating system. The magnetic local contact interactions, which address only the internal degrees of freedom of the fermionic atoms, have led to a fundamental change of the global insulating property of the investigated large-scale system. Our results underline the capability of fermionic spinor gases in optical lattices for the investigation of complex many-body phenomena that are governed by a strong interplay of external and internal degrees of freedom, for example, spin transport, spin–charge separation, spin waves or spin–orbit coupling. In the strongly correlated regime, spin dynamics could serve as an intriguing tool to directly probe superexchange processes. The tunability of the effective spin opens up the route to further study high-spin magnetism in comparison with conventional spin 1/2 electrons and shows the ability to access and engineer fundamentally new multi-flavour spin systems. Especially for high-spin systems, our experiments also pave the way for coherent control of entanglement and studies dedicated to quantum coherence properties.

Figure 4 | Transition from on-site to many-body spin dynamics. a, Relative populations of different spin states $n(9/2) + n(1/2)$ (red), $n(7/2) + n(3/2)$ (blue), $n(5/2)$ (green) and $n(-1/2)$ (purple) as a function of time. Solid lines are fits to the data, from which we extract oscillation amplitudes and frequencies (see Supplementary Information S5). The magnetic field is $B_{\text{exp}} = 0.186 \, \text{G}$. The lattice depth $V_L$ along one dimension is indicated on the right-hand side; for the other two dimensions the lattice depth is $35 \, E_r$. The transition from single-site-dominated high-contrast oscillations to diffusing spins in the high-tunnelling regime is clearly visible with decreasing lattice depth. In the latter case, spin oscillations in the initial channels are strongly damped, while at the same time tunnelling leads to the occupation of new spin states. b, Extracted oscillation frequencies from a, compared with a two-particle calculation (solid line). Error bars representing two standard deviations lie within the data points. The width of the curve is given by the lattice depth uncertainty.
Figure 5 | Simplified sketch of processes originating from the combination of spin-changing collisions and tunnelling. Shown are two lattice sites coupled by finite tunnelling. Two characteristic situations are distinguished, each of them leading to a decrease of the oscillation amplitude and to the generation of new spin states, which are forbidden on perfectly isolated lattice sites. a. At edges and defects, tunnelling from the doubly occupied to the singly occupied site occurs. b. In the core of the system, there is tunnelling between doubly occupied sites. The dominating processes are first-order tunnelling (left), inducing particle number fluctuations, and superexchange (right) where the on-site particle number is constant.

Methods
Preparation of a fermionic quantum gas in an optical lattice. By sympathetic cooling with $^{87}$Rb, about $N = 2 \times 10^6$ spin-polarized $^{40}$K atoms are cooled to quantum degeneracy in a magnetic trap. The potassium atoms are transferred adiabatically to a crossed optical dipole trap operated at 812 nm. After switching off the magnetic trap, a series of radiofrequency pulses and sweeps is applied to prepare an equal mixture of two hyperfine states. The atoms are evaporatively cooled by exponentially ramping down the trap laser intensity within 2 s. The resulting spin mixture now consists of about $N = 4 \times 10^4$ atoms at typical temperatures between 0.15 $T_j$ and 0.25 $T_j$. For the presented experiments, the magnetic field after the evaporation is 7 G for the mixture $|1/2, -1/2\rangle$ and 3 G for $|3/2, -7/2\rangle$. To increase the chemical potential, we ramp up the depth of the optical dipole trap in 50 ms and obtain final trapping frequencies of $\omega_z = 2\pi \times 125$ Hz in the vertical direction and $\omega_x = 2\pi \times 41$ Hz and $\omega_y = 2\pi \times 32$ Hz in the horizontal plane. Afterwards, we adiabatically ramp up a 3d cubic optical lattice in 150 ms. It consists of three orthogonal retro-reflected laser beams at $\lambda = 1.030$ nm with a $1/e^2$ radius of 200 $\mu$m, detuned with respect to each other by several tens of megahertz. The uncertainty of our lattice depth calibration is $\pm 2\%$. The lattice depth and chemical potential are chosen such that in the centre of the system a band insulator is formed containing about 40% of the atoms, which we infer from the measured double occupancy. Note that only at low enough temperatures a sufficiently pure two-component band insulator is formed in the core of the lattice, which is crucial to observe high-contrast spin oscillations. After the preparation procedure, we initialize spin dynamics by rapidly decreasing the magnetic field to a value $B_{\text{off}}$ with an accuracy of about $\pm 3$ mG.

Microwave transfer of singly occupied sites. Atoms on singly occupied sites do not participate in the spin-changing dynamics and contribute only as a constant offset to the signal. We circumvent this by applying microwave pulses to transfer all singly occupied sites to the $f = 7/2$ hyperfine manifold, where the atoms are not resonant with our detection light (the $f = 7/2$ states are separated by a frequency of 1.3 GHz from the $f = 9/2$ states). For this, we prepare the atoms in the state $|3/2, 7/2\rangle$ and let the system evolve on resonance for one-quarter of a spin-oscillation period. Now, the doubly occupied sites are in the state $|1/2, 9/2\rangle$. At this moment we stop the dynamics by ramping up the magnetic field to $B = 1.69$ G. Then, we apply four linearly polarized microwave pulses with a length between 35 and 50 $\mu$s and transfer all atoms on singly occupied sites in the remaining spin states ($|1/2, 3/2\rangle$, $|5/2\rangle$ and $|7/2\rangle$) into the $f = 7/2$ manifold. Subsequently, we restart the spin dynamics.
from a very pure state, where all atoms contributing to the signal are in the two-particle state |1/2, 9/2⟩. Thus, we observe high-contrast spin oscillations.

In addition, the admixture of new spin components due to defect tunnelling is strongly suppressed because atoms in |j = 7/2⟩ do not exchange their spin with atoms in |j = 9/2⟩. Note that this procedure was applied for the measurements starting from the initial state |1/2, 9/2⟩ in Figs 2 and 4.

Counting of spin components. After a variable evolution time, we stop the spin oscillations and detect the relative occupations in the different states |m⟩. To stop the oscillations we increase the magnetic field rapidly, which pins the atoms to their momentary state. Afterwards, we ramp down the lattice potential in 500 μs and hence map the quasimomenta of the atoms onto real momenta. After all optical potentials are switched off, we perform a Stern–Gerlach separation of the different spin states within a time-of-flight of 18.5 ms. The atoms are finally detected by absorption imaging with a short pulse of resonant laser light. We count the number of atoms in all spin states, accounting for saturation effects due to the spatial variation of the pulse intensity.

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