CONCEPTUAL MATTER PHYSICS

Enhanced quantum coherence in exchange coupled spins via singlet-triplet transitions

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Manipulation of spin states at the single-atom scale underlies spin-based quantum information processing and spintronic devices. These applications require protection of the spin states against quantum decoherence due to interactions with the environment. While a single spin is easily disrupted, a coupled-spin system can resist decoherence by using a subspace of states that is immune to magnetic field fluctuations. Here, we engineered the magnetic interactions between the electron spins of two spin-1/2 atoms to create a “clock transition” and thus enhance their spin coherence. To construct and electrically access the desired spin structures, we use atom manipulation combined with electron spin resonance (ESR) in a scanning tunneling microscope. We show that a two-level system composed of a singlet state and a triplet state is insensitive to local and global magnetic field noise, resulting in much longer spin coherence times compared with individual atoms. Moreover, the spin decoherence resulting from the interaction with tunneling electrons is markedly reduced by a homodyne readout of ESR. These results demonstrate that atomically precise spin structures can be designed and assembled to yield enhanced quantum coherence.

INTRODUCTION

The coherent control of spin states is a prerequisite for the use of spins in quantum information technologies (1–3). However, the quantum properties of spin states in solid-state nanostructures are easily disrupted by interactions with the environment such as electric or magnetic field noise (4), as well as unwanted coupling to nearby spins (5, 6). To protect the spin states against decoherence, ion traps (7, 8), silicon-based qubits (9), and quantum dots (10–12) adopted particular spin transitions called “clock transitions,” which are inherently robust against magnetic field fluctuations (7, 13). By carefully tuning the parameters in the spin Hamiltonian of a coupled electron-nuclear (7, 8) or electron-electron system (9), these clock transitions have been created and shown to be insensitive to magnetic field noise, at least to first order.

To experimentally address sources of decoherence, well-controlled studies of individual spin centers are critical (13). Scanning tunneling microscopy (STM) has been intensively used to construct and characterize spin structures (14, 15). While the spin relaxation time \( T_1 \) of individual atoms (16, 17), molecules (18), and nanostructures (19–21) has been studied using STM, the spin coherence time \( T_2 \) of surface atoms is mostly discussed for individual atoms (17, 22) and in theoretical works (23–25). Recently, electron spin resonance (ESR) in STM has been applied to electrically sense and control individual magnetic atoms on the surface (26) as well as interactions between them (27–29). Combining the high-energy resolution of ESR and the capability of STM to position individual spin centers with atomic precision, ESR-STM now enables the exploration of decoherence in assembled nanostructures.

In this work, we create a two-level system using magnetic-field–independent spin states of two magnetically coupled spin-1/2 titanium (Ti) atoms. The spacing between the atoms is precisely chosen to create a relatively strong magnetic coupling (~30 GHz) that protects the spin states from fluctuating magnetic fields. The two-level system consists of the singlet and triplet states having magnetic quantum number \( m = 0 \), and thus, it is not sensitive to magnetic field fluctuations to first order (3). This gives a spin coherence time that is more than one order of magnitude longer compared with other states in this system of coupled atoms and with individual Ti atoms. We further improve the coherence time by setting the DC bias voltage to zero to reduce decoherence induced by tunneling electrons (22). This is achieved by using homodyne detection, a mechanism previously used in electrical detection of ferromagnetic resonance (30, 31) and here applied to ESR-STM.

RESULTS

Spin resonance of singlet and triplet states

We used a low-temperature STM that allows imaging, atom manipulation, and single-atom ESR (Fig. 1A) (26–29). One or a few Fe atoms were transferred to the tip apex to create a magnetic tip for ESR driving and sensing. We deposited Ti atoms on two monolayers (ML) of MgO grown on Ag(001) (see Materials and Methods and section S1). On this surface, Ti atoms have two binding sites: on top of the oxygen atom (Ti\textsubscript{O}) and at the bridge site between two oxygen atoms (Ti\textsubscript{B}). Both species have a spin of 1/2, most likely due to an attached H atom (29), and show negligible magnetic anisotropy; thus, to good approximation, the spins align to the uniform external magnetic field \( \mathbf{B}_{\text{ext}} \) (fig. S2).

We positioned two Ti atoms to form Ti\textsubscript{O}–Ti\textsubscript{B} dimers (Fig. 1A) and characterized the magnetic interactions between Ti atoms using ESR. During ESR data acquisition, the STM tip is positioned over the Ti\textsubscript{O} atom because it yields a better ESR signal amplitude than Ti\textsubscript{B} (fig. S10). Furthermore, this position compensates for the subtle difference in the gyromagnetic ratios of Ti\textsubscript{O} and Ti\textsubscript{B} atoms (figs. S2 and S6). When two spin-1/2 atoms are magnetically coupled, the eigenstates are given by the singlet (\(| S \rangle \)) and triplet (\(| T_0 \rangle, | T_+ \rangle, | T_- \rangle \)) states. While two of the triplet states are the Zeeman product states (\(| T_- \rangle = |0\rangle \otimes |0\rangle \) and \(| T_+ \rangle = |1\rangle \otimes |1\rangle \)), the spin–spin interaction causes the superposition of |0\rangle and |1\rangle states and results in the remaining two eigenstates: \(| S \rangle = (|0\rangle \otimes |1\rangle) / \sqrt{2} \) and \(| T_0 \rangle = (|0\rangle + |1\rangle) / \sqrt{2} \). Here, 0 and 1 designate the spin-up and spin-down states of the constituting spins, respectively.

Figure 1B shows an ESR spectrum obtained from a Ti\textsubscript{O}–Ti\textsubscript{B} dimer with the atomic separation \( r = 0.92 \) nm. Four ESR peaks arise from the four transitions that change the total magnetic quantum
number $m$ by ±1, as given in the schematic energy diagram (Fig. 1D). The peak heights are proportional to the difference in thermal occupation of the initial and final states (29); thus, the two tallest peaks correspond to transitions out of the ground state. The difference between resonance frequencies ($f_{T_0T} - f_{ST}$ or, equivalently, $f_{ST} - f_{T_0T}$) directly gives the magnetic interaction energy $J$ between the two spins (29), which is 0.77 ± 0.02 GHz for this dimer spacing (fig. S5). Because the interaction energy $J$ is smaller than the Zeeman energy, the $|T_0\rangle$ state becomes the ground state.

We find that the Ti$_O$-Ti$_B$ dimer can be positioned close enough to yield coupling strengths sufficiently strong to shift the singlet state down in energy to become the ground state (Fig. 1E). This interaction strength was not accessible for the TiO-TiO dimers (29). Decreasing the spacing of the atoms in the Ti$_O$-Ti$_B$ dimer to $r = 0.72$ nm (Fig. 1C) results in three ESR peaks in our measurement range (5 to 30 GHz). In addition to the two triplet-triplet transitions ($f_{T_0T}$ and $f_{T_0T}$), the singlet-triplet (S-T$_0$) transition is now visible as resonance at $f_{ST} \approx 29$ GHz. Here, the resonance frequency $f_{ST}$ directly gives the value of $J$ when the detuning (see below) is negligible.

While a traditional ESR measurement applies a radio frequency (RF) magnetic field, in the ESR-STM technique used here, the RF magnetic field at the Ti atom arises from the modulation of the atom’s position (32) in the nonuniform magnetic field $B_{tip}$ (29, 33) generated by the STM tip. This enables the ESR transition of $\Delta m = 0, \pm 1$. The triplet-triplet transitions in the dimer are driven by a gradient in $B_{tip}$, the component perpendicular to $B_{ext}$. This is the same component required for driving the $|0\rangle$ to $|1\rangle$ transition for an individual Ti atom ($\Delta m = \pm 1$; section S6). In contrast, driving the S-T$_0$ transition ($\Delta m = 0$) requires a gradient in $B_{tip}$, the component of $B_{tip}$ parallel to $B_{ext}$ (12). In this work, we chose a tip having both spatial components of $B_{tip}$ which therefore can drive the transitions of $\Delta m = 0, \pm 1$ in the dimer.

**Heisenberg exchange coupled spin-1/2 Ti atoms**

From the ESR peak splitting, we determined the magnetic interaction energy $J$ for 30 dimers with different separations and orientations (section S3). To exclude the effects of $B_{tip}$ on the resonance frequencies, we measured the ESR spectra as a function of $B_{tip}$ and determined the value of $J$ from the fit based on the spin Hamiltonian (section S4). The measured values of $J$ are given in Fig. 2A as a function of atomic separations ($r$, ranging from 0.72 to 1.3 nm). We find that for atomic distances of less than 1 nm, the Ti$_O$-Ti$_B$ dimers are dominantly coupled by the Heisenberg exchange interaction $J S_1 S_2$, where $S_1$ and $S_2$ are the spin operators. Moreover, the interaction is found to be isotropic (fig. S3).

The exchange interaction generally shows exponential dependence on the separation between spins (34). Given the isotropic interaction energy $J = J_0 \exp[-(r - r_0)/d]$ (34) and taking $r_0 = 0.72$ nm, we obtain for Ti$_O$-Ti$_O$ dimers a decay constant $d = 64.6 \pm 4.9$ pm and a prefactor $J_0 = 28.9 \pm 1.3$ GHz. The decay constant matches well with reported values for exchange interactions across a vacuum gap (29, 33, 35). For Ti$_O$-Ti$_B$ and Ti$_B$-Ti$_B$ dimers, we obtain $d = 40.0 \pm 2.0$ pm (29) and $94.0 \pm 0.3$ pm (fig. S3D), respectively. This difference in decay constant between the dimer types indicates the sensitivity of the exchange interaction to either the orbitals being involved in the interaction or the spatial distribution of spin density (36), resulting from the different interaction potentials (34) and the different magnetic ground states (29). As determined from the intensity of peaks in the ESR spectra (fig. S3) (27, 29), $J_0$ is positive, and thus, the coupling between Ti atom spins is antiferromagnetic.

**Energy detuning of superposition states**

While the nonuniform magnetic field arising from the STM tip is necessary to drive the spin resonance, this tip magnetic field also provides a means to control the quantum states by applying a local...
magnetic field to one atom in the dimer. The eigenstates deviate from ideal singlet and triplet states because of an energy detuning \( \varepsilon \), which is the difference in Zeeman energies between the two atoms. The detuning arises from two sources: (i) the tip magnetic field that is applied only on one of the atoms (29) and (ii) a slight difference in the gyromagnetic ratios \( \gamma_1 \) and \( \gamma_2 \) for the two atoms at different binding sites (fig. S2). The Hamiltonian (in units of angular frequency) describing the two spins dominantly coupled by the exchange interaction is then given by

\[
H = \gamma_1 S_1 \cdot (B_{\text{ext}} + B_{\text{tip}}) + \gamma_2 S_2 \cdot B_{\text{ext}} + 2\pi J S_1 \cdot S_2
\]  

Under the approximation that \( B_{\text{tip}} \) is parallel to \( B_{\text{ext}} \), the energy detuning is given by \( \varepsilon = |(\gamma_1 - \gamma_2)B_{\text{ext}} + \gamma_1 B_{\text{tip}}|/2\pi \) (29), resulting in the quantum eigenstates

\[
|T_0(\xi)\rangle = \sin \xi |01\rangle + \cos \xi |10\rangle
\]

\[
|S(\xi)\rangle = \cos \xi |01\rangle - \sin \xi |10\rangle
\]

where \( \xi \) is a mixing parameter given by \( \tan \xi = J/\varepsilon \). When the energy detuning is negligible \( (J \gg \varepsilon) \), the eigenstates are the ideal singlet and triplet states: \( |S\rangle = (|01\rangle - |10\rangle)/\sqrt{2} \) and \( |T_0\rangle = (|01\rangle + |10\rangle)/\sqrt{2} \). In contrast, increasing the energy detuning leads to the Zeeman product states: \( |01\rangle \) and \( |10\rangle \).

The effect of this energy detuning on the eigenstates is a shift of their energy levels, which results in a corresponding ESR frequency shift of the \( S\text{–}T_0 \) transition \( (\Delta f_{\text{ST}0}) \) from the minimum value of \( f_{\text{ST}0} \). Figure 2B shows the measured \( \Delta f_{\text{ST}0} \) for TiO-TiB dimers as a function of \( B_{\text{tip}} \) for different values of \( J \). The minimum in \( f_{\text{ST}0} \) is reached at \( B_{\text{tip}} = 38 \pm 12 \text{ mT} \) (Fig. 2C), where the transition constitutes a magnetic field–independent clock transition to first order. At this field, the detuning is absent, that is, \( \varepsilon = 0 \), because the tip field fully compensates for the subtle difference in magnetic moments of the TiO and TiB atoms (fig. S2).

We calculated the eigenvalues and eigenstates (fig. S4) using the Hamiltonian in Eq. 1 to fit the experimental results. When \( B_{\text{tip}} \) is parallel to \( B_{\text{ext}} \), the singlet-triplet energy detuning is given by \( \Delta f_{\text{ST}0} = \sqrt{f^2 + \varepsilon^2} - J \). However, it is necessary to account for the tilting of \( B_{\text{tip}} \) with respect to \( B_{\text{ext}} \) to obtain adequate fits to the ESR frequencies for all allowed transitions in each dimer of Fig. 2. For the STM tips used in Fig. 2, we find that the angles between \( B_{\text{tip}} \) and \( B_{\text{ext}} \) fall in the range of 21° to 51° depending on the tip apex (section S4).

For weakly coupled dimers \( (J < 1 \text{ GHz}) \), a slight increase of energy detuning due to \( B_{\text{tip}} \), shifts \( f_{\text{ST}0} \) considerably from its minimum value. Using the model Hamiltonian of Eq. 1, at the typical tip field of 110 mT, the eigenstates of the dimer with \( J = 0.8 \text{ GHz} \) are \( |S(\xi)\rangle = 0.92|01\rangle - 0.39|10\rangle \) and \( |T_0(\xi)\rangle = 0.39|01\rangle + 0.92|10\rangle \), which more closely resemble the Zeeman product states.

We find that the effects of Zeeman energy detuning \( (\varepsilon) \) on \( f_{\text{ST}0} \) can be regulated by the coupling strength of two atoms. As shown in Fig. 2C, increasing \( J \) to 30 GHz \( (J \gg \varepsilon) \) markedly reduces the sensitivity of \( f_{\text{ST}0} \) on the magnetic field variation. At the same tip field \( (B_{\text{tip}} = 110 \text{ mT}) \), the eigenstates now remain almost in the ideal singlet and triplet states \( (|S(\xi)\rangle = 0.71|01\rangle - 0.70|10\rangle \) and \( |T_0(\xi)\rangle = 0.70|01\rangle + 0.71|10\rangle \)). Thus, in the following, we ensure that \( J \gg \varepsilon \) by keeping \( B_{\text{tip}} \) small \( (< 150 \text{ mT}) \) and by using large \( J \) (30 GHz) and show that this choice results in a decoherence-free subspace.

Enhanced spin coherence using magnetic field–independent states

On the basis of the results from the previous sections, we now focus on the spin coherence times of strongly coupled TiO-TiB dimers \( (r = 0.72 \text{ nm}, J \approx 30 \text{ GHz}) \). The spin coherence for the singlet-triplet transition and its sensitivity to the external and local magnetic fields are compared in Fig. 3 with (i) the triplet-triplet transition of the same dimer and (ii) the \( |0\rangle \) to \( |1\rangle \) transition of an individual TiO atom. We obtained the spin coherence time of each transition by fitting the
ESR linewidth $\Gamma$ to the Bloch equation model (26) (section S5), as a function of RF voltage ($V_{RF}$) (Fig. 3A). In the limit of small $V_{RF}$, the coherence time is given by $1/\Gamma$. This coherence time includes the effect of inhomogeneous line broadening and is designated $T_2^*$ to distinguish it from the intrinsic spin coherence time $T_2$. In our single-spin experiment, inhomogeneous broadening may be due to any time-varying magnetic fields that are present to give temporal ensemble broadening (37).

For typical ESR conditions and $B_{tip} = 110$ mT, we find $T_2^* = 99.0 \pm 9.7$ ns for the S-T0 transition (Fig. 3A). Under the same conditions, $T_2^*$ of the triplet-triplet ($T_0^-T_0^+$) transition and of the individual TiO is ~8 and ~20 times smaller, at only $13.0 \pm 0.3$ ns and $5.3 \pm 0.7$ ns, respectively.

The spin coherence time is closely related to the sensitivity of spin states to the time-varying external and local magnetic fields. The sensitivity to uniform external magnetic fields was characterized by varying the external field magnitude $B_{ext}$ from 0.5 to 1.1 T (Fig. 3B). ESR frequencies $f_{T_0^-T_0^+}$ and $f_{T_0^-T_0^+}$ ($\Delta m = \pm 1$ transition) shift linearly with $B_{ext}$ due to the Zeeman effect on the states $|T_0^-\rangle$ and $|T_0^+\rangle$. In contrast, $f_{ST_0}$ ($\Delta m = 0$ transition) shows no Zeeman shift and remains nearly independent of $B_{ext}$, an essential property of a clock transition (7–9).

We now investigate the effect of a local magnetic field by varying $B_{tip}$ over a large range, extending from 10 mT to 0.45 T (Fig. 3C). For the transitions between triplet states, the resonance frequencies $f_{T_0^-T_0^+}$ and $f_{T_0^-T_0^+}$ again increase steadily by the Zeeman energy owing to $B_{tip}$ applied to one atom in the dimer. In contrast, $f_{ST_0}$ remains essentially constant when $B_{tip}$ is lower than ~150 mT. The results in Fig. 3 show that the singlet-triplet transition is insensitive to both external and local magnetic field fluctuations, which results in the measured increase of its spin coherence time.

**Homodyne detection as a means to spin decoherence reduction**

In addition to magnetic field fluctuations, tunneling electrons are a major source of decoherence of the surface atom’s spin in magnetoresistively...
assembling custom spin chains and arrays with enhanced quantum magnetic structures may serve as the smallest component for generating an additional way to improve the spin coherence. As a result, we find that at $V_{\text{DC}} = 0$ is only possible for the S-T0 transition (Fig. 4B, top). For the S-T0 transition, the time-average population of spin states of the atom under the tip does not vary; thus, it cannot be detected by DC conductance changes. However, the magnetization of the atom along the quantization axis oscillates in time during ESR. The oscillating magnetoelectric field at the frequency of the driving voltage $V_{\text{RF}}$ is multiplied by $V_{\text{RF}}$ to produce a DC tunnel current, which can thus be detected. This rectification is known as a homodyne detection (for a full description of the ESR line shape, see section S5) (30, 31). Thus, in the case of the S-T0 transition, both driving and sensing the spin resonance signal can be achieved by using $V_{\text{RF}}$ only, enabling us to set $V_{\text{DC}}$ to zero. In Fig. 4B, we find that, for the S-T0 resonance signal, the peak width is narrower for lower $V_{\text{DC}}$. As a result, we find that at $V_{\text{DC}} = 0$, the ESR signal of the S-T0 transition is the sharpest because the tunneling current due to $V_{\text{DC}}$ is absent.

As seen in Fig. 4C, the coherence times $T_2^*$ for all transitions observed increase rapidly with decreasing $V_{\text{DC}}$. Since nearly every tunneling electron induces decoherence of the surface spin (22), reducing the number of tunneling electrons improves the spin coherence significantly. At $V_{\text{DC}} = 0$, we obtain $T_2^* = 257 \pm 80$ ns for the S-T0 transition. Note that the ESR measurement at $V_{\text{DC}} = 0$ is only possible for the S-T0 transition (Fig. 4B). Although we set $V_{\text{DC}}$ to zero, the remaining tunneling current generated by $V_{\text{RF}}$, the finite temperature (22), and the relatively short spin relaxation time $T_1$ (29, 38) limit the spin coherence time of the S-T0 transition, resulting in the deviation of $T_2^*$ from the reciprocal curve in Fig. 4C.

**DISCUSSION**

By controlling the magnetic coupling between electron spins of two atoms, we have demonstrated robust singlet and triplet states and achieved a significantly enhanced spin coherence time. Both driving and sensing the singlet-triplet transition do not require a DC voltage, providing an additional way to improve the spin coherence. As a result, we achieved a large improvement of spin coherence by a factor of about 10 compared with the triplet-triplet transition in the same dimer. Moreover, this exceeds the spin coherence time previously determined for individual Fe atoms (26), despite the much shorter spin relaxation time $T_1$ for individual Ti atoms (29). These engineered atomic-scale magnetic structures may serve as the smallest component for assembling custom spin chains and arrays with enhanced quantum coherence times. The ability of ESR-STM to construct desired multispin systems and to electrically access their many-body states might enable the exploration of quantum phases, spintronic information processing, and quantum simulation.

**MATERIALS AND METHODS**

Experiments were performed using a home-built STM system at the IBM Almaden Research Center. We evaporated Ti atoms onto a cold (<10 K) 2 ML MgO grown on Ag(001). The MgO layer was used to decouple the spin of Ti atoms from the underlying substrate electrons (39). Previous works showed that the Ti atoms are likely hydrogenated because of residual hydrogen gas in the vacuum chamber (29, 40), and here, we denote the hydrogenated Ti atoms simply as Ti. An external magnetic field ($B_{\text{ext}}$) was applied nearly in-plane. An RF voltage $V_{\text{RF}}$ was applied across the tunnel junction for driving spin resonance, and a DC bias voltage $V_{\text{DC}}$ was applied for the DC magnetoelastic sensing of the spin states (Fig. 1A) (26).

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/11/eaau4159/DC1

Section S1. Binding site of Ti atoms on 2 ML MgO/Ag(001)
Section S2. ESR of individual Ti atoms
Section S3. Magnetic interaction of Ti dimers
Section S4. Spin Hamiltonian of two coupled Ti spins
Section S5. Detection mechanism of ESR
Section S6. Driving mechanism of ESR

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