Utilizing a single atom magnet and oscillating electric fields to coherently drive magnetic resonance in single atoms

Authors:
Soo-hyon Phark1,2,3†, Hong T. Bui1,4†, Alejandro Ferrón5, Joaquin Fernández-Rossier6, Jose Reina-Gálvez1,2, Christoph Wolf1,2, Yu Wang1,2, Kai Yang3,7, Andreas J. Heinrich1,4*, Christopher P. Lutz3*

Affiliations:
1Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Korea
2Ewha Womans University, Seoul 03760, Korea
3IBM Research Division, Almaden Research Center, San Jose, CA 95120, USA
4Department of Physics, Ewha Womans University, Seoul 03760, Korea
5Instituto de Modelado e Innovación Tecnológica (CONICET-UNNE) and Facultad de Ciencias Exactas, Naturales y Agrimensura, Universidad Nacional del Nordeste, Avenida Libertad 5400, W3404AAS Corrientes, Argentina
6QuantaLab, International Iberian Nanotechnology Laboratory (INL), 4715-330 Braga, Portugal
7Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing, China.

*Corresponding authors. Email: heinrich.andreas@qns.science, cplutz@us.ibm.com
†These authors contributed equally to this work.

Abstract: Scanning tunneling microscopes (STM) equipped with pulsed electron spin resonance (ESR) have paved a way to coherently control individual atomic and molecular spins on surfaces. A recent breakthrough was to drive ESR of a spin outside the tunnel junction by locating a single atom magnet in proximity to a qubit, composing a 'spin-magnet pair'. Here we present a combined experimental and model study on the ESR driving mechanism in such a spin-magnet pair. Pulsed ESR of a single hydrogenated Ti atom on MgO with an Fe atom located between 6 and 8 Å away showed a non-vanishing Rabi rate even when the tip is substantially retracted, comparable in strength with that driven by the interaction with the tip's magnetic moment under normal tunnel conditions. We reveal that this ESR driving field is contributed by Fe through the spin-spin interaction in the pair and show its tunability using a vector magnetic field. The spin-magnet pair therefore expands ESR-STM to address and coherently control on-surface atomic and molecular spins independent of the tip's magnetic apex. Together with existing atom manipulation techniques in STM, our study establishes a feasible method to design spin-based multi-qubit systems on surfaces.

One sentence summary: We coherently drive electron spin resonance of on-surface spins using a local magnetic field from nearby single atom magnets coupled with radio-frequency electric fields, enabling the use of ESR-STM for a wide range of surface spins.
INTRODUCTION:

Atomic and molecular spins on surfaces can provide a solid state qubit platform [Heinrich2021, Thiele2014] with unique bottom-up design capabilities. The coherent manipulations of single spins in a scanning tunneling microscope (STM) equipped with pulsed electron spin resonance (ESR) opened a way to utilize individual atomic [Yang2019] and molecular [Willke2021] spins on surfaces as feasible qubits for quantum information and computation. To date, magnetic resonance in STM [Baumann2015, Seifert2020, Stein2021, Veldman2021, KimJK2021] has relied on the electric-field driven modulation of the atomic scale magnetic interaction between the spin on the surface and the STM tip [Lado2017, Yang2019prl, Galvez2019]. A coherence time of a few tens of ns has been reported for the spins in the tunnel junction [Yang2019, Willke2018, Willke2021]. This coherence is mainly limited due to the scattering of the spin states by tunneling electrons [Willke2018, Willke2021], limiting the number of available quantum gate operations within the coherence time. Moreover, such a 'tip-field-driven ESR' allows only one spin in the tunnel junction to be coherently controlled at a time, which has so far prohibited quantum manipulation of multiple spin qubits in STM. Therefore, developing a new ESR driving mechanism of a spin in STM, free from the tip and thus from the measurement schemes confined in the tunnel junction, is required. Controlling spin-spin couplings with Angstrom-scale precision has recently enabled the precise and simultaneous quantum-coherent control of multiple qubits [Wang2022].

In this work, we present continuous wave (CW) and pulsed ESR of hydrogenated Ti atoms ($S = 1/2$) on MgO [Yang2017], with Fe atoms located in close proximity, in an ultra-high vacuum STM operating at 1 K. Pulsed ESR of Ti atoms in such atomic pairs showed a non-vanishing Rabi rate up to $\Omega/2\pi \sim 25$ MHz even when the tip-Ti distance was large, comparable with that driven by the interaction with the tip magnetic moment [Yang2019, Willke2021]. CW-ESR on the Ti-Fe pair of a separation of 0.6 – 0.8 nm in a vector magnetic field revealed that the spin-spin interaction ranges up to 20 GHz and the driving strength is tunable as well. Combined with a model study, we show that such a tip-independent driving field is contributed by the Fe spin due to its quasi-static spin states [Paul2017, Willke2019]. A single atom magnet, such as Fe on MgO, provides an
inhomogeneous local field, which is able to replace the role of the tip in the conventional ESR scheme in a STM [Lado2017,Yang2019prl, Galvez2019, Ferron2019, Seifert2020, KimJK2021].

**EXPERIMENTAL RESULTS:**

Our ESR experiment in STM on a pair of Ti and Fe atoms, hereafter referred to as 'Ti-Fe pair', is illustrated in Fig. 1A. Here both the tip and the Fe are coupled with the Ti spin via exchange interactions $J_{\text{Ti-tip}}$ and $J_{\text{TiFe}}$, respectively. The RF bias voltage $V_{\text{RF}}$ applied between the tip and substrate can drive a spin resonance of the Ti, which is measured by the spin-polarized tunneling current [Baumann2015] (Materials & Methods). By utilizing established atom manipulation techniques [Eigler1990], we constructed such Ti-Fe pairs of three different separations (Figs. 1B-D) and performed ESR on the Ti atom of each pair as shown in Fig. 1E. The ESR spectrum from the pair with the largest separation in Fig. 1B (black curve) showed only one resonance peak, determined by the Zeeman energy of an isolated Ti [Seifert2020, KimJK2021], which indicates a vanishingly small interaction with Fe.

On the other hand, Ti-Fe pairs of smaller separations (Figs. 1C and D) showed a clear splitting ($\Delta f$) of resonance peaks (red and blue curves in Fig. 1E). This splitting increased for the pair with the smaller separation (Fig. S1), revealing a sizable spin-spin interaction between Ti and Fe in the pair. Our model study, presented below, suggests that the spin of Fe in such a Ti-Fe pair remains quasi-static for sufficiently long times in either spin up ($\uparrow; m_{\text{Fe}} = +2$) or down ($\downarrow; m_{\text{Fe}} = -2$) states (see MODEL STUDY section; Fig. S4). This is similar to isolated Fe atoms on MgO, which have a large uniaxial magnetic anisotropy energy (MAE) [Baumann2015prl] and a spin relaxation time of a few tens of microseconds at the magnetic fields used here [Paul2017, Willke2019]. Henceforth in this work, we treat the Fe in the pair as a single atom magnet with two long-lived magnetic states that are aligned perpendicular to the surface, resulting in two peaks in each ESR spectrum on the Ti in the pairs. Besides the splitting, we also observed significant shifts of both resonance peaks on the pairs by up to a few GHz, as indicated by $f_c$. This can be understood as a change of the direction of the Ti spin stemming from the Ti-Fe interaction, which increases its Zeeman energy due to the anisotropy of its $g$-factor [KimJK2021] (Fig. S6).
Similar to the role of the exchange field from the tip-Ti interaction in ESR of an isolated single spin [Lado2017,Yang2019prl,Galvez2019], the Ti-Fe interaction can also provide an effective local magnetic field at the position of Ti. This may effectively convert the applied RF electric field into an oscillating transverse driving field, which is able to drive the spin resonance of Ti. To verify this, we performed pulsed ESR on the three Ti atoms in Fig. 1 at $V_{RF} = 100$ mV (Fig. S7) and measured the Rabi rate ($\Omega$) as a function of tunnel conductance ($\sigma_{\text{tun}}$), i.e. the tip-Ti distance, as shown in Fig. 2. At a large tunnel conductance, the measured Rabi rates were comparable for all three cases (Fig. 2A). However, we found a considerable difference of Rabi rates at a small tunnel conductance (Fig. 2B). The Rabi rate of the isolated Ti showed a linear decrease for a decreasing tunnel conductance (gray in Fig. 2C), intersecting around $\Omega \sim 0$ at zero tunnel conductance, which implies a vanishing driving field at a very large tip-atom distance. This linear behavior indicates an exponential dependence of the driving field on the tip height, a fingerprint for the exponential nature of the driving field generated by the Ti-tip exchange interaction [Yang2019prl].

In contrast, the Rabi rate of Ti close to Fe (Figs. 1C and D) showed a finite Rabi rate, $\Omega_0$, when the tunnel conductance approached to zero (blue and red in Fig. 2C). This strongly suggests that the ESR of Ti was driven effectively by another source, namely the quasi-static Fe spin moment in the pair, at a tip height where the driving field from the tip vanishes. Furthermore, we found that the zero conductance Rabi rate measured on the pair with a separation of 0.72 nm was smaller than that from the pair with 0.59 nm. The Rabi rate saturates at $\Omega_0/2\pi = 25.0 \pm 6.4$ MHz for the closer pair (Figs. S8–S10), which is sufficient to coherently invert the spin in $\sim 20$ ns. We note that this Rabi rate is comparable to that for the isolated Ti when the tip is brought relatively close.

The contribution of Fe to the spin resonance of a Ti atom induced unusual features also in the intensity of CW-ESR spectra. Figure 3A shows an evolution of ESR spectra as a function of tunnel conductance obtained from the pair with a separation 0.72 nm (Fig. 1C). The peak $f_\uparrow$ showed a monotonic decrease of its ESR intensity with decreasing conductance, reflecting a decreasing driving field from the tip spin as the tip was moved away from the surface, however, not completely vanishing as the tunnel conductance approached 0. In contrast, the ESR intensity of peak $f_\downarrow$ vanished at an intermediate tunnel conductance of about 0.15 nS and reappeared as the tip was withdrawn further from the atom. This behavior
differs markedly from the ESR intensity of isolated Ti atoms (Fig. S11) or Ti-Ti pairs [Yang2017, Bae2018], where the peak intensities decrease monotonically and vanish at very low conductance. As a consequence, the intensity ratio of the two peaks in a Ti-Fe pair strongly deviates from that of a Ti-Ti pair, where it is mostly determined by the thermal occupations of its four spin states.

The features in the ESR spectra of the Ti-Fe pair can be qualitatively understood as a vector sum of two driving fields, one stemming from the interaction with the tip and the other with the spin of the Fe single atom magnet. This is illustrated in the three insets in Fig. 3B. If we assume that the two driving fields are antiparallel, they can completely cancel each other at a critical tip height, which clarifies the disappearance of the peak for $f_\downarrow$ at about 0.15 nS. On the other hand, the opposite orientation of the Fe spin state leads to a parallel alignment of the two driving fields, which explains the monotonic decrease but the non-vanishing peak height for $f_\uparrow$ as the tunnel conductance approaches 0.

Our measurements have shown that the spin-spin interaction in a Ti-Fe pair is the origin of the peak splitting in the ESR spectra as well as the finite Rabi rates at zero tunnel conductance. The quasi-static nature of the Fe spin, which is always aligned perpendicular to the surface, suggests that the Ti-Fe interaction $J_{\text{TiFe}} S_{\text{Ti}} \cdot S_{\text{Fe}}$ only depends on the direction of the Ti spin, which is controllable by the direction of the external magnetic field. To characterize the Ti-Fe interaction, we therefore measured the dependence of the peak splitting in the ESR spectra of the two pairs on the field angle (Figs. 1C and D) (Fig. S12). Figures 4A and B show the resonance peak frequencies and peak splitting, respectively, measured as a function of the angle between the magnetic field and the surface normal ($\theta_{\text{ext}}$).

The splitting in both pairs reaches a maximum when the field is normal to the surface ($\theta_{\text{ext}} = 0$) and vanishes for in-plane fields ($\theta_{\text{ext}} = +90^\circ$ or $-90^\circ$), confirming that the Fe spin is always pointing along the surface normal. Of more interest is the fact that it is mirror symmetric about the field angle of an in-plane direction, where the splitting is 0. The peak splitting ($\Delta f$) from an exchange type Ti-Fe interaction in general will show a cosine-dependence on the angle between two spins ($\eta$; see Fig. 4C). However, note that both Ti and Fe spins flip simultaneously when $B_{\text{ext}}$ rotates across an in-plane direction (Fig. 4D), leading to peak splitting $\Delta f \propto |\cos \eta|$. This excellently explains the features in the field angle dependence of the splitting in Fig. 4B and suggests that the peak splitting is originated only
from the Ti-Fe interaction. Fitting the data in Fig. 4B resulted in exchange couplings $J_{\text{TiFe}} = 1.1 \pm 0.1$ and $6.8 \pm 0.5$ GHz for the pairs of 0.72 nm and 0.59 nm, respectively. We note that the resonance frequencies showed a periodic evolution with a change in field angle by 180° due to bi-directional nature of the magnetic moment of the tip [KimJK2021] (Fig. S13). In addition, the asymmetries of the resonance frequencies about the field angles close to the in-plane directions (+90° and −90°) were mainly contributed by the tip-induced magnetic field (see MODEL STUDY section).

A single atom magnet can indeed provide an atomic-scale and reliable local transverse field, which can be used to coherently drive ESR of a nearby spin. To enhance the driving strength, a straightforward strategy is positioning more single atom magnets in proximity. We constructed a Fe-Ti-Fe structure by adding one more Fe atom near a Ti-Fe pair of 0.72 nm, as shown in Fig. 5A, and performed ESR measurements on the Ti atom. The ESR spectrum from this spin complex showed four resonance peaks (Fig. 5B), corresponding to four spin orientations of two non-interacting Fe spins, $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, and $|\downarrow\downarrow\rangle$ (Fig. S5). To figure out the effect of two Fe atoms on the ESR of the Ti spin, we performed pulsed ESR on this complex at the resonance peak $f_3$ (Fig. S15). Figure 5C shows the Rabi rate as a function of the tunnel conductance (purple) as well as that measured on a Ti-Fe pair with the same tip (red). This comparison directly reveals an enhancement of the Fe-induced driving field by a factor of about two, indicating that our atomic-scale and bottom-up approach is plausible to tailor magnetic resonances in artificial atomic structures.

**MODEL STUDIES & DISCUSSION:**

To survey spin eigenstates of a Ti-Fe pair, we set up a Hamiltonian of the spin pairs

$$\hat{H}_{\text{pair}} = D_{\text{Fe}} S_T^2 + C_{\text{Fe}} (S_T^4 + S_T^2) + J_{\text{TiFe}} S_T \cdot S_{\text{Fe}} - g_{\text{Ti}} \mu_B S_T \cdot B_{\text{ext}} - g_{\text{Fe}} \mu_B S_{\text{Fe}} \cdot B_{\text{ext}} \quad (1),$$

composed of out-of-plane MAE of Fe $D_{\text{Fe}} = -4.7$ meV [Baumann2015], four-fold in-plane anisotropy of Fe $C_{\text{Fe}} = 41$ neV [Paul2017], Zeeman terms for Ti and Fe spins with $B_{\text{ext},x} = 0.891$ T and $B_{\text{ext},z} = 0.125$ T, and Ti-Fe exchange term with the coupling $J_{\text{TiFe}}$ ranging 0 – 80 $\mu$eV, which corresponds to the peak splitting of 0 – 20 GHz observed from the ESR spectra (see Fig. 4). $g_{\text{Ti}}$ and $\mu_B$ are the g-factor of the Ti atom and Bohr magneton, respectively. The
Hamiltonian turns out to be almost diagonal in the basis set $|m_{\text{Ti}}m_{\text{Fe}}\rangle$, where $m_{\text{Ti}}$ and $m_{\text{Fe}}$ are magnetic quantum numbers of Ti and Fe spins, respectively (see Fig. 1F). By diagonalizing the Hamiltonian, we found the four lowest eigenstates of the pair to be linear combinations of $|+\ 1/2, +2\rangle, |-\ 1/2, +2\rangle, |+\ 1/2, -2\rangle$, and $|-\ 1/2, -2\rangle$, whose eigenenergies are lower by $\sim 13$ meV than the other 6 eigenstates (Fig. S4). Together with the long spin relaxation time [Paul2017, Willke2019], this guaranties to drop off the MAE and four-fold in-plane anisotropy terms from the Hamiltonian, such that the Fe spin in the ESR of the Ti spin is treated as an Ising spin. The ESR peak originated from the resonance of the Ti spin between its two eigenstates of $m_{\text{Ti}}= +1/2$ (+) and $-1/2$ (−) [Yang2017], thus we attribute the two peaks of each spectrum from the pairs (Fig. 1E) to the resonances of Ti spin corresponding to the two distinct quasi-static Fe spin states, $|\uparrow\rangle$ and $|\downarrow\rangle$.

For the influence of the Ti-Fe interaction on the Ti spin, we consider a simple picture that the Fe spin contributes an additional field $B_{\text{Fe}}$ at the Ti position, as sketched in Fig. 4C. The Ti-Fe interaction $J_{\text{TiFe}}S_{\text{Ti}} \cdot S_{\text{Fe}}$ provides an additional Zeeman energy to the Ti spin by the exchange field generated from the Fe

$$B_{\text{Fe}} = (J_{\text{TiFe}}/\mu_B) g_{\text{Ti}}^{-1} \cdot S_{\text{Fe}}$$

where $g_{\text{Ti}}$ is the g-tensor of the Ti spin. Considering the Fe spin as a classical magnetic dipole pointing $\pm \hat{z}$ direction, we take an approximation of $S_{\text{Fe}} = \langle S_{\text{Fe},z}\rangle$. This simplifies the equation (2) such that the Fe-induced field is $B_{\text{Fe}} = \pm J_{\text{TiFe}}/\mu_B g_{\text{Ti},z} \hat{z}$, as denoted in Fig. 4C by $B_{\text{Fe},\uparrow}$ and $B_{\text{Fe},\downarrow}$ for the $|\uparrow\rangle$ and $|\downarrow\rangle$ states, respectively. Hence, the Ti spin is aligned to the net field $B_{\text{net}} = B_{\text{ext}} + B_{\text{Fe}}$, and the two sets of energy eigenvalues for $B_{\text{net},\uparrow}$ and $B_{\text{net},\downarrow}$ determine two resonant frequencies $f_{\uparrow}$ and $f_{\downarrow}$ in each spectrum, with a peak splitting $\Delta f = f_{\uparrow} - f_{\downarrow}$. For a coupling strength $J_{\text{TiFe}}$ given by the Ti-Fe separation, the Ti-Fe interaction will show a cosine-dependence on the angle between two spins, $\eta = \theta_{\text{ext}} - \alpha$, where $\alpha$ is the angle formed by the exchange field contributed from Fe. Simultaneous flipping of both Ti and Fe spins across field angle of an in-plane direction is responsible for the mirror symmetric peak splitting (Fig. 4B) and formation of a dead-angle $2\alpha_0$ as well (see Fig. 4D).

All the experimental data presented in Figs. 1–4 point a fact that a single atom magnet is able to drive ESR of a single atom spin nearby. However, a rigorous understanding
on the spin system in this work requires an extended model, counting the contribution from the tip as well. First, to have a quantitative insight on the spin interactions, we set up a time-independent Hamiltonian $\tilde{H}_0$ by adding Ti-tip interaction to the equation (1)

$$\tilde{H}_0 = -g_\text{Ti}\mu_\text{B}S_{\text{Ti}} \cdot B_{\text{ext}} + J_{\text{TiFe}} S_{\text{Ti}} \cdot S_{\text{Fe}} + J_{\text{til}} S_{\text{Ti}} \cdot S_{\text{tip}}$$

where $J_{\text{Ti,tip}}$ represents the exchange coupling between the spins of the Ti and tip. Following the discussion on the eigenstates of the pair, we exclude the Zeeman and anisotropy terms for the Fe and tip spins. Due to the bi-directional nature of the tip magnetic moment [KimJK2021], like the Fe spin, here we treat the tip spin as a classical magnetic moment of uniaxial anisotropy with two expectation values $\pm |\langle S_{\text{tip}} \rangle|$ along a direction

$$n_{\text{tip}} = (n_x, n_y, n_z) = (\sin \theta_{\text{tip}} \cos \phi_{\text{tip}}, \sin \theta_{\text{tip}} \sin \phi_{\text{tip}}, \cos \theta_{\text{tip}})$$

where $\theta_{\text{tip}}$ and $\phi_{\text{tip}}$ are the polar and azimuthal angles of the unit vector $n_{\text{tip}}$ in the Cartesian coordinates. The Hamiltonian reduces to a simple form:

$$\tilde{H}_0 = S_{\text{Ti}} \cdot \tilde{B}_{0,\pm}$$

$$\tilde{B}_{0,\pm} = (-g_x \mu_\text{B} B_x + J_{\text{til}} \langle S_{\text{tip}} \rangle n_x, -g_y \mu_\text{B} B_y + J_{\text{til}} \langle S_{\text{tip}} \rangle n_y, -g_z \mu_\text{B} B_z + J_{\text{til}} \langle S_{\text{tip}} \rangle n_z \pm 2J_{\text{TiFe}})$$

where the $\pm$ sign distinguishes the two quasi-static states of the Fe spin. Diagonalization of $\tilde{H}_0$ served as eight eigenstates in total, leading four available ESR transitions of the Ti spin corresponding to four distinct combinations of the Fe and tip spin states, $\{\n\}_{\text{Fe}} \{\n\}_{\text{tip}}$, $\{\u\}_{\text{Fe}} \{\n\}_{\text{tip}}$, $\{\n\}_{\text{Fe}} \{\u\}_{\text{tip}}$, and $\{\u\}_{\text{Fe}} \{\u\}_{\text{tip}}$. The two resonant frequencies $f_\parallel$ and $f_\perp$ in each spectrum read $|\tilde{B}_{0,+}|$ and $|\tilde{B}_{0,-}|$, respectively, which determines peak splitting $\Delta f = f_\parallel - f_\perp$.

Using the field-angle dependences of the eigenenergies of $\tilde{H}_0$ (Eqn. (5)) and tuning $J_{\text{TiFe}}$, we fit the resonance frequencies and peak splitting in Figs. 4A and B. First, to characterize the tip spin, we utilized field-angle dependences of ESR frequency, intensity, and Rabi rate measured on an isolated Ti atom, which provided the magnetic anisotropy of the tip spin ($\theta_{\text{tip}} = 65^\circ$ and $\phi_{\text{tip}} = 75^\circ$; Fig. S13). Note that a combinatorial fine tuning of $J_{\text{til}}$, $\theta_{\text{tip}}$, and $\phi_{\text{tip}}$ excellently reproduced detailed experimental features, e.g. the asymmetry in the resonance frequencies across $\theta_{\text{ext}} = +90^\circ$ and $-90^\circ$ measured from the pair of 0.72 nm (red markers), which got weaker as the tip moved out from the Ti atom. This is a proof for a
high validity of our model as described in the equations (5) and (6). In contrast, we ignored the Ti-tip interaction in the model for the pair of 0.59 nm (blue markers). Together with a small tunnel conductance ($V_{DC} = 200$ mV, $I_{DC} = 10$ pA) during the ESR of this pair, the much larger Ti-Fe interaction guaranteed a condition of $J_{TiFe} \gg J_{Ti,tip}$ (Fig. S14), leading a negligible contribution of the tip to the ESR of Ti, as supported by a high symmetry observed in the resonance frequencies across $\theta_{ext} = +90^\circ$.

Drawn from the observations shown in both pulsed- and CW-ESR experiments, we model the driving field for the ESR of the Ti spin in the presence of the Fe. In STM, ESR has been driven by radio-frequency electric bias applied between tip and substrate, i.e. a time-varying electric field $E(t)$, which couples with an inhomogeneous local spin-spin interaction such as $J_{Ti,tip}$ between the Ti and tip, generating an effective time-varying magnetic field $B_1(t)$ [Lado2017,Galvez2019]. In the spin system here, by considering the Ti-Fe interaction as well as Ti-tip interaction, the driving terms of the Hamiltonian consists of two time-varying fields, one induced by the tip ($B_{1,tip}$) and the other by Fe ($B_{1,Fe}$), as illustrated in Fig. 3C. The total driving field is $B_1 = B_{1,tip} + B_{1,Fe}$, leading that the Rabi rate $\Omega$ is derived by $\hbar \Omega = g \mu_B \langle + | B_1 \cdot S_{Ti} | - \rangle$ for the ESR transition between two eigenstates of the Ti spin, $| + \rangle$ and $| - \rangle$. Only the components of $B_{1,tip}$ and $B_{1,Fe}$ perpendicular to the total static field $B_0$ will contribute to the transverse time-varying field $B_{1,perp}$, such that the Rabi rate reads

$$\Omega = g \mu_B m_s |B_{1,tip,perp} + B_{1,Fe,perp}|/\hbar \quad (7).$$

The driving field from the tip $B_{1,tip}$ is tunable by the tip-Ti distance while $B_{1,Fe}$ is fixed for a given Ti-Fe separation. Hence, survey on a dependence of ESR amplitude on tunnel conductance will allows us to discriminate the contribution of the Fe from that of the tip.

For a careful selection of tip, one can set the two driving fields to be parallel ($\phi = 0$) or antiparallel ($\phi = 180^\circ$), adding to each other for the $| \uparrow \rangle$ state or canceling each other for the $| \downarrow \rangle$ state of the Fe, respectively. For the antiparallel case, the two contributions completely cancel each other at a critical tip-Ti distance, i.e. critical tunnel conductance which is $\sim 0.15$ nS in Fig. 3. As a tip-Ti distance increases across this critical distance, contribution of Fe ($B_{1,Fe,perp}$) becomes dominant. As a result, the net driving field ($B_{1,perp}$) changes its sign, leading to the opposite Rabi rotation of the Bloch vector with a reappearance of the ESR signal (see
Fig. 3B), which will be a proof on the presence of two competing transverse RF driving fields. With a collinear alignment of the two driving fields, $B_{1,\text{tip} \perp}$ and $B_{1,\text{Fe} \perp}$, the Rabi rate of the Ti spin in Eqn. (7) can be rewritten as a simple sum of two contributions

$$\Omega_\pm = |\Omega_{\text{tip} \pm} \Omega_{\text{Fe}}| \quad (8),$$

where the $+(-)$ sign represents the parallel (antiparallel) case. The contribution of the Rabi rate to the ESR peak height is described by $\Omega^2 T_1 T_2 / (1 + \Omega^2 T_1 T_2)$, where $T_1$ and $T_2$ are the energy relaxation time and dephasing time, respectively [Delgado2017,KimJK2021]. Using a unitless Rabi rate $\Omega'_{\pm} \equiv \Omega_\pm \sqrt{T_1 T_2}$ as fitting parameters, our model excellently reproduced the tunnel conductance dependence of the ESR peak heights (solid curves in Fig. 3B). This specific behavior was observed only with certain tips, providing a collinear alignment to the Fe spin directions, while other tips showed a similar behavior but generally yielded incomplete cancellation (Fig. S16). The finite ESR peak heights at zero conductance demonstrates a sizable driving field by the nearby Fe. The extrapolation of the fit curves to zero conductance reads finite intercepts of a ratio $\sim 0.64$ that is mostly determined by the thermal occupations of the two Fe states at the measurement temperature of 1.2 K.

CONCLUDING REMARKS:

Utilizing a single atom magnet for ESR driving liberates on-surface spin qubits, which have been confined to the tunnel junction in the conventional ESR-STM configuration. Furthermore, this approach enables the separation of driving and detection of ESR, providing longer relaxation and quantum coherence times [Wang2022]. Importantly, it enables atomic and molecular spins on surfaces as a platform of multi qubits operation. In this work, we investigated the electron spin resonance of a single Ti spin on MgO with Fe atoms in close proximity, unraveling the underlying driving mechanism of ESR in such a 'spin-magnet pair'. Together with atom manipulation technique and the availability of further single atom magnets [Donati2016,Natterer2017,Singha2021], our work sheds light on atomic precision design of on-surface spin multi qubit structures with longer relaxation and coherence times.
REFERENCES:

[Bae2018] Y. Bae, K. Yang, P. Willke, T. Choi, A. J. Heinrich, C. P. Lutz, Enhanced quantum coherence in exchange coupled spins via singlet-triplet transitions. *Science Advances* **4**, eaau4159 (2018).

[Baumann2015] S. Baumann, W. Paul, T. Choi, C. P. Lutz, A. Ardavan, A. J. Heinrich, Electron paramagnetic resonance of individual atoms on a surface. *Science* **350**, 417-420 (2015).

[Baumann2015prl] S. Baumann, F. Donati, S. Stepanow, S. Rusponi, W. Paul, S. Gangopadhyay, I. G. Rau, G. E. Pacchioni, L. Gragnaniello, M. Rivetta, J. Dreiser, C. Piamonteze, C. P. Lutz, R. M. Macfarlane, B. A. Jones, P. Gambardella, A. J. Heinrich, and H. Brune, Origin of Perpendicular Magnetic Anisotropy and Large Orbital Moment in Fe Atoms on MgO. *Phys. Rev. Lett.* **115**, 237202 (2015).

[Donati2016] F. Donati, S. Rusponi, S. Stepanow, C. Wäckerlin, A. Singha, L. Persichetti, R. Baltic, K. Diller, F. Patthey, E. Fernandes, J. Dreiser, Ž. Šljivančanin, K. Kummer, C. Nistor, P. Gambardella, H. Brune, Magnetic remanence in single atoms. *Science* **352**, 318–321 (2016).

[Delgado2017] F. Delgado, J. Fernández-Rossier, Spin decoherence of magnetic atoms on surfaces. *Prog. Surf. Sci.* **92**, 40–82 (2017).

[Eigler1990] D. M. Eigler, E. K. Schweizer, Positioning single atoms with a scanning tunnelling microscope. *Nature* **344**, 524–526 (1990).

[Ferron2019] A. Ferrón, S. A. Rodríguez, S. S. Gómez, J. L. Lado, J. Fernández-Rossier, Single spin resonance driven by electric modulation of the g-factor anisotropy. *Phys. Rev. Research* **1**, 033185 (2019).

[Galvez2019] J. Reina Gálvez, C. Wolf, F. Delgado, N. Lorente, Cotunneling mechanism for all-electrical electron spin resonance of single adsorbed atoms. *Physical Review B* **100**, 035411 (2019).

[Heinrich2021] A. J. Heinrich, W. D. Oliver, L. Vandersypen, A. Ardavan, R. Sessoli, D. Loss, A. B. Jayich, J. Fernandez-Rossier, A. Laucht, A. Morello, Quantum-coherent Nanoscience. *Nature Nanotechnology* **16**, 1318–1329 (2021).

[Hwang2022] J. Hwang, D. Krylov, R. Elbertse, S. Yoon, T. Ahn, J. Oh, L. Fang, W. Jang, F. H. Cho, A. J. Heinrich, Y. Bae, Development of a scanning tunneling microscope for variable temperature electron spin resonance. *Rev. Sci. Instrum.* **93**, 093703 (2022).

[KimJK2021] J. K. Kim, W. Jang, H. T. Bui, D. Choi, C. Wolf, F. Delgado, D. Krylov, S. Lee, S. Yoon, C. P. Lutz, A. J. Heinrich, Y. Bae, Spin Resonance Amplitude and Frequency of a Single Atom on a Surface in a Vector Magnetic Field. *Phys. Rev. B* **104**, 174408 (2021).

[Lado2017] J. L. Lado, A. Ferrón, J. Fernández-Rossier, Exchange mechanism for electron paramagnetic resonance of individual adatoms. *Physical Review B* **96**, 205420 (2017).

[Natterer2017] F. D. Natterer, K. Yang, W. Paul, P. Willke, T. Choi, T. Greber, A. J. Heinrich, C. P. Lutz, Reading and writing single-atom magnets. *Nature* **543**, 226-228 (2017).

[Paul2017] W. Paul, K. Yang, S. Baumann, N. Romming, T. Choi, C. P. Lutz, A. J. Heinrich, Control of the millisecond spin lifetime of an electrically probed atom. *Nat. Phys.* **13**, 403–407 (2017).
[Seifert2020] T. Seifert, S. Kovarik, D. M. Juraschek, N. A. Spaldin, P. Gambardella, S. Stepanow, Longitudinal and transverse electron paramagnetic resonance in a scanning tunneling microscope, *Sci. Adv.* 6, eabc5511 (2020).

[Singha2021] A. Singha, P. Willke, T. Bilgeri, X. Zhang, H. Brune, F. Donati, A. J. Heinrich, T. Choi, Engineering atomic-scale magnetic fields by dysprosium single atom magnets. *Nat. Comm.* 12, 1-6 (2021).

[Stein2021] M. Steinbrecher, W. M. J. van Weerdenburg, E. F. Walraven, N. P. E. van Mullekom, J. W. Gerritsen, F. D. Natterer, D. I. Badrtdinov, A. N. Rudenko, V. V. Mazurenko, M. I. Katsnelson, Ad van der Avoird, G. C. Groenenboom, A. A. Khajetoorians, Quantifying the interplay between fine structure and geometry of an individual molecule on a surface. *Phys. Rev. B* 103, 155405 (2021).

[Thiele2014] S. Thiele, F. Balestro, R. Ballou, S. Klyatskaya, M. Ruben, W. Wernsdorfer, Electrically driven nuclear spin resonance in single-molecule magnets. *Science* 344, 1135-1138 (2014).

[Wang2022] Y. Wang, Y. Chen, H. T. Bui, C. Wolf, M. Haze, C. Mier, J. Kim, D.-J. Choi, C. P. Lutz, Y. Bae, A. J. Heinrich, S. Phark, An electron-spin qubit platform crafted atom-by-atom on a surface. *arXiv*:2108.09880 (2022).

[Willke2018] P. Willke, W. Paul, F. D. Natterer, K. Yang, Y. Bae, T. Choi, J. Fernández-Rossier, A. J. Heinrich, C. P. Lutz, Probing quantum coherence in single-atom electron spin resonance, *Sci. Adv.* 4, eaaq1543 (2018).

[Willke2019] P. Willke, A. Singha, X. Zhang, T. Esat, C. P. Lutz, A. J. Heinrich, T. Choi, Tuning single-atom electron spin resonance in a vector-magnetic field. *Nano Lett.* 19, 8201−8206 (2019).

[Willke2021] Philip Willke, Tobias Bilgeri, Xue Zhang, Yu Wang, Christoph Wolf, Herve Aubin, Andreas Heinrich, and Taeyoung Choi, Coherent Spin Control of Single Molecules on a Surface. *ACS Nano* 15, 17959–17965 (2021).

[Yang2017] K. Yang, Y. Bae, W. Paul, F. D. Natterer, P. Willke, J. L. Lado, A. Ferrón, T. Choi, J. Fernández-Rossier, A. J. Heinrich, C. P. Lutz, Engineering the Eigenstates of Coupled Spin-1/2 Atoms on a Surface. *Physical Review Letters* 119, 227206 (2017).

[Yang2019prl] Kai Yang, William Paul, Fabian D. Natterer, Jose L. Lado, Yujeong Bae, Philip Willke, Taeyoung Choi, Alejandro Ferrón, Joaquin Fernández-Rossier, Andreas J. Heinrich, and Christopher P. Lutz, Tuning the Exchange Bias on a Single Atom from 1 mT to 10 T. *Phys. Rev. Lett.* 122, 227203 (2019).

[Yang2019] K. Yang, W. Paul, S. H. Phark, P. Willke, Y. Bae, T. Choi, T. Esat, A. Ardavan, A. J. Heinrich, C. P. Lutz, Coherent spin manipulation of individual atoms on a surface. *Science* 366, 509-512 (2019).

**Author Contributions:** C.P.L., S.P., and A.J.H. conceived the experiment. S.H., H.T.B., Y.W., K.Y., and C.P.L. performed experiments and data analysis. J.R.-G., C.W., and H.T.B. carried out numerical simulations and data fitting. A.F. and J.F-R. developed the analytical model for spin-spin interaction. All authors discussed the results and prepared the manuscript.
Fig. 1: (A) Schematic showing a Ti spin ($S = 1/2$) coupled with Fe on MgO in electron spin resonance (ESR). $J_{TiFe}$ and $J_{Ti\text{tip}}$ are Ti-Fe and Ti-tip spin-spin interactions, which drive ESR of the Ti spin when coupled with the RF voltage $V_{RF}$ in the tunnel junction. (B-D) STM images of Ti atoms with Fe positioned at distances of 1.9 nm, 0.72 nm, and 0.59 nm away from Ti, respectively ($V_{DC} = 50$ mV, $I_{tun} = 10$ pA). (E) Continuous wave ESR spectra measured on Ti atoms in (B) (black), (C) (red), and (D) (blue) at $B_{ext} = 0.9$ T with the polar angle $\theta_{ext} = 82^\circ$ ($V_{DC} = 50$ mV, $V_{RF} = 20$ mV, $I_{tun} = 15$ pA, $T = 1.2$ K). The frequency $f_c$ indicates the center of the two peaks in each curve of the pairs. (F) Checkerboard representation of a spin Hamiltonian for a Ti-Fe pair in the basis set $|m_{Fe}\rangle$ for the $|+\rangle$ state of Ti. Note that the Hamiltonian is almost diagonal, and the energies of the two states $|+\rangle|+2\rangle$ and $|+\rangle|-2\rangle$ are lower by $\geq 10$ meV compared to the other states.
Fig. 2: (A and B) Rabi oscillations using pulsed ESR on Ti atoms shown in Figs. 1(B)–(D) at tunnel conductance of (A) 0.5 nS and (B) 0.15 nS, at the RF voltage $V_{RF} = 100$ mV. Oscillation period of each curve is marked by the vertical bar. The curves are successively shifted vertically by 40 fA for clarity. (C) Rabi rate $\Omega$ extracted from Rabi oscillation measurements of isolated Ti (gray circles), Ti-Fe of 0.72 nm (red crosses), and Ti-Fe of 0.59 nm (blue triangles) ($I_{tun} = 10$ pA, $V_{RF} = 100$ mV, $T = 1.2$ K, $B_{ext} = 0.9$ T, $\theta_{ext} = 82^\circ$). The dotted circles denote the data points corresponding to the curves in (A) and (B) with the same color codes. Two insets illustrate the magnetic interactions, $J_{TiFe}$ and $J_{Ti,tip}$, for small (left) and large (right) tunnel conductance regimes, respectively. Solid curves are fits using the model described in the Supplementary 5, resulting in zero conductance Rabi rates $\Omega_0 / 2\pi$ of $27 \pm 2$ and $14 \pm 2$ MHz for the pairs of 0.59 and 0.72 nm, respectively.
Fig. 3: (A) cw-ESR spectra measured on the Ti-Fe of 0.72 nm at frequency ranges across the peaks $f_\uparrow$ (red) and $f_\downarrow$ (blue) for a varying tunnel conductance ($V_{DC} = 50$ mV, $I_{tun} = 15$ pA, $V_{RF} = 20$ mV, $T = 1.2$ K, $B_{ext} = 0.9$ T, $\theta_{ext} = 82^\circ$). The solid curves are Lorentzian fits. The curves are successively shifted vertically by 0.3 pA for clarity. (B) Peak heights vs. tunnel conductance extracted from cw-ESR spectra in (A). Fits according to the discussion in the main text are overlaid (solid curves). Note that the extrapolation of the fit curves to zero conductance reads finite intercepts $I_{sat} = 0.226$ pA ($f_\uparrow$) and 0.142 pA ($f_\downarrow$). The three insets illustrate the vectorial relationships between the ESR driving fields from the Fe (red or blue) and the tip (light green). (C) Vectorial relationship of driving fields from Fe ($B_{1Fe}$) and tip ($B_{1tip}$). $B_0$ is the total static field at the Ti position, composed of external ($B_{ext}$), tip-induced field ($B_{tip}$), and Fe-induced ($B_{Fe}$) fields. $B_{1Fe\perp}$ and $B_{1tip\perp}$ denote projections of $B_{1Fe}$ and $B_{1tip}$, respectively, to a plane perpendicular to the total static field $B_0$. (D) A schematic of Bloch sphere in a condition that $B_{1Fe\perp}$ and $B_{1tip\perp}$ are antiparallel ($\phi = 180^\circ$) and showing resultant Rabi rotations of the Bloch vector of the Ti spin (brown thick arrow). Note that the net driving field $B_{1\perp}$ changes its direction depending on the magnitude of the tip-induced driving field $B_{1tip\perp}$, leading to corresponding change in the Rabi rotation as well.
Fig. 4: (A and B) ESR resonance peak frequencies (A) and splitting $\Delta f$ (B) in cw-ESR spectra measured from Ti-Fe pairs of 0.72 nm (red) and 0.59 nm (blue) at a varying polar angle ($\theta_{\text{ext}}$) of $B_{\text{ext}}$. Solid curves are fits using the model described in the equations (5) and (6). Red solid (dashed) curves correspond to the up (down) state of the tip spin. ($V_{\text{DC}} = 200$ mV, $I_{\text{tun}} = 10$ pA, $V_{\text{RF}} = 30$ mV for the 0.59 nm pair; $V_{\text{DC}} = 30$ mV, $I_{\text{tun}} = 20$ pA, $V_{\text{RF}} = 30$ mV for the 0.72 nm pair; $T = 0.4$ K, $B_{\text{ext}} = 0.6$ T). (C and D) Schemes of net magnetic field $B_{\text{net}}$ at the Ti position, composed of the external field $B_{\text{ext}}$ and Fe-induced field $B_{\text{Fe}}$ in the plane of $B_{\text{ext}}$ vector with an arbitrary (C) and 90° (D) field angle $\theta_{\text{ext}}$. $\alpha$ and $\alpha_0$ denote the angles added by the Fe-induced field.
Fig. 5: (A) A Fe-Ti-Fe spin complex with Ti-Fe separations of 0.72 nm. (B) Continuous wave ESR spectra measured on Ti of the complex in (A) (purple) and a TiFe pair of 0.72 nm (red). (C) Rabi rates obtained from pulsed ESR of the complex (purple) and TiFe pair of 0.72 nm (red), and an isolated Ti (gray) at a varying tunnel conductance. The insets schematically depict the Ti-Fe interactions for three groups of Rabi rates ($V_{RF} = 100$ mV, $T = 1.2$ K, $B_{ext} = 0.9$ T, $\theta_{ext} = 82^\circ$).
SUPPLEMENTARY MATERIALS

Materials and Methods

Supplementary 1: ESR peak splitting measured on Ti-Fe pairs

Supplementary 2: Eigenstates and ESR transitions of Ti spins in Ti-Fe and Fe-Ti-Fe complexes

Supplementary 3: Model simulations of ESR transitions in Ti-Fe pairs

Supplementary 4: Pulsed ESR data corresponding to all the Rabi rates in Fig. 2C

Supplementary 5: Analysis of Rabi rates vs. tunnel conductance in Fig. 2C

Supplementary 6: ESR spectra of a single Ti vs. conductance with the same tip in Fig. 3

Supplementary 7: CW- and pulsed-ESR on Ti-Fe pairs and a single Ti vs. field angle

Supplementary 8: Pulsed ESR data corresponding to all the Rabi rates in Fig. 5C

Supplementary 9: A case of non-perfect cancellation between $B_{1tip}$ and $B_{1Fe}$. 
Materials and Methods:

An atomically clean Ag(100) substrate was prepared by alternating Ar+ sputtering and annealing cycles. MgO films were grown on the Ag substrate at 580 K by evaporating Mg in an O\(_2\) atmosphere of 1.1×10\(^{-6}\) Torr. Then, Fe and Ti atoms were deposited on the MgO surface pre-cooled at about 50 K. All measurements were performed on Ti atoms bound to a bridge site of the MgO surface (that is, in the middle of two oxygen sites). Before measurements, the STM tip made by Pt/Ir wire was poked into the Ag(001) surface until satisfactory topographic and spectroscopic features were observed on atoms on MgO, after which Fe atoms were picked up by the STM tip from MgO (by applying a DC voltage pulse of 0.3 V) to create a spin-polarized tip. The tip’s spin polarization was calibrated with the asymmetry around zero bias in the \(dl/dV\) spectra of Ti on MgO.

Measurements were performed in two ultrahigh-vacuum (< 10\(^{-10}\) mbar) scanning tunneling microscopes (STMs). The data presented in Figs. 1, 2, 3, 5 were measured in a home-built \(^3\)He-cooled STM at \(T = 1.2\) K equipped with a single-axis superconducting magnet. The data presented in Fig. 4 and Figs. S12–S14 were measured in a commercial \(^3\)He-cooled STM (Unisoku, USM1300) at \(T = 0.4\) K equipped with two-axis superconducting magnets. High-frequency transmission cables were installed on the STM systems as described in detail elsewhere [Paul2017, Hwang2022]. Continuous wave electron spin resonance (ESR) spectra were acquired by sweeping the frequency of an RF voltage \(V_{RF}\) generated by an RF generator (Agilent E8257D) across the tunneling junction and monitoring changes in the tunneling current. For pulsed ESR, the RF generator was gated by the pulse outputs programed in arbitrary waveform generators (Tektronix 7122C for 1K-system; Tektronix 5000 for 0.4K-system). The output signal of the RF generator was combined with a DC bias voltage through a bias tee (SigaTek, SB15D2). In both CW- and pulsed-ESR measurements, the RF signals were chopped at 95 Hz and sent to a lock-in amplifier (Stanford Research Systems SR860) and recorded by a DAQ-Box (National Instruments 6363). The bias voltage \(V_{DC}\) refers to the sample voltage relative to the tip. The STM constant-current feedback loop was set to a low gain during the measurements.
Supplementary 1: ESR peak splitting measured on Ti-Fe pairs

Section 1.1. Dependence on Ti-Fe separation

As discussed in the main text (Fig. 1E), the ESR peak splitting measured on a Ti-Fe pair monotonically increased for a decreasing separation between two atoms. Figures S1A and B show ESR spectra measured on Ti-Fe pairs of five different separations and peak splitting with respect to the Ti-Fe separation.

Fig. S1. ESR peak splitting of pairs with various Ti-Fe separations. (A) Spectra measured on seven pairs of different Ti-Fe separations ($V_{RF} = 20$ mV, $T = 1.2$ K, $B_{ext} = 0.9$ T, $\theta_{ext} = 82^\circ$). The inset is a zoom-in of the spectrum on the pair with a separation of 1.13 nm. (B) Dependence of peak splitting on Ti-Fe separation, extracted from 24 pairs. The red solid curve is a fit to an exponentially decaying function in the equation (S1), resulting in the decay length $d_{ex}$ of the Ti-Fe interaction shown in the table (red, inset), with those from the other spin pairs for comparison [Yang2017,Bae2018].
Assuming an isotropic exchange interaction, we applied an exponential dependence of splitting ($\Delta f$) on Ti-Fe separation ($r_{\text{TiFe}}$) [Yang2017]

$$\Delta f(r_{\text{TiFe}}) = J_0 \exp[-(r_{\text{TiFe}} - r_0)/d_{\text{ex}}] \quad (S1)$$

where $J_0$ and $d_{\text{ex}}$ are the exchange coupling energy at $r_{\text{TiFe}} = r_0$ and the decay length of the Ti-Fe interaction. Fitting the data in Fig. S1B to the equation (S1) with $r_0 = 0.59$ nm resulted in $d_{\text{ex}} = 86 \pm 27$ pm and $J_0 = 3.22 \pm 0.45$ GHz (red curve).

Section 1.2. Dependence on pair orientations

We note that the pairs with a separation of 0.72 nm showed a mean splitting of 0.9 GHz but with a large dispersion of about 0.5 GHz (Fig. S1). Due to the out-of-plane MAE of the Fe spin, a possible contribution from dipole-dipole interaction should have the same form as that from exchange interaction, leading to a total spin-spin interaction to be a simply combined shape ($J - D$) $S_{\text{Ti}} \cdot S_{\text{Fe}}$, where $D$ is dipole-dipole coupling strength for a given Ti-Fe separation. Thus, we expect that the total spin-spin interaction should be isotropic for the azimuthal orientation of a pair. To survey this, we measured ESR peak splitting on Ti-Fe pairs of 0.72 nm with different pair orientations as shown in Fig. S2. The splitting is anisotropic and doesn’t show any regular dependence on the pair orientation. We speculate that this is from the difference in local environment of each pair and/or existence of defects nearby, unresolvable using normal STM microscopy and spectroscopy.

Section 1.3. Dependence on tips

One possible origin of such a large dispersion of peak splitting observed from pairs of 0.72 nm is the anisotropic local field from the tip spin since it contributes an additional Zeeman energy to the atomic spins, which is comparable in size to the spin-spin interactions in the pair. We performed ESR on four pairs (‘1’ – ‘4’) indicated in Fig. S2 with 21 different tips and show the measured splitting in Fig. S3. The mean splitting of the four pairs showed a large variation of 0.3 – 1.3 GHz as that shown in Fig. S2 for the same pairs. However, the tip-dependent variation of the splitting on each pair is in the order of 0.1 GHz, indicating that the ESR splitting was mainly contributed by the Ti-Fe interaction inherent in each pair when it formed on the surface.
Fig. S2. Dependence of ESR peak splitting on pair orientations. (A) A schematic illustrating a Ti-Fe pair with separation of 0.72 nm. The position of Ti is measured in the polar coordinate (\( \mathbf{r}_{\text{TiFe}} \), \( \phi_B \)), measured from the position of Fe and in-plane component of the external field (\( \mathbf{B}_{\text{ext},\parallel} \)). The gray mesh represents the underlaying MgO(100) lattice. (B) A collection of ESR peak splitting measured on pairs with separation of 0.72 nm as a function of azimuthal angle (\( \phi_B \)) of Ti atom. The radius of the graph denotes the splitting (\( \Delta f \)) (\( B_{\text{ext}} = 0.9 \) T, \( \theta_{\text{ext}} = 82^\circ \)).

Fig. S3. Dependence of ESR peak splitting on different tips used. ESR peak splitting measured from the four pairs with the same separation of 0.72 nm, denoted ‘1’ – ‘4’ in Fig. S2, using 21 different tips (\( B_{\text{ext}} = 0.9 \) T, \( \theta_{\text{ext}} = 82^\circ \)). Note that splitting measured from pairs of 0.72 nm scatters in the range of 0.2 – 1.4 GHz (see also Figs. S1 and S2), however, each pair showed a much weaker dependence on the tips used, with a dispersion of ~ 0.1 GHz from its mean splitting. The open circle represents the average splitting of each pair with its error bar.
**Supplementary 2:** Eigenstates and eigenvalues of Ti-Fe pairs and Fe-Ti-Fe complex

**Section 2.1. Ti-Fe pair**

![Model Hamiltonian of Ti-Fe pairs](image)

**Fig. S4. Model Hamiltonian of Ti-Fe pairs.** Checkerboard representations of (A) the Hamiltonian shown in the equation (1) and (B) the eigenstates obtained by the diagonalization of (A).

**Section 2.2. Fe-Ti-Fe complex**

To survey the eigenstates and possible ESR transitions of the Ti spin in a Fe-Ti-Fe pair shown in Fig. 5, we set up a model Hamiltonian

$$
\hat{H}_{\text{FTF}} = D_{\text{Fe}} S_{1Z}^2 + C_{\text{Fe}} (S_{1+}^4 + S_{1-}^4) + D_{\text{Fe}} S_{2Z}^2 + C_{\text{Fe}} (S_{2+}^4 + S_{2-}^4) - g_{\text{Fe}} \mu_B (S_1 + S_2) \cdot B_{\text{ext}}
+ J_{\text{TiFe1}} S_{\text{Ti}} \cdot S_1 + J_{\text{TiFe2}} S_{\text{Ti}} \cdot S_2 - g_{\text{Ti}} \mu_B S_{\text{Ti}} \cdot B_{\text{ext}} \quad (S2),
$$

where two Fe spins, $S_1$ and $S_2$, are identical with two exchange couplings, $J_{\text{TiFe1}}$ and $J_{\text{TiFe2}}$, with the Ti spins, respectively. We used the same $D_{\text{Fe}}$, $C_{\text{Fe}}$, and $g_{\text{Fe}}$ as those in the Hamiltonian for a Ti-Fe pair (equation (1)) and fixed $J_{\text{TiFe1}}$ to be $J_{\text{TiFe}} = 1.1$ GHz as extracted from Fig. 4. We used $J_{\text{TiFe2}}$ as a parameter to study on a dependence of possible ESR transitions on the difference in the two Ti-Fe couplings. Similar to the case that a single Fe is coupled with the Ti spin, the Hamiltonian is almost diagonal in the basis set of the Zeeman product states of the three spins, $|S_z(\text{Ti}) S_z(\text{Fe}_1) S_z(\text{Fe}_2)>$, (Fig. S5A). The eigenstates of $\hat{H}_{\text{FTF}}$ are well decoupled by the four distinct spin orientations of two Fe spins, $|\uparrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$, $|\uparrow\downarrow\rangle$, and $|\downarrow\uparrow\rangle$, as shown in Fig. S5B.
Fig. S5. Model Hamiltonian of a Fe-Ti-Fe spin complex. Checkerboard representations of (A) subset of the Hamiltonian matrix for the first Fe (Fe₁) is up (ℏ; ℓ = 2) and (B) the eigenstates of the eight lowest energies. The spin states are labelled according to the array of three spins, |S_z(Ti) S_z(Fe₁) S_z(Fe₂)>.
**Supplementary 3:** Model simulations of ESR transitions in Ti-Fe pairs

Fig. S6. Model simulations of ESR transitions in Ti-Fe pairs. (A) Eigenstates and (B) peak splitting as a function of Ti-Fe separation using the model Hamiltonian in equation (5). In (A), we shifted the four as-calculated eigenenergies by adding the Zeeman energies of the Fe spin for the two ESR transitions ($f_\uparrow$ and $f_\downarrow$) to be clearly visible.
**Supplementary 4:** Pulsed ESR data corresponding to all the Rabi rates in Fig. 2C

![Graphs showing pulsed ESR data](image)

**Fig. S7.** Tunnel conductance dependence of Rabi oscillations measured on the Ti-Fe pairs shown in Figs. 1b-d. Solid curves are the fits using an exponentially decaying sinusoidal function, resulting in the Rabi rates in Fig. 2c. \(I_{DC} = 10\ \text{pA},\ T = 1.2\ \text{K},\ B_{ext} = 0.9\ \text{T},\ \theta_{ext} = 82^\circ\).
**Supplementary 5**: Analysis of Rabi rates vs. tunnel conductance in Fig. 2C

Section 5.1. Two driving fields

The Rabi rate (Ω) of the Ti spin is directly proportional to the magnitude of the effective total driving field $B_{1\perp}$, residing in the plane perpendicular to the total static field $B_0$, with a proportionality constant α. The $B_{1\perp}$ is the vector sum of the tip-originated ($B_{1\text{tip}\perp}$) and Fe-originated ($B_{1\text{Fe}\perp}$) ones as depicted in Fig. 3C, hence its magnitude is calculated by

$$B_{1\perp} = \sqrt{B_{1\text{Fe}\perp}^2 + B_{1\text{tip}\perp}^2 + 2B_{1\text{Fe}\perp}B_{1\text{tip}\perp}\cos\phi} \; (S4),$$

where $\phi$ is the angle between the two vectors $B_{1\text{tip}\perp}$ and $B_{1\text{Fe}\perp}$. By assuming that the contribution from Fe ($B_{1\text{Fe}\perp}$) for a given Ti-Fe separation is independent of the tunnel conductance ($\sigma_{\text{tun}}$), i.e. Ti-tip distance ($r_{\text{Ti-tip}}$), we show in Fig. S8B the magnitude of the total driving field ($B_{1\perp}$) as a function of that of the tip-originated driving field $B_{1\text{tip}\perp}$. First, we note two special cases: (i) with $\phi = 0$ ($B_{1\text{tip}\perp}$ and $B_{1\text{Fe}\perp}$ are parallel.), $B_{1\perp}$, and thus $\Omega$, is maximum for the whole range of $B_{1\text{tip}\perp}$. (ii) with $\phi = 180^\circ$ ($B_{1\text{tip}\perp}$ and $B_{1\text{Fe}\perp}$ are antiparallel.), $B_{1\perp}$ becomes zero at $B_{1\text{tip}\perp} = B_{1\text{Fe}\perp}$ and bounces back for $B_{1\text{tip}\perp} < B_{1\text{Fe}\perp}$. Both cases show saturations of $B_{1\perp}$ to $B_{1\text{Fe}\perp}$ at $B_{1\text{tip}\perp} = 0$, which is trivial. These two cases lead to the tunnel conductance dependence of the ESR peak amplitudes shown in Fig. 3B. For an arbitrary choice of two driving vectors, neither parallel nor antiparallel to each other, $B_{1\perp}$ shows a trend in between abovementioned two cases and also saturates to the same point at $B_{1\text{tip}\perp} = 0$.

Section 5.2. Fit the Rabi rates vs. tunnel conductance in Fig. 2C

Using the linear dependence of the Rabi rate of an isolated Ti spin on the tunnel conductance (gray in Fig. 2C), we can scale the conductance axis to the Rabi rate contributed only by tip ($\Omega_{\text{tip}}$), as discussed in the following. The tunnel conductance $\sigma_{\text{tun}}$ and tip-originated driving field $B_{1\text{tip}\perp}$ can be described using the tip height ($z$) as follows [Yang2019prl]:

$$\sigma_{\text{tun}} = \sigma_0 e^{-z/d_0} \; (S5),$$

$$B_{1\text{tip}\perp} = c_{1\text{tip}\perp} e^{-z/d_{\text{ex}}/z} \; (S6),$$

where $\sigma_0$, $d_0$, and $d_{\text{ex}}$ are the tunnel conductance at the point contact, decay lengths of tunneling probability, and that of exchange coupling between tip and Ti spins. Here, we take the tip height to be zero at the tip-Ti point contact. Using (S5), (S6) is transformed into

$$B_{1\text{tip}\perp} = \frac{c_{1\text{tip}\perp}}{d_0} \left( \frac{\sigma_{\text{tun}}}{\sigma_0} \right) \frac{d_0/d_{\text{ex}}}{\ln(\sigma_0/\sigma_{\text{tun}})} \; (S7).$$

Set the parts of the tunnel conductance in (S7) as $x_{\text{tip}}$ and $c_{1\text{tip}\perp}/d_0$ as $\beta$, together with the relation in (S3), we obtain

27
\[ \Omega_{\text{tip}}/\alpha\beta = x_{\text{tip}}, \text{ where } x_{\text{tip}} \equiv \left( \frac{\sigma_{\text{tun}}}{\sigma_0} \right)^{d_0/d_{\text{ex}}} \frac{1}{\ln(\sigma_0/\sigma_{\text{tun}})} \] (S8).

Then using (S4) and (S8), the Rabi rate \((\Omega; y\text{-axis of Fig. 2C})\) is scaled into

\[ \Omega/\alpha\beta = \left( x_{\text{Fe}}^2 + x_{\text{tip}}^2 + 2 x_{\text{Fe}} x_{\text{tip}} \cos \phi \right)^{1/2} = y \] (S9),

where \(x_{\text{Fe}} \equiv B_{1\text{Fe}, \perp}/\beta\). The plots \(\Omega(\sigma_{\text{tun}})\) in Fig. 2C will transform into \(y(x_{\text{tip}})\) using (S8) and (S9), and \(\alpha\beta\) can be extracted from the data for an isolated Ti spin using the relation in (S7).

We obtained \(d_0 = 43.4\ \text{pm}\) by using the equation (S5) with \(\sigma_0 = 0.6\ \text{mS}\) from the point contact experiment data on a bridge-site Ti atom [Bae2018]. The \(d_{\text{ex}}\) was extracted using a tunnel conductance dependence of ESR resonance frequency \(f_{\text{res}}\) measured on an isolated Ti, as shown in Fig. S9A. The linear dependence of \(f_{\text{res}}\) on the tunnel conductance suggests the exchange interaction between tip and Ti as a dominant contribution to the shift of the resonance frequency. From a linear fit, we obtained the \(f_{\text{res}}\) of 22.485 GHz at zero conductance and calculated \(f_{\text{res}}\) shift \((\Delta f_{\text{res}})\) at each measurement tunnel conductance. Assuming an exponential dependence of \(\Delta f_{\text{res}}\) on the tip height \((z)\)

\[ \Delta f_{\text{res}}(z) \propto \exp\left[-(z - z_0)/d_{\text{ex}}\right] \] (S10),

we obtained the decay length of \(d_{\text{ex}} = 38.2\ \text{pm}\) for the tip-Ti interaction. Together with the as-obtained \(d_0\) and \(\sigma_0\), we scaled the \(\Omega(\sigma_{\text{tun}})\) plots in Fig. 2C into the \(y(x_{\text{tip}})\) plane as shown in Fig. S10. We fit the data using (S9) with \(x_{\text{Fe}}\) and \(\phi\) as fitting parameters, resulting in the zero conductance Rabi rates \(\Omega_0\) for the two pairs of 0.59 and 0.72 nm.

**Fig. S8.** (A) A schematic illustration of two driving fields \((B_{1\text{tip}, \perp}, B_{1\text{Fe}, \perp})\) in ESR of Ti in a Ti-Fe pair. The red and blue arrows denote two Fe-induced driving fields for two quasi-static spin states of Fe, \(|\uparrow\rangle\) and \(|\downarrow\rangle\), respectively. \(B_0\) is the total static field applied to the Ti spin, composed of three contributions from external, tip-induced, and Fe-induced fields. (B) Magnitude of total driving field \((B_{1\perp})\) following the formula (S4) as a function of tip-originated driving field \((B_{1\text{tip}, \perp})\), depending on the angle \((\phi_{\parallel}, \phi_{\perp})\) between \(B_{1\text{tip}, \perp}\) and \(B_{1\text{Fe}, \perp}\).
Fig. S9. ESR resonance frequency on an isolated Ti. (A) Resonance frequency ($f_{\text{res}}$) as function of tunnel conductance. (B) Shift of $f_{\text{res}}$ ($\Delta f_{\text{res}}$) measured from the $f_{\text{res}}$ at zero conductance, as indicated in (A), as a function of tip height ($z$). Using as-obtained $d_0$ and $\sigma_0$ in the discussion above, we converted the measurement tunnel conductance into the tip height ($z$). The solid lines are the linear fits of the plots.

Fig. S10. Fits of Rabi rates vs. tunnel conductance for the three cases in Fig. 2C. Replot the data in Fig. 2C using the model discussed in the above text with the x- and y-axis variables defined in equations (S8) and (S9). The fits resulted in the parameters $\Omega_0$ and $\phi$, as shown in the inset.
Supplementary 6: CW-ESR on a single Ti atom vs. conductance, with the same tip in Fig. 3

Fig. S11. Tunnel conductance dependence of ESR spectra of a single Ti measured with the same tip used in Fig. 3. As the tip was move out from the Ti spin, the peak height monotonically decreases, and it vanishes. ($V_{DC} = 50 \text{ mV}, T = 1.2 \text{ K}, B_{ext} = 0.9 \text{ T}, \theta_{ext} = 82^\circ$).
Supplementary 7: CW- and pulsed-ESR on Ti-Fe pairs and a single Ti vs. external field angle

Section 7.1. Dependence of ESR spectra on external field angle

Fig. S12. ESR of Ti-Fe pairs with a varying angle of external field ($\theta_{\text{ext}}$). ESR spectra on (A) an isolated Ti ($I_{\text{tun}} = 10$ pA, $V_{\text{DC}} = 30$ mV, $V_{\text{RF}} = 30$ mV) and (B and C) Ti-Fe pairs with separations of 0.72 nm ($I_{\text{tun}} = 20$ pA, $V_{\text{DC}} = 30$ mV, $V_{\text{RF}} = 30$ mV) and 0.59 nm ($I_{\text{tun}} = 10$ pA, $V_{\text{DC}} = 200$ mV, $V_{\text{RF}} = 30$ mV), respectively. Each spectrum is indicated by the polar angle, in degree ($^\circ$), of the external field used in measurement and presented with a $I_{\text{ESR}}$-offset for clarity. Gray dotted line indicates artifacts from the transfer function (TF) of the RF transmission at 14.6 GHz.
Section 7.2. Analysis of CW- and pulsed-ESR data of an isolated Ti spin

As discussed in the 'MODEL STUDY' section in the main text, the spins of the tips used in this work were well described, like the Fe spin, by a classical magnetic moment of uniaxial anisotropy along a direction \( \mathbf{n}_{\text{tip}} \) (Eqn. (4)). To have a quantitative insight on the contribution of the tip spin to the measurements of the Ti-Fe pairs (Fig. 4), we performed both CW- and pulsed-ESR measurements on an isolated Ti with the same tip. In Fig. S13, we show resonance frequency \( f_{\text{ESR}} \), Rabi rate, and CW-ESR amplitude as a function of the angle \( (\theta_{\text{ext}}) \) of the external field. A time-independent Hamiltonian \( \hat{H}_0 \) of this system can be written as

\[ \hat{H}_0 = -g_{\text{Ti}} \mu_B \mathbf{S}_{\text{Ti}} \cdot \mathbf{B}_{\text{ext}} - g_{\text{Fe}} \mu_B \mathbf{S}_{\text{tip}} \cdot \mathbf{B}_{\text{ext}} + J_{\text{Ti,tip}} \mathbf{S}_{\text{Ti}} \cdot \mathbf{S}_{\text{tip}} \quad (S11), \]

where the first two terms represent the Zeeman energies of the Ti and tip spins, and the last term does the interaction between two spins. We referred the \( g \)-factor of Ti spin as reported previously [KimJK2021].

We choose Cartesian coordinates, where the sample surface and external field are confined in the \( xy \)- and \( xz \)-plane, respectively, so that the directions of the external field and Ti spin are described by conventional definitions of the polar and azimuthal angles, \( \theta_{\text{ext}} \) and \( \mathbf{n}_{\text{tip}}(\theta_{\text{tip}}, \phi_{\text{tip}}) \), as illustrated in Fig. S13A. Since the Ti-tip interaction was smaller than the Zeeman energy of the Ti by about two orders of magnitude (Fig. S14), we approximate the direction of the Ti spin to be along the external magnetic field. Hence, the external field and tip spin can be written as

\[ \mathbf{B}_{\text{ext}} = B_{\text{ext}} \mathbf{n}_{\text{ext}}, \quad \text{where} \quad \mathbf{n}_{\text{ext}} = \hat{x} \sin \theta_{\text{ext}} + \hat{z} \cos \theta_{\text{ext}} \quad (S12), \]

\[ \mathbf{S}_{\text{tip}} = \pm \langle S_{\text{tip}} \rangle | \mathbf{n}_{\text{tip}} \rangle \quad (S13). \]

Diagonalization of the Hamiltonian (S11) results in four eigenstates \( |+\rangle |\uparrow\rangle, \langle -|\uparrow\rangle, |+\rangle |\downarrow\rangle, \langle -|\downarrow\rangle \), where the first and second parts of each represent Ti and tip spin states, and corresponding eigenenergies parameterized by \( \theta_{\text{tip}}, \phi_{\text{tip}}, \) and \( J_{\text{Ti,tip}} \langle S_{\text{tip}} \rangle \). We obtain two possible ESR transitions of the Ti spin for two spin states of the tip \( |+\rangle |\uparrow\rangle \leftrightarrow |+\rangle |\downarrow\rangle \) and \( |+\rangle |\downarrow\rangle \leftrightarrow |+\rangle |\uparrow\rangle \) as a function of \( \theta_{\text{ext}} \). A simulation of the resonance frequency using this model with \( \theta_{\text{tip}} = 65^\circ \) and \( \phi_{\text{tip}} = 75^\circ \) is in a great agreement with the experiment (red curves in Fig. S13B). The overall \( \theta_{\text{ext}} \)-dependent variation of the resonance frequency ranging about 2 GHz stemmed from the anisotropy in the \( g \)-factor of the Ti spin \( (g_{\text{Ti}}) \) [KimJK2021]. We note a crossing of two simulation curves at about \( \theta_{\text{ext}} = -25^\circ \), across which the Zeeman energy forces the tip’s spin state to flip from \( |\uparrow\rangle \) to \( |\downarrow\rangle \) (or vice versa).

Time-dependent perturbation theory deduces the Rabi rate \( (\Omega) \) of a spin \( S \) \( (S = 1/2) \)

\[ \hbar \Omega = g \mu_B (|+\rangle \langle B_1 | \cdot \mathbf{S} | -\rangle) \quad (S14) \]

for a given time-varying magnetic field \( \mathbf{B}_1 \). Only the component of \( \mathbf{B}_{\text{tip}} \) perpendicular to the total static magnetic field \( \mathbf{B}_0 \) drives the ESR of the spin, leading to a dependence of the Rabi rate on the angle \( \gamma \) between \( \mathbf{B}_0 \) and \( \mathbf{B}_{\text{tip}} \).
Here $\gamma$ is approximately determined by the directional cosine between two unit vectors, $\mathbf{n}_{\text{ext}}$ and $\mathbf{n}_{\text{tip}}$, \( \cos \gamma = \mathbf{n}_{\text{ext}} \cdot \mathbf{n}_{\text{tip}} \). The $\theta_{\text{ext}}$-dependence of the Rabi rate is then described by

\[
\Omega(\theta_{\text{ext}}) = \Omega_{\gamma=90^\circ} \sqrt{1 - (\sin \theta_{\text{tip}} \cos \phi_{\text{tip}} \sin \theta_{\text{ext}} + \cos \theta_{\text{tip}} \cos \theta_{\text{ext}})^2}
\] (S16)

with the polar and azimuthal angles of the tip ($\theta_{\text{tip}}$, $\phi_{\text{tip}}$). Note that the tip spin flips when $\gamma$ changes across 90° or 270°, where the Rabi rate is the maximum ($\Omega_{\gamma=90^\circ}$), leading to a continuous evolution of the Rabi rate with a periodicity of 180° in the $\theta_{\text{ext}}$ axis. With the $\theta_{\text{tip}}$ and $\phi_{\text{tip}}$ from the analysis of the resonance frequency (Fig. S13B), we simulated the $\theta_{\text{ext}}$-dependence of the Rabi rate as shown in Fig. S13C (solid orange curve), which is in good agreement with the experimental data.

The influence of the uniaxial anisotropy of the tip spin also appears in the $\theta_{\text{ext}}$-dependence of the ESR peak amplitude. We calculated a simulation curve (purple curve in Fig. S13D) using the model introduced in a previous report [KimJK2021] with the anisotropy of our tip, $\mathbf{n}_{\text{tip}}(\theta_{\text{tip}}, \phi_{\text{tip}})$, extracted from the analysis of the resonance frequency (Fig. S13B). The result fits in great agreement with the experimental data, with assignments of critical angles where the ESR amplitude becomes minima when the spins of the tip and Ti are either parallel or perpendicular due to its dependence on the magnetoresistance of the tunnel junction. These angles for the perpendicular and parallel configurations of the two spins coincide with the angles for the maximum ($\Omega_{\text{max}}$) and minimum ($\Omega_{\text{min}}$) Rabi rates, a compelling evidence that the tip spin provides the driving field of the ESR on the Ti spin, which is maximum when it is perpendicular to the total static magnetic field.
Fig. S13. Analysis of data from an isolated Ti spin. (A) A schematic showing the definitions of the angles used in the model. Plots of (B) ESR frequencies, (C) Rabi rates, and (D) ESR amplitudes measured from an isolated Ti spin as a function of the polar angle of the external field ($\theta_{\text{ext}}$) ($I_{\text{tun}} = 10$ pA, $V_{\text{DC}} = 30$ mV). Solid curves are the fits using the model discussed in the text.
Section 7.3. CW- and pulsed-ESR data of an isolated Ti: tunnel conductance dependence

Figures S14A and B show CW- and pulsed-ESR data measured at four different tunnel conductance on the same isolated Ti atom and with the same tip used for the measurements in Fig. 4 and Fig. S13. In Fig. S14C, we show the dependence of the resonance frequency on the tunnel conductance. From a linear fit, we extracted the resonance frequency at zero conductance and estimated the tip-field-induced Zeeman energy of 0.008 GHz at 0.05 nS. This is smaller by ~600 times than the Ti-Fe interaction ($J_{TiFe}$) of 4.91 GHz in the pair with a separation of 0.59 nm and at the field angle of 72°, as can be seen in the data in Fig. 4 and Fig. S12C where the same tunnel conductance (0.05 nS: $V_{DC} = 200$ mV, $I_{DC} = 10$ pA) was used. This fulfills the condition $J_{TiFe} \gg J_{Tiltip}$ in the discussion of the main text. In addition, only a noisy fluctuation was observed in the Rabi measurement at 0.1 nS (Fig. S14B), which is also a compelling evidence, supporting the negligible contribution of the tip-induced field to the data in Fig. S12C.

Fig. S14. Tunnel conductance dependence of CW- and pulsed-ESR on an isolated Ti. (A) CW-ESR spectra and (B) Rabi oscillations measured on the same Ti atom as used for the data in Fig. S13. (C) ESR resonance frequencies extracted from the spectra in (A). Solid line is a linear fit. ($I_{DC} = 10$ pA, $T = 0.4$ K, $B_{ext} = 0.6$ T, $\theta_{ext} = 72^\circ$).
Supplementary 8: Pulsed ESR data corresponding to all the Rabi rates in Fig. 5C

Fig. S15. Tunnel conductance dependence of Rabi oscillations measured on the Fe-Ti-Fe complex shown in Fig. 5. Solid curve are the fits using an exponentially decaying sinusoidal function, resulting that the Rabi rates $\Omega$ in Fig. 5c were extracted. ($I_{\text{DC}} = 10 \text{ pA}, T = 0.6 \text{ K}, B_{\text{ext}} = 0.9 \text{ T}, \theta_{\text{ext}} = 82^\circ$).
**Supplementary 9:** A case of non-perfect cancellation between $B_{1\text{tip}}$ and $B_{1\text{Fe}}$

![ESR Spectra](image_url)

**Fig. S16. Tunnel conductance dependence of ESR spectra on a Ti-Fe pair.** (left) ESR spectra showing both lower ($f_{\uparrow}$) and higher ($f_{\downarrow}$) resonances. (right) Zoom-in across the peak at the higher resonance ($f_{\downarrow}$). Note that both peaks showed monotonic decrease of the height as the tip was moved out from the Ti spin, in a tunnel conductance regime comparable to that in Fig. 3 ($I_{\text{DC}} = 10 \text{ pA}, \, T = 1.2 \text{ K}, \, B_{\text{ext}} = 0.9 \text{ T}, \, \theta_{\text{ext}} = 82^\circ$).