

Efficient driving of a spin qubit using single-atom magnets

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The realization of electron-spin resonance at the single-atom level using scanning tunneling microscopy has opened avenues for coherent quantum sensing and quantum state manipulation at the ultimate size limit. This allows us to build many-body Hamiltonians and the study of their complex physical behavior. Recently, a qubit platform has emerged from this field, raising questions about the driving mechanism from single-atom magnets. In this work, we demonstrate how single-atom magnets can be used to drive a nearby single spin qubit efficiently. We show that the modulation of exchange coupling is the primary driving force, which successfully reproduces Rabi rates in the tens of MHz range, consistent with experimental data, while also addressing critical aspects related to the optimization of experimental parameters.

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Introduction. Since the first demonstration of electron-spin resonance (ESR) on the level of individual atoms using a scanning tunneling microscope (STM), the combination of ESR and STM has been expanded to the study of single atoms, pairs, and molecules [1–24]. Pulsed ESR techniques have also been used to coherently control the quantum state of single [25,26] and multiple electrons on surfaces [27]. At the same time, the mechanism that allows to drive ESR in an STM using time-varying electric fields has remained somewhat elusive.

Originally, it was proposed that a modulation of the crystal field by the applied electrical field is responsible for creating the necessary transition matrix elements [1]. Then, it was suggested that the applied electrical field modulates the coupling between the magnetic STM tip apex and the spin [28] or changes the g factor of the adatom [29]. And recently, it was suggested that the ac magnetic field component of the electromagnetic field supplied by an antenna in the STM junction region could serve as a driving field [30]. Some of us showed that an ac voltage is able to reduce or increase the tunneling barrier, which modulates the coupling between the tip and the

spin [31–34]. This modulation in the presence of a polarized electrode leads to a time-dependent exchange field that drives ESR when on resonance with a spin transition [35,36]. In all cases, the model incorporated some influence from the STM tip, which, in typical STM fashion, can be expected to have a highly localized interaction with the spin on the surface. All of the previous approaches were challenged when it was discovered that a spin can be coherently driven *without* being directly under the tip apex, by placing a magnetic atom nearby [27,37–39].

A well-demonstrated system is the engineered Fe-Ti pair [37]. In this system, the Ti spin state can be driven either by the tip or by the nearby Fe atom. Here, the Fe atom can be approximated as a single-atom magnet since it possesses an anisotropy barrier against spin reversal, and the lifetime of its spin state is $\sim 10^5$ times longer than the coherence time of the Ti spin [40]. Using two Ti spins, where one spin played the role of the sensor while the other formed an atomic pair with an Fe atom, it was also shown that the driving mechanism works when the controlled spin is outside the tunnel junction [26,27,38,39]. This experimental observation is not compatible with the assumption of an exchange modulation between the tip apex and the surface spin, which has a range of a few angstroms. However, the electric field that emanates from the wider tip body or a nearby antenna covers an area of possibly several millimeters squared, which allows it to drive spins outside of the tunnel junction [26,27,38,39]. This field could modulate the g factor of the surface spins, the on-site crystal-field anisotropy terms, or the exchange coupling between Ti and Fe. However, the absence of any measurable effect of the electric field on the magnetic moment observed

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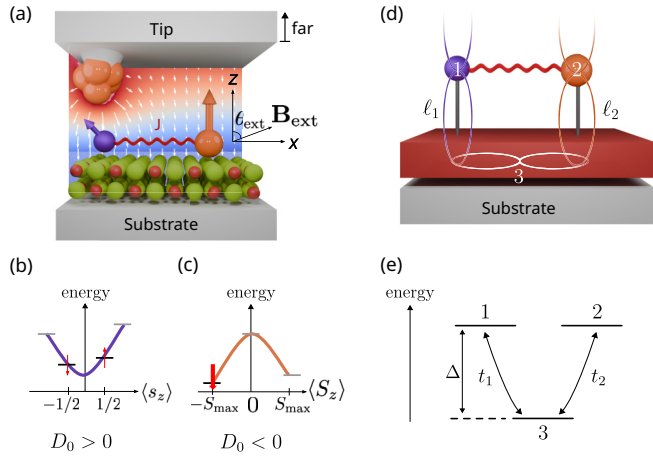


FIG. 1. Spin-qubit system: (a) Front view of two atoms subject to an electric field in the STM junction. External magnetic field direction as indicated. The atom pair is exchange coupled with coupling strength J . (b) Spin qubit, i.e., a spin-1/2 with magnetic stability $D_0 > 0$. (c) Single-atom magnet $S > 1/2$ with an energy barrier $D_0 < 0$. (d) Substrate-mediated superexchange of an atomic pair. Atoms 1 and 2 are connected via the substrate wavefunction represented by a single effective orbital 3. $\ell_{1(2)}$ is the vertical distance from orbital 1 (2) to the effective orbital 3. (e) $t_{1(2)}$ represents the hopping amplitude from orbital 1 (2) to effective orbital 3 and Δ the site potential difference.

experimentally rules out the g -factor modulation as the main driving mechanism [22], leaving two remaining possibilities.

In this work, we show that although crystal field modulation can drive a spin, the resulting Rabi rates are approximately an order of magnitude smaller for the Fe-Ti system, where the most reliable experimental data are available. To alleviate this discrepancy between experiment and model, we suggest a driving mechanism that is entirely based on electric field modulation of the exchange coupling between Ti and Fe. We will demonstrate that this approach leads to Rabi rates in the range of 10–30 MHz, compatible with experimental data, and provides a better understanding of how spins on surfaces are driven by electric fields in ESR-STM.

Exchange-coupled spin pair. We first focus on a generic pair of two spins coupled on a surface via an isotropic Heisenberg interaction and subjected to an external magnetic field as indicated in Fig. 1(a). Throughout this work, we will discard the driving field provided by the exchange with the magnetic tip entirely, i.e., we consider the limit where the tip is far, and focus on the Heisenberg coupling between the two spins parametrized by J . One spin is either a spin $s = 1/2$ that does not have any anisotropy or a system with a ground-state doublet not separated by an anisotropy barrier (i.e., $D_0 > 0$); see Fig. 1(b). This renders it an ideal candidate for a spin-qubit system and, therefore, it can be efficiently driven using ESR-STM [20,25,41]. The other spin in the pair is a single-atom magnet with a large anisotropy barrier [42]; see Fig. 1(c). The simplest effective spin Hamiltonian to describe it is of the form $H_{D_0} = D_0 S_z^2$, where $D_0 < 0$ is the uniaxial anisotropy parameter, S_z is the spin projection of S along quantization axis z , and $S \geq 1$. To distinguish these two spins, we will denote the spin qubit with lowercase s and the single-atom

magnet with capital S throughout the remainder of the text. The Zeeman Hamiltonian for each spin reads $H_Z = g\mu_B \mathbf{B}_{\text{ext}} \cdot \mathbf{s}$ (or S), with μ_B the Bohr magneton and $\mathbf{B}_{\text{ext}}$ the external static magnetic field defined in polar coordinates $(B_{\text{ext}}, \theta_{\text{ext}})$.

The spin pair has a Hilbert space of the form $\mathcal{H} = s \otimes S$. In the following, we will employ the notations $|\uparrow\rangle$ and $|\downarrow\rangle$ for spin-qubit states, and $|\uparrow\rangle$ and $|\downarrow\rangle$ for single-atom magnet states. In this context, $|\uparrow, \uparrow\rangle \leftrightarrow |\downarrow, \uparrow\rangle$ corresponds to a spin-flip on the spin qubit, while the single-atom magnet remains unaffected. Conversely, $|\downarrow, \uparrow\rangle \leftrightarrow |\downarrow, \downarrow\rangle$ would indicate that the single-atom magnet has reversed its spin. A physical system corresponding to this spin pair consists of an iron (Fe, $S = 2$) as single-atom magnet and a titanium (Ti, $s = 1/2$) atom as spin qubit [27,37]. We will discuss the specific parameters for this system at the end. The static Hamiltonian of this spin pair is

$$H_0 = g\mu_B \mathbf{s} \cdot \mathbf{B}_{\text{ext}} + g\mu_B \mathbf{S} \cdot \mathbf{B}_{\text{ext}} + D_0 S_z^2 - J_0 \mathbf{S} \cdot \mathbf{s}, \quad (1)$$

where J_0 indicates the static exchange coupling of the two spins. The underlying physical mechanism in the systems considered here is substrate-mediated superexchange [43] which can be described by an effective three-orbital model, as illustrated in Fig. 1(d). In the limit of low-energy physics where the on-site repulsion U is the dominant energy scale, J_0 can be represented as [44,45]

$$J_0 = \frac{4|t_1 t_2|^2}{(U + \Delta)^2} \left(\frac{1}{U} + \frac{1}{U + \Delta} \right), \quad (2)$$

where $t_{1(2)}$ is the hopping amplitude for orbital 1 (2) and Δ is the on-site energy difference between the spin centers 1 and 2 and the mediating orbital 3, depicted in Fig. 1(e). A detailed derivation is presented in the subsection A of Ref. [46].

Modulation of the anisotropy and exchange term. The last two terms in Eq. (1) show two venues for modulation, D_0 and J_0 . We note that it has been demonstrated that dc electric control of D_0 and J_0 allows for full control of a spin-1/2 system under suitable conditions [47,48]. In the following, we want to show that an ac modulation will exhibit the same physics and will allow to directly drive s .

We first consider that an applied electrical field can modulate the crystal field of the adatom via the piezoelectric effect [49]. Assuming that the adatom is not entirely neutral or has a nonvanishing (induced) dipole moment, the electric field could either pull the atom away from the substrate or push it closer. This process has to be adiabatic [28,50], which is a reasonable assumption since the resonances (phonon modes) of the substrate-adatom are typically in the THz range, while ESR operates in the regime of a few tens of GHz. As a result, the ac voltage $\cos(\omega_0 t + \varphi)$ modulates the anisotropy in-phase, so the resulting time-dependent Hamiltonian is given by

$$H_{D_1}(t) = D_1 \cos(\omega_0 t) S_z^2, \quad (3)$$

where we have set $\varphi = 0$, which corresponds to a fixed axis for the rotation. Varying φ allows for full control of quantum state on the Bloch sphere [51]. To satisfy the resonance condition, the modulation frequency equals transition frequency between $|i\rangle = |\uparrow, \uparrow\rangle$ and $|j\rangle = |\downarrow, \uparrow\rangle$, in which the single-atom magnet remains in its spin-up state, i.e., $\omega_0 = \omega_{ij}$ (the spin-down state provides similar physics).

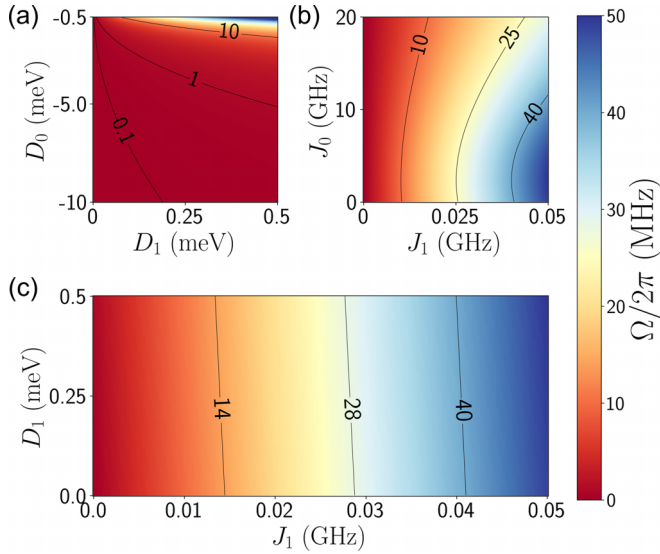


FIG. 2. Achievable Rabi rates from D , J , and DJ modulation models: (a), (b) Dependence of the Rabi rate by separate tuning of D and J on keeping the respective other parameters fixed. (c) Dependence of the Rabi rate on J_1 and D_1 while choosing D_0 and J_0 values compatible with the physical Fe-Ti system. Fixed parameters are as follows: (a) $J_0 = 5$ GHz and $J_1 = 0$ GHz; (b) $D_0 = -5$ meV and $D_1 = 0$ meV; and (c) $D_0 = -5$ meV and $J_0 = 5$ GHz. Parameters related to the external magnetic field are $B_{\text{ext}} = 0.9$ T and $\theta_{\text{ext}} = 80^\circ$. We use isotropic g tensor $g = 2$.

Second, if the exchange between adatoms is modulated by an electric field [52], a modulation of J_0 —given certain other geometric conditions—could provide a driving field with sufficient magnitude to drive ESR on the Ti atom given by

$$H_{J_1}(t) = -J_1 \cos(\omega_{ij}t) \mathbf{S} \cdot \mathbf{s}. \quad (4)$$

Using Eq. (2) we assume that the modulation of exchange coupling via electric field is induced by the variation of on-site potential Δ , which leads to $J_1 \propto J_0 V_{\text{AC}}$. We note that the linear relation holds as long as the electric energy is much smaller than on-site repulsion U , i.e., $e(\ell_1 + \ell_2)|\mathbf{E}|/U \ll 1$, where $\ell_{1(2)}$ is the distance from orbital 1 (2) to effective orbital 3 and e is the electron charge. This is valid for the typically small amplitudes of the ac voltage where the change in on-site energies remains negligible compared to the on-site repulsion energy in the doubly occupied orbitals; see subsection B in Ref. [46]. Finally, we emphasize that, although we are modulating the anisotropy of $\mathbf{S}$ and exchange coupling between the two spins, we are ultimately interested in the time evolution of $\mathbf{s}$.

Qualitative estimate of Rabi rates. We now aim to qualitatively predict Rabi rates of $\mathbf{s}$. We solve the full Hamiltonian $H_0 + H_{D_1}(t) + H_{J_1}(t)$ to obtain the time evolution of both $\mathbf{S}$ and $\mathbf{s}$. The Rabi rate Ω can be determined via the off-diagonal terms of time-dependent Hamiltonian, which read

$$\hbar\Omega = \langle \uparrow, \uparrow | (D_1 S_z^2 - J_1 \mathbf{S} \cdot \mathbf{s}) | \downarrow, \uparrow \rangle. \quad (5)$$

We first consider each driving contribution in Eq. (5) separately, namely D modulation and J modulation, for reasonable direction and magnitude of a magnetic field [37]. Figure 2(a) shows the Rabi rate solely from D modulation. To maximize

the Rabi rate, the ratio D_1/D_0 has to be maximized; i.e., the static contribution D_0 should be small relative to the modulation amplitude D_1 . However, in our model the limit of $D_0 \rightarrow 0$ is not valid as the system would not result in a magnet with large magnetic anisotropy energy and in any physical system D_0 is given by the material and adsorption site. The result associated with the J modulation in Fig. 2(b) shows a different trend. Here, a relatively small ratio J_1/J_0 is able to achieve Rabi rate values significantly larger than the ones obtained by the D modulation. This is further supported by Fig. 2(c), where we combined both DJ modulations for a given J_0 and D_0 , since the Rabi rate remains nearly unchanged along D_1 direction, while it changes dramatically along J_1 direction. Figure 2(c) implies that, in the context of J modulation, a magnitude able to explain the experimental data can be easily achieved or exceeded. Whilst each individual spatial component of the exchange modulation can drive the spin, the direction of the modulation that aligns with the anisotropy axis of the single-atom magnet will dominate as we analyze in Ref. [53].

Quantitative estimate of Rabi rates. We now apply our model to a physical system. To mimic the experiment, we will use parameters that are typical for a Fe-Ti pair on two monolayers of MgO on a silver substrate. Fe is a well-characterized single-atom magnet with $D_0 = -4.7$ meV [40]. For a small distance, the magnitude of J_0 depends exponentially on the separation between the two atoms on the surface [3]. In Ref. [37] exchange coupling values of $J_0 = 1.1 \pm 0.1$ and 6.8 ± 0.5 GHz were measured for $d = 0.72$ and 0.59 nm, respectively. These couplings were extracted from the splitting of the resonance frequencies considering the influence of the external magnetic field angle θ_{ext} , which effectively modifies the net exchange coupling between atoms. In the following, we will apply our model to a pair of $d = 0.59$ nm.

Regarding the driving contributions, we first assess the D_1 modulation of Fe on MgO/Ag. For details of the model, including calculations and experimental data, we refer to Ref. [12]. Here, it should suffice to calculate the modulation D_1 as a function of the displacement of the Fe adatom, i.e., $D_1(z)$, for which we performed a series of multiplet calculations using a multi-orbital Hubbard model [54]. Using typical rf field strengths of 0.1 V/nm, we expect a displacement of at most 0.05 pm, as shown in Ref. [55]. This leads to a maximal variation of $D_1 = 0.41$ μeV (≈ 100 MHz), contributing much less than 0.1 MHz to the Rabi rates, according to the result shown in Fig. 2(a). We note that in this displacement range, the relation between D and z is strictly linear with a proportionality of 0.107 meV/nm.

We have confirmed that the magnitude of J_1 required to achieve the experimental Rabi rates is reasonable in this system. Based on experimental data for the Rabi rate of Fe-Ti pairs, the magnitude of $J_1/\Delta E$ should be on the order of 20 MHz/ 0.1 V/nm [39]. This is compatible with results from our density functional theory (DFT) calculations on a magnetically coupled Fe-Fe and Fe-Ti pair adsorbed on the oxygen-top site of a MgO/Ag slab. More details of DFT calculations can be found in Ref. [55]. We note that on-surface exchange for Fe-Ti and Fe-Fe pairs might be highly anisotropic, considering the ground-state wavefunction of, for example, Ti [15,56]. Typical values for the field modulation

of J reported in literature for other systems are approximately 6–40 times larger than those required for Fe-Ti pairs, and can exceed them by several orders of magnitude for molecular magnets. A comparison for various magnetic systems is provided in Ref. [57]. Therefore, it is reasonable to assume that the order of J_1 in Fig. 2, from 5 to 50 MHz, is very plausible in our model. We also emphasize that, considering the results shown in Figs. 2(a) and 2(c), the D modulation from the piezoelectric displacement provides nearly zero driving on the spin qubit; however, we will retain it for consistency. Finally, we note that detuning of the resonance frequency $\Delta f = |\omega_0 - \omega_{ij}|$ increases the effective Rabi rate $\Omega' = \sqrt{\Omega^2 + \Delta f^2}$, although the amplitude of spin components is reduced to Ω/Ω' as expected, as shown in Ref. [51].

Angle dependence of the Rabi rate. Figure 3 presents the Rabi driving process of a Fe-Ti pair. We first illustrate the net magnetic field on the position of the Ti spin, $\mathbf{B}_{\text{net}} = \mathbf{B}_{\text{ext}} + \mathbf{B}_{0,\text{Fe}(\uparrow)}$, where $\mathbf{B}_{0,\text{Fe}(\uparrow)}$ is the static magnetic field induced effectively by the spin-up state of Fe. The static fields are drawn proportionally with their real magnitude so that the initial direction of the Ti spin, which is parallel to $\mathbf{B}_{\text{net}}$, is self-explanatory. Thus, $\mathbf{B}_{\text{net}}$ constitutes the quantization axis where the Hamiltonian of the Ti alone is diagonal. Figures 3(b) and 3(c) show the time evolution of Ti and Fe spins. In Fig. 3(b), we plot the corresponding Rabi rotations in the Bloch sphere under resonant condition, while Fig. 3(c) shows the evolution of the Fe under the same condition, remaining almost unchanged in its initial spin-up state. Nonetheless, the modulation also induces small oscillations in the Fe spin, as depicted in the inset of Fig. 3(c). Using realistic parameters (see figure caption), we obtain Rabi rate of Ti spin $\Omega/(2\pi) \approx 10$ MHz, which compares well to experimental values of 10 to 30 MHz, depending on the specific driving condition [37–39].

Figure 3(a) also illustrates the induced driving field $\mathbf{B}_{1,\text{Fe}(\uparrow)}$ from the spin-up state of the Fe. Its amplitude reads

$$\begin{aligned} B_{1,\text{Fe}(\uparrow)} &= B_{0,\text{Fe}(\uparrow)} \frac{J_1}{J_0} \cos(\theta_{\text{Ti}} - \pi/2) \\ &= \frac{\langle S_z \rangle J_1}{g\mu_B} \sin(\theta_{\text{Ti}}), \end{aligned} \quad (6)$$

where the polar angle of the initial Ti spin, θ_{Ti} , can be determined via the static magnetic fields as follows:

$$\theta_{\text{Ti}} = \cos^{-1} \left[\frac{B_{\text{ext}} \cos(\theta_{\text{ext}}) - B_{0,\text{Fe}(\uparrow)}}{B_{\text{net}}} \right],$$

where the minus sign in front of $B_{0,\text{Fe}(\uparrow)}$ originates from the negative sign in the exchange coupling term in Eq. (1). The strength of the driving field that determines the Rabi rate is the perpendicular projection of the driving field onto the Ti spin direction, as indicated by its proportionality to $\sin(\theta_{\text{Ti}})$. In this context, Eq. (6) highlights a crucial factor for enhancing the Rabi rate: its behavior with respect to the angle θ_{Ti} . This angle depends on the external magnetic field; thus, for a given exchange coupling J_0 , there is an optimal direction of the external magnetic field that enables the Fe to drive the Ti most efficiently. Notably, $\theta_{\text{ext}} = 0^\circ$ results in $\theta_{\text{Ti}} = 0^\circ$ and therefore, no driving field acts on the spin qubit, emphasizing

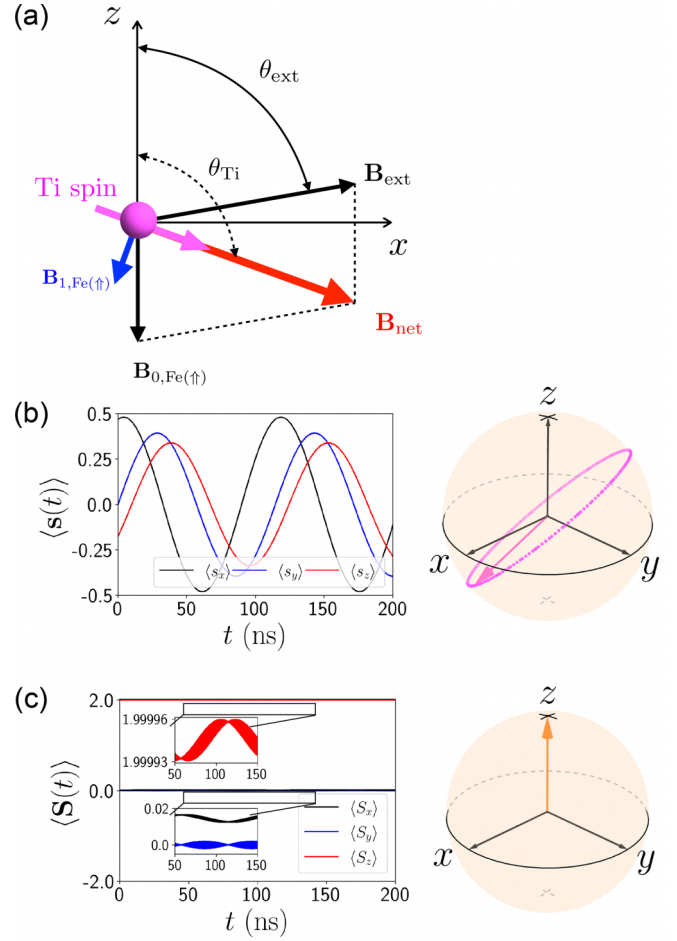


FIG. 3. Spin dynamics of a Fe-Ti pair using DJ modulation: (a) Illustration of the static net magnetic field composed of the external field $\mathbf{B}_{\text{ext}}$ and the field induced by the spin-up state of Fe, $\mathbf{B}_{0,\text{Fe}(\uparrow)}$, acting on the Ti spin. $\mathbf{B}_{1,\text{Fe}(\uparrow)}$ is the time-dependent driving field induced by Fe spin dynamics. Time dependence of (b) the Ti spin in the rotating frame and (c) the Fe spin and their representations on a Bloch sphere. The arrows indicate the initial spin configurations of Ti and Fe. As we can see, the Fe spin does not undergo any measurable time dependence. The transition frequency is $\omega_{ij}/(2\pi) \approx 26$ GHz, and the Rabi rate is $\Omega/(2\pi) = 9.41$ MHz. Simulation parameters: $D_0 = -4.7$ meV, $D_1 = 0.41$ μ eV, $J_0 = 6.8$ GHz, $J_1 = 0.01$ GHz. $B_{\text{ext}} = 0.9$ T, $\theta_{\text{ext}} = 80^\circ$. We employ isotropic g tensor $g = 2$.

the importance of selecting the proper external magnetic field angle.

To better illustrate the role of θ_{ext} , Fig. 4(a) presents a color map of the Rabi rate as a function of the exchange coupling J_0 and the external magnetic field angle θ_{ext} . Since $J_1 \propto J_0$, we use a fixed ratio for the modulation strength of $J_1 = J_0/680$, so that $J_0 = 6.8$ GHz results in a modulation of 10 MHz. The color map confirms Eq. (6): For a given static exchange coupling, the optimal magnetic field angle is the one that aligns the Ti at 90° , as indicated in Fig. 4(b), unless the magnetic field from the Fe is strongly dominating. In such scenarios, it is evident by Fig. 4(b) that the Rabi rate reaches its maximum value for an external field angle that aligns the Ti spin as close as possible to 90° . Since both J_0 and θ_{ext} are adjustable

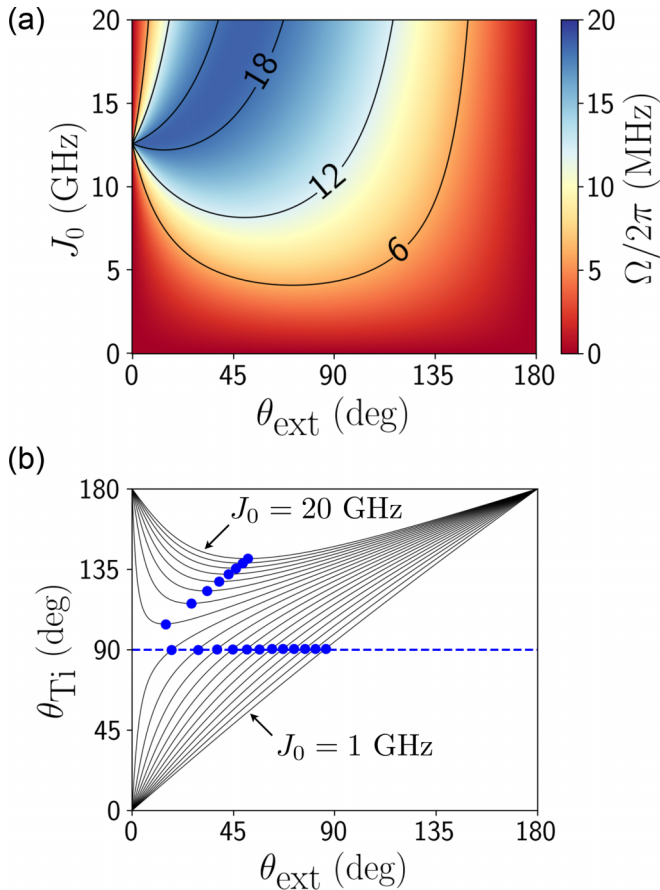


FIG. 4. Optimization of Rabi rates. (a) Dependence of Rabi rates on the static coupling strength J_0 and the external magnetic field angle θ_{ext} . Here we use a fixed ratio of $J_1/J_0 = 1/680$. (b) Dependence of the Ti spin's angle θ_{Ti} on the external magnetic field angle θ_{ext} . Each line corresponds to a fixed value of J_0 , ranging from 1 to 20 GHz in steps of 1 GHz. Blue dots indicate the maximum Rabi rates. The remaining parameters are $B_{\text{ext}} = 0.9$ T, isotropic g tensor $g = 2$, $D_0 = -4.7$ meV, and $D_1 = 0.41$ μeV .

parameters in the experiment, these findings open the door for optimizing the driving from a single-atom magnet. In practice, this has already been implemented experimentally, as it is often necessary to tune θ_{ext} to properly drive the spin qubit adjacent to the quantum magnet. An additional observation regarding Eq. (6) is the dependence on $\langle S_z \rangle$, suggesting that magnets with higher spin exhibit enhanced Rabi rates for a given modulation J_1 .

In summary, we have explored that in an atomic pair formed by a single-atom magnet and a spin-1/2 on a surface, ESR can be achieved by modulation of the crystal field anisotropy of the single-atom magnet or by modulation of the Fe-Ti exchange coupling. Modulation of the anisotropy alone results in Rabi rates that are 1–2 orders of magnitude smaller than those observed in the experiment [26,27,37]; however, even a small modulation of the exchange coupling results in a highly efficient Rabi process, with Rabi rates easily reaching tens of MHz in agreement with the experiment. We have also highlighted the significance of adjustable parameters, such as the static exchange coupling, the external magnetic field angle, and the spin of the magnet, in maximizing the Rabi rate. We therefore conclude that surface spin qubits built by combining a single-atom magnet and a spin-1/2 can be efficiently controlled purely by electrical fields that modulate the exchange coupling between the two spins.

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