Nonlocally sensing the magnetic states of nanoscale antiferromagnets with an atomic spin sensor

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The ability to sense the magnetic state of individual magnetic nano-objects is a key capability for powerful applications ranging from readout of ultradense magnetic memory to the measurement of spins in complex structures with nanometer precision. Magnetic nano-objects require extremely sensitive sensors and detection methods. We create an atomic spin sensor consisting of three Fe atoms and show that it can detect nanoscale antiferromagnets through minute, surface-mediated magnetic interaction. Coupling, even to an object with no net spin and having vanishing dipolar stray field, modifies the transition matrix element between two spin states of the Fe atom–based spin sensor that changes the sensor’s spin relaxation time. The sensor can detect nanoscale antiferromagnets at up to a 3-nm distance and achieves an energy resolution of 10 μeV, surpassing the thermal limit of conventional scanning probe spectroscopy. This scheme permits simultaneous sensing of multiple antiferromagnets with a single-spin sensor integrated onto the surface.

INTRODUCTION

Properties of quantum spin systems, such as magnetic stability (1) and spin coherence (2, 3), depend sensitively on their local conditions. Therefore, they can be used to great effect as sensors for magnetic environments. Great effort is directed toward increasing the spatial resolution and sensitivity of spin detection with a variety of techniques using quantum systems, such as magnetic resonance force microscopy (4, 5), scanning nano-SQUIDs (superconducting quantum interference devices) (6), and nitrogen vacancy (NV) centers in diamond (7–10), which have achieved nanometer resolution. A scanning tunneling microscope (STM) is capable of atomic resolution, but the direct interaction of the microscope tip can have an invasive influence on spins (11). Controllably placing the sensor in close proximity, ideally with atomic precision, is crucial and is an intensely studied problem for NV centers and other nanofabricated field sensors. This atomic–scale precision is routine in STM-based atom manipulation, albeit at cryogenic temperatures and on monocristalline surfaces (12, 13). Manipulation of magnetic adatoms allows the creation of quantum spin systems that can be addressed by single-atom magnetometry (14, 15), inelastic electron tunneling (16, 17), electronic pump-probe spectroscopy (1, 18), and electron paramagnetic resonance (19). Magnetic atoms on surfaces couple to their local environment via a broad range of interactions, for example, by exchange (20), superexchange (21), nonlinear exchange (22), or RKKY (Ruderman-Kittel-Kasuya-Yosida) coupling (15, 23); via Kondo scattering (24, 25); with electric (19) or magnetic fields; and to surface strain through magnetic anisotropy (17, 26). Therefore, adatom sensors have the potential to realize detection schemes via any of these coupling mechanisms.

RESULTS AND DISCUSSION

Here, we show that measuring the spin dynamics of a few-atom spin system permits it to function as a highly sensitive, surface-integrated sensor capable of detecting the presence and state of nearby magnetic nano-objects. Using a three-Fe atom chain (trimer), we can detect the switching of multiple nano-antiferromagnets via a substrate-mediated interaction (Fig. 1A). Both the trimer and the nano-antiferromagnets are assembled using a low-temperature STM with Fe atoms on a monolayer copper nitride (Cu2N) surface on a Cu(100) substrate (17, 25). The nano-antiferromagnets are constructed with an even number of antiferromagnetically coupled atoms so that any single one has two stable magnetic states, Néel states, with zero net spin. This is signified in spin-polarized STM topographs by alternating the apparent height of the constituent Fe atoms (Fig. 1B). These states also exhibit spontaneous switching (13). The solely internal magnetic structure and absence of a net magnetic moment makes state switching in antiferromagnets difficult to detect by conventional magnetometry (5–7).

The trimer was chosen as the inaugural sensor because it features an avoided level crossing in its two low-energy spin states at zero magnetic field (20). The crossing mixes the |+2 +2⟩ and | −2 +2⟩ states of the trimer (±2 denotes the expectation value of each Fe atom’s spin along the easy magnetic axis). This mixing permits transitions between the two states with a rate that is strongly dependent on magnetic field or any other local magnetic perturbation (Fig. 1C). A small magnetic field parallel to the z axis, B0 > 0, biases these two states so that the ground state, |⃗ν⟩, becomes mostly | +2 +2⟩ (see fig. S1), making it detectable by low–bias voltage, spin-polarized STM imaging (Fig. 1B; see Materials and Methods). Spin-polarized pump-probe measurements can then be used to pump the trimer to the excited state, |ν⟩, and probe the spin relaxation time, T1, back to the ground state (18). Monitoring variations in T1 reveals magnetic perturbations caused by changes in the trimer’s local environment (Fig. 1B).

Sensing the magnetic state of a nearby nano-antiferromagnet

With an external magnetic field of 0.25 T, the Fe trimer has an average spin relaxation time, T1, of 78 ns. We find that the Fe trimer switches between two different spin relaxation times as a nano-antiferromagnet constructed nearby switches between its two Néel states (see section S1). The decay curve measured on the Fe trimer yields a longer T1 when the nano-antiferromagnet is in Néel state “0” and a shorter T1 when it is in Néel state “1” (Fig. 1B). The difference in spin relaxation time, ΔT1, is 26 ± 3 ns measured for a separation of 3 nm between the...
Fe trimer and the nano-antiferromagnet. This demonstrates that the Fe trimer can measure the magnetic state of a nearby antiferromagnet and act as a spin sensor.

This capability allows us to detect spontaneous switching of the nano-antiferromagnet by continuously monitoring the Fe trimer’s spin relaxation time. When the nano-antiferromagnet switches from one Néel state to the other, the electronic pump-probe signal jumps between two decay curves (Fig. 1B). A trace of this two-state switching can be obtained by keeping the pump-probe delay fixed and measuring the pump-probe signal amplitude (Fig. 1D). Large pump pulse amplitudes induced additional switches in these traces (see fig. S2). To minimize this effect, we used minimal amplitude pump pulses (10 mV). The amplitude of the two-state signal depends on the delay time chosen. Increasing the delay time from 150 to 250 ns reduces the signal, and
it vanishes when the Fe trimer’s spin is fully relaxed at a delay of 500 ns (Fig. 1D). This further confirms that the two-state switching is encoded in the dynamic response of the Fe trimer.

The variation in $T_1 (\Delta T_1)$ can be increased by reducing the separation of the trimer and the nano-antiferromagnet. The measurements investigating the Fe trimer-nano-antiferromagnet separation were performed with the same nano-antiferromagnet on the same copper nitride patch, by simply moving the Fe trimer to different locations by STM atom manipulation (Fig. 2A). $\Delta T_1$ becomes larger as the separation between the nano-antiferromagnets and Fe trimer decreases (Fig. 2B): At 3.0-nm separation, $\Delta T_1$ is 29 ns and increases to 466 ns at 1.1-nm separation.

**Exploring the sensing mechanism**

To understand the sensing mechanism, we inspect the spin relaxation process, which occurs predominantly by electron-spin scattering (18, 20). $T_1$ is determined by the product of the transition matrix element between the trimer’s two low-energy spin states, $|\phi\rangle$ and $|\rho\rangle$, and the number of substrate electrons that can scatter (see section S2) (20). It is minimal at zero magnetic field, where transverse magnetocrystalline anisotropy axis of the Fe atoms creates an energy splitting, $E_{\text{an}}$, between the two low-energy states, reduces state mixing, and increases $T_1$ to a detectable level (Fig. 3A). This magnetic bias field sets a working point within the Fe trimer’s two-level system. All other magnetic perturbations introduce an additional variation in the energy splitting, $\Delta E$, that leads to a variation in the spin relaxation time, $\Delta T_1$.

We use a spin Hamiltonian model to calculate the response of $T_1$ to different magnetic perturbations, including a longitudinal magnetic field ($B_||$), transverse magnetic fields ($B_\perp$) that add to the bias field ($B_0$), and a Heisenberg-type exchange interaction ($J_{nAF}$) with the nano-antiferromagnet (Fig. 3B) (see section S2). We find that $T_1$ is very sensitive to $B_||$ and $J_{nAF}$, but it is not sensitive to $B_\perp$. This rules out the possibility that the changes in $T_1$ of the Fe trimer are induced by transverse dipolar magnetic fields from the nearby nano-antiferromagnet.

Notably, the variation of the spin relaxation time, $\Delta T_1$, with the perturbation energy, $\Delta E$, induced by $B_||$ and $J_{nAF}$ is expected to be linear over a large range of perturbation magnitudes. We use an external superconducting vector magnet to apply the bias field and an additional perturbation field in the same direction, $B_0 + B_||$. The perturbation energy of $B_||$ is given by the Zeeman energy of the Fe trimer, $\Delta E = 4\mu_B B_||$ (the magnetic moment of the Fe trimer is $4\mu_B$) so that we can plot $\Delta T_1$ as a function of perturbation energy, $\Delta E$, for each experimental configuration. We find that $\Delta T_1/\Delta E (B_||)$ is linear (Fig. 3B).

The two curves, $\Delta T_1/\Delta E (B_||)$ and $\Delta T_1/\Delta E (J_{nAF})$, are almost identical (Fig. 3B). This similarity permits us to use the external magnetic field as a quantitative reference to deduce the interaction energy between trimer and nearby nano-antiferromagnet from the observed changes in $T_1$ induced by Néel state switching (see section S3 and fig. S3). We verified

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**Fig. 2. Distance dependence of nonlocal spin sensing.** (A) Spin-polarized STM topographs of different separations, $d$, between Fe trimer and nano-antiferromagnet. Tunnel junction set point, 5 mV and 20 pA. (B) Pump-probe measurements on the Fe trimer for each arrangement shown in (A), as the nano-antiferromagnet is in Néel state “0” (red dots) or “1” (blue dots). The pump-probe signal is normalized to 1 at zero delay time for clarity. Solid lines are exponential fits. Pulse amplitude and duration: pump pulse, 8 mV and 80 ns; probe pulse, 3 mV and 100 ns. Magnetic bias field during measurement, 0.25, 0.5, 0.75, and 0.75 T (left to right). (C) Magnetic interaction energy between Fe trimer and nano-antiferromagnet for the arrangements shown in (A) as a function of sensor-antiferromagnet separation (blue points). The blue line is an exponential fit to the measured interaction energies. The black line is the calculated magnetic dipolar interaction between the Fe trimer and nano-antiferromagnet.
that the choice of magnetic bias field \( B_0 \) has no influence on the responsivity of the Fe trimer by finding that \( \Delta T_1 \) induced by Néel state switching is constant over a large range of bias fields for all nano-antiferromagnet–sensor arrangements that we studied (Fig. 3C), although the absolute magnitude of the Fe trimer’s lifetime changes significantly with bias field (Fig. 1C). This also demonstrates that perturbations from magnetic fields and exchange interaction add linearly. It is worth noting that, at large negative \( \Delta E \), the variation of \( T_1 \) levels off and reverses when the perturbation is sufficiently large to compensate the bias field and cause \( |\psi\rangle \) and \( |\phi\rangle \) to cross (Fig. 3B). In all measurements, we kept the bias field large enough to avoid this region.

This makes it possible to deduce a simple empirical equation to quantitatively extract the magnetic interaction energy between the Fe trimer and the nano-antiferromagnet from measured values of \( \Delta T_1 \). Because the nano-antiferromagnet is in Néel state “1,” the magnetic interaction with the Fe trimer will decrease the energy difference between \( |\psi\rangle \) and \( |\phi\rangle \) by \( -\Delta E/2 \), whereas it will increase by \( \Delta E/2 \) for Néel state “0” (Fig. 3A). Because \( T_1 \) is linearly dependent on both \( B_0 \) and \( J_{\text{nAF}} \), and \( \Delta T_1/\Delta E (B_0) \) and \( \Delta T_1/\Delta E (J_{\text{nAF}}) \) are almost identical, we can express magnetic interaction in terms of the Zeeman energy of \( B_|| \) and obtain the empirical relation

\[
\Delta E = \frac{4 g \mu_B}{\frac{\Delta T_1}{\Delta B_0}} \Delta T_1 \tag{1}
\]

where \( \mu_B \) is the Bohr magneton and \( g \) is the Fe trimer’s \( g \) factor (\( g = 2 \)). \( \frac{\Delta T_1}{\Delta B_0} \) is the responsivity of \( T_1 \) to longitudinal magnetic field and was...
obtained experimentally by varying the magnitude of $B_0$. It ranges from 450 to 830 ns/T for Fe trimers in different locations on the Cu$_2$N surface (see fig. S3). Figure 2C shows the extracted magnetic interaction as a function of the separation between the Fe spin sensor and nano-antiferromagnet. To remove possible influence from variations in the crystal-field environment of the sensor, we performed the calibration of $(\frac{dN}{dF})$ for each sensor--nano-antiferromagnet distance. The magnetic interaction energy increases quickly from 12 to 224 μeV as the separation decreases from 3.0 to 1.1 nm.

This interaction is surprisingly long-ranged when compared to the previously reported superexchange interaction on the Cu$_2$N/Cu(100) surface (27, 28). Dipolar magnetic interaction can be calculated from the known positions of all Fe atoms in the two structures but accounts only for ~5% of the measured interaction energy because the structures' antiferromagnetic order cancels most dipolar fields (black line in Fig. 2C). This suggests the presence of an indirect exchange interaction through the Cu$_2$N/Cu(100) surface. Long-range indirect exchange interaction has thus far been found only for adatoms on metallic surfaces, with a surface state that mediates oscillatory RKKY-type interaction (15, 23). This interaction is absent for adatoms on Cu(100) (29, 30), but the influence of the Cu$_2$N decoupling layer has not been considered yet.

We therefore performed ab initio all-electron density functional theory (DFT) calculations to elucidate the microscopic origin of this long-range indirect exchange interaction mechanism. Because of the long-range nature of the interaction, we cannot treat large isolated objects deposited on the surface in our supercell calculation approach. Consequently, we modeled the experimental geometry by one Fe atom ladder (to simulate the nano-antiferromagnet) and one Fe atom chain (to simulate the Fe trimer) (see Materials and Methods and the Supplementary Materials for details). The magnetic interaction energy between the Fe ladder and the Fe chain was calculated as the total energy difference between their ferromagnetic (state “1”) and antiferromagnetic (state “0”) alignment (Fig. 4A, inset) within the broken symmetry approach. We find antiferromagnetic interaction between the Fe ladder and the Fe chain, and the interaction strength decreases with distance, in agreement with the experiment (see Fig. 4A). To gain further insight into the long-distance limit (beyond 1.45 nm) of this interaction, we approximated the Fe ladder by a Fe chain, which reduced the supercell size and enabled calculations up to 2.6-nm chain-chain separation. Remarkably, the decay trend of the magnetic interaction is comparable to the experiment: It is antiferromagnetic and nonoscillatory and decays with a characteristic length scale of approximately 1 nm (Fig. 4A). Some deviations from an approximate exponential trend are found for the smallest interchain distance. However, this can be attributed to finite size effects in the calculations (see the Supplementary Materials for details).

**Fig. 4. Calculated long-range magnetic interaction.** (A) Magnetic interaction energy between a Fe nano-antiferromagnet (blue atoms in inset) and a Fe chain sensor (orange atoms in inset) calculated by DFT using a broken symmetry approach (see Materials and Methods for details). Energies are calculated analogous to Fig. 3A. Within the DFT calculation, the Fe trimer sensor was approximated by an infinite one-dimensional chain of Fe atoms and the nano-antiferromagnet by either an infinite ladder (purple squares) or another one-dimensional chain (blue dots). In the latter approximation, a significantly smaller supercell could be used. This enables calculations for large distances but overestimates interaction energies because of the absence of the second antiferromagnetically aligned chain in the nano-antiferromagnet (see fig. S6 for supercell geometries). (B) Density of states of the Cu$_2$N surface projected on the Cu atoms (orange) and N atoms (blue). Energies are given with respect to the Fermi energy. States between −4.5 and −1.5 eV are localized on Cu and have $d$-orbital character. States between −6 and −4.5 eV and between −1.5 and +0.5 eV are $p$-$d$ hybrids between Cu and N of $\alpha$ and $\beta$ character. In particular, the antibonding $\alpha$ bands give a sharp peak at −1 eV, and the $\beta$ bands extend across the Fermi level. (C) Top: Spin density distribution for two Fe atom chains at a 1.1-nm distance. Isosurfaces show $10^{-3} e/Å^2$ majority spin (red) and minority spin (blue) densities that extend well beyond the Fe atoms into the Cu-N network. Bottom: Side view of the DFT supercell on the same scale as the top panel, showing Fe atoms (red spheres), Cu atoms (orange spheres), and N atoms (blue spheres).
We note that our DFT calculations do overestimate the magnitude of the interaction. This can be attributed to three effects: first, the used exchange-correlation density functional (specifically the generalized gradient approximation) that tends to delocalize the electronic wave functions; second, the small structural uncertainties in the vertical position of the Fe atoms above the Cu$_2$N; third, the lateral periodic boundary conditions that result in calculating Fe chains of infinite length instead of a finite nano-object. All three effects likely increase the calculated magnetic interaction energy compared to the experiment. We remark that both the sign and decay behavior of the magnetic interaction are predicted accurately.

From our DFT calculations, we can unambiguously link the long-range magnetic interaction to the specific electronic properties of the surface. Hybridization between the Cu-$d$ and the N-$p$ orbitals in the Cu$_2$N monolayer results in bonding and antibonding bands similar to bands reported for Cu$_3$N films (see Fig. 4B) (31). The bonding bands are 6 eV below the Fermi energy, $E_F$, but the antibonding bands extend from approximately 1 eV below $E_F$ up to and across it. These bands are the source of the long-range magnetic interaction.

The antibonding Cu-N bands have either $\sigma$ or $\pi$ character. We find that the $\sigma$ band is mostly localized 1 eV below $E_F$, whereas the $\pi$ band extends over a wider energy range because of broadening induced by hybridization with the Cu substrate. The parallel Fe atom chains give rise to an electronic confinement in the Cu$_2$N layer that generates spin-polarized confined states stemming from these $\sigma$ and $\pi$ bands. The degree of confinement and the spin polarization depend critically, and nontrivially, on the energy position of the Fe-$d$ orbitals with respect to the $\sigma$ and $\pi$ bands that the $d$ orbitals are allowed to hybridize with by symmetry. The resulting quantum well states are ultimately responsible for the interaction between the chains. This turns out to be antiferromagnetic and, most importantly, long-range over a scale of 1 nm, as illustrated in Fig. 4C, which displays the spin density profile and its decay in between two Fe atom chains.

To prove the key role that the Cu-N network plays in setting the long-range magnetic interaction, we repeated the calculation breaking the Cu-N network by removing one row of N atoms between the two Fe atom chains. Now, the energy difference between the Néel states “1” and “0” drops below the numerical accuracy. In this case, the only way to achieve magnetic interaction is via the Cu substrate, but this interaction is expected to be much smaller, short-range, and ferromagnetic (29). We were able to detect weak ferromagnetic interaction (~12 ± 3 μeV at a 2.1-nm distance) across two Cu$_2$N patches experimentally by using a Fe trimer and a large 18-atom nano-antiferromagnet (Fig. 5).

It is worth noting that the magnetic interaction through $\sigma$ and $\pi$ states found for the Cu$_2$N monolayer bears similarities to exchange coupling between the opposite edges of zigzag graphene nanoribbons, where confined states originating from the carbon $\pi$ band near the Fermi energy also cause exponentially decaying antiferromagnetic interaction (32, 33).

**Achievable sensitivity**

The weakest magnetic interaction that is still detectable by the Fe trimer is determined by the smallest $\Delta T_1$, that can be measured above the noise of the pump-probe measurement. The pump-probe measurements transduce $\Delta T_1$ into a variation of the number of electrons tunneling during the probe pulses so that the signal can be optimized by adjusting the probe pulse duration to be approximately equal to $T_1$. Increasing the pump pulse amplitude or duration did not improve the signal but could induce additional switching of the nearby nano-antiferromagnets (see fig. S2). We therefore kept the pump pulses minimal. The time traces measured with the Fe trimer at a 3.0-nm distance from the nano-antiferromagnet have a signal amplitude of 0.55 e$^-$/pulse (electrons per probe pulse) and a noise level of 0.11 e$^-$/pulse for the chosen integration time of 0.1 s (Fig. 1C). This measurement indicates a magnetic interaction strength of 12 μeV and has a signal-to-noise ratio of 5. Hence, the sensitivity of this particular measurement was 0.9 μeV/√Hz, which relates to an equivalent magnetic field sensitivity of 3.8 mT/√Hz. This sensitivity is sufficient that single spins could be detected at a distance of 2 nm exclusively via dipolar fields with 60-s integration time. This method surpasses the energy resolution of conventional scanning tunneling spectroscopy. This is possible because it measures the dynamics of the few-atom quantum magnet rather than static spectral features that are thermally broadened by 150 μeV for elastic and 230 μeV for inelastic tunneling spectroscopy at 0.5 K.

Fundamentally, the sensitivity of this spin sensing scheme is limited by the shot noise of the tunneling electrons that probe the Fe trimer. Leaving all experimental parameters the same, a shot-noise limited detection would yield a sensitivity of 27 neV/√Hz (110 μT/√Hz). This...
Simultaneously sensing the magnetic states of multiple nano-antiferromagnets

The sensitivity that we achieved allows monitoring of several antiferromagnets within the detection range of one Fe trimer spin sensor. As a test, we assembled one 12-atom and one 10-atom nano-antiferromagnet around a Fe trimer (Fig. 6A). Each has two Néel states, giving four distinct configurations [labeled (0, 0), (0, 1), (1, 0), and (1, 1) in Fig. 6A].

The intrinsic lifetime of each state is sufficiently long that we can identify each of the four magnetic configurations and correlate each with a distinct $T_1$ of the Fe trimer by interleaving a series of pump-probe measurements with topographic imaging (Fig. 6B). Time traces of the pump-probe signal detect four-state switching, demonstrating that the Fe trimer spin sensor can simultaneously detect the states of two nano-antiferromagnets (Fig. 6C).

Thereby, we overcome a fundamental limitation of STM measurements: the single-point measurement. Typically, information gained by the STM is local and limited to the single location of the STM tip. By using the Fe trimer spin sensor, we can simultaneously interrogate nano-objects at two different locations using a single probe and perform nonlocal measurements that give access to spin-spin correlations. These measurements are difficult to achieve by using conventional spin-polarized STM because scanning the probe tip over the nano-antiferromagnets would perturb their state population too much to reveal weak correlations.

We can test for these correlations with a histogram of the four-state switching signal measured for more than 1 hour (see fig. S5). It reveals...
the probability, \( P \), for each of the four states to occur (Fig. 6D). This probability distribution contains the probability with which the ferromagnetic and antiferromagnetic configurations of the two nano-antiferromagnets occur. We find

\[
\frac{P_{(0,1)} + P_{(1,0)}}{P_{(0,0)} + P_{(1,1)}} = 1.12 \pm 0.09
\]

where \( \pm 0.09 \) is the standard error (\( \pm 1 \sigma \)) resulting from uncertainty in \( P \) because of the finite measurement duration of 4000 s. The value of 1.12 hints at a small but measurable antiferromagnetic correlation between the two nano-antiferromagnets.

**CONCLUSION**

In conclusion, we have demonstrated that few-atom spin systems on surfaces can be used as an atomic-scale spin sensor to sense the magnetic state of single or multiple nano-antiferromagnets with micro–electron volt energy sensitivity. In addition to surpassing the energy resolution of conventional STM spectroscopy, the sensing scheme mitigates the dynamic and invasive influence of the tip on the sensed object. Here, we applied the sensing scheme to quantify the distance-dependent magnetic interaction between a nano-antiferromagnet and a Fe trimer sensor on a Cu\(_2\)N surface. The interaction was found to be long-range and antiferromagnetic. DFT calculations identify that the spin-dependent confinement of the \( \sigma \) and \( \pi \) bands of the Cu\(_2\)N surface is the source for this antiferromagnetic interaction. This Cu\(_2\)N substrate–mediated antiferromagnetic interaction presents an intriguing possibility to use artificial arrays of magnetic adatoms on Cu\(_2\)N for studies of magnetic interaction across (quasi-) two-dimensional \( \pi \) electron systems and to explore some functionalities that have been originally proposed for two-dimensional materials, such as graphene nanoribbons (33).

The submicro–electron volt sensitivity of the sensing scheme enables indirect measurements of single-molecule magnets (34) that are difficult to address by direct tunneling spectroscopy and of ferromagnetic nanomagnets (35, 36) through their large, dipolar stray field. These measurements do not require coherent control of the spin sensor and can, in principle, be implemented on other sample systems, such as metallic (14), semiconducting (37), and thin insulating film (38) surfaces, and with other few-atom sensors that feature two spin states at low energy. The ability to simultaneously sense the magnetic states of multiple nano-antiferromagnets enables atomic-scale studies of spin-spin correlations for classical and quantum magnetic objects.

**MATERIALS AND METHODS**

**Experimental design**

This experiment aimed to demonstrate the possibility of creating integrated adatom spin sensors that allow spin-polarized STM to make highly sensitive, but noninvasive, measurements of other adatom magnetic structures, including those with no net spin, on a surface. A series of experiments were devised to show an adatom chain coupled to a nano-antiferromagnet and to explore this coupling by tuning separation and magnetic field. An additional experiment was designed, placing the sensor in proximity with two nano-antiferromagnets, to show the potential for sensing multiple magnetic adatom structures.

All experiments were conducted using a low-temperature and ultrahigh-vacuum STM equipped with a 2-T vector magnetic field (Unisoku USM1300 \(^3\)He). For all the measurements, the temperature was maintained at 0.5 K, and the external magnetic field was aligned to the easy magnetic axis of Fe atoms in Fe trimer spin sensor. The easy axis was parallel to the direction of the two nitrogen atoms neighboring each Fe atom, and the alignment accuracy of the magnetic field was \( \pm 3^\circ \).

The nano-antiferromagnet switched spontaneously between two Néel states (labeled “0” and “1”) but at a sufficiently low rate that allowed topographic imaging (typical duration, 20 s) and pump-probe spectroscopies (typical duration, 60 s) to be acquired without a switch occurring during the acquisition. For measurements that required the nano-antiferromagnet to be in a particular state (Figs. 1B, 2B, 3C, and 6B), we acquired a fast topography (10 s) before and after to verify that the nano-antiferromagnet did not switch. To attain statistical significance, these measurements were repeated at least 10 times.

**Sample preparation**

PtIr tips were sputtered with argon and flashed by e-beam bombardment for 10 s before use. The Cu(100) crystal was cleaned by several Ar-sputtering and annealing (850 K) cycles. After the last sputtering and annealing cycle that creates a clean Cu(100) surface, the monatomic copper nitride, Cu\(_2\)N, layer was prepared by nitrogen sputtering at 1 kV and annealing to 600 K. Then, the sample was precooled to 4 K, and Fe atoms were deposited onto the cold sample by positioning it in a low flux of Fe vapor from a Knudsen cell evaporator.

The Fe trimer spin sensor was built by positioning Fe atoms 0.72 nm apart on the Cu binding sites of the Cu\(_2\)N surface by vertical STM atom manipulation (13). The Fe trimer was built along the direction of the easy axis of Fe atoms in it. The nearby antiferromagnets were assembled from Fe atoms by the same technique. The spin-polarized tips were prepared by picking up three to four Fe atoms to the apex of the tip, which yielded spin polarization \( \eta \approx 0.1 \) to 0.3 (calibrated with 2-T external magnetic field). Spin-polarized topographic images were acquired using a bias voltage below the 7.5-mV inelastic excitation thresholds of the trimer and nano-antiferromagnets to avoid inelastic excitation of either structure.

**All-electronic pump-probe measurement**

An all-electronic pump-probe method was used to measure the spin relaxation time of the Fe trimer (18). A sequence of alternating pump and probe voltage pulses was created by a pulse pattern generator (Agilent 81110A) and sent to the sample using semirigid coaxial wires. The pump pulses excite the Fe trimer by inelastic scattering of tunneling electrons. The probe pulses detect the spin state of the Fe trimer because the tunnel magnetoresistance differs when the Fe trimer is in the excited state versus the ground state. Tunnel current resulting from the probe pulses was measured by lock-in detection at 690.6 Hz.

For the measurements shown in Figs. 2B, 3C, and 6B, the probe pulses were modulated on and off. This method removes the tunnel current contribution of the pump pulses from the lock-in signal. For the measurements shown in Figs. 1 (B and C), 5B, and 6C, the time delay between pump and probe pulses was modulated. This method removes any tunnel current contributions that are not due to time-dependent dynamics and records background-free time traces of the pump–probe signal (18). The average dynamical evolution of the Fe trimer was measured by slowly varying the time delay between pump and probe pulses (\( \Delta t \)). For increasing delay time, the probability of Fe trimer still being in the excited state decreases exponentially. The spin relaxation time, \( T_1 \), was determined as the decay constant of an exponential decay function fitted to the delay time–dependent tunnel relaxation time data.
current I(Δt). The typical time for taking the pump-probe spectrum (like Figs. 1B and 2B) and STM topograph is about 2 or 3 min.

DFT calculations
All-electron DFT calculations were performed with the Fritz Haber Institute ab initio molecular simulations package (FHI-aims) code (39). The Perdew-Burke-Ernzerhof (40) generalized gradient approximation to the exchange-correlation density functional was used. FHI-aims uses numerical atom-centered orbitals as the basis set. Here, the “light” settings, which include the tier 1 basis set for all atoms, were chosen. Nevertheless, for some selected cases, it was checked that energy differences changed by less than 1 meV per supercell when the tier 2 basis set was used. This value (1 meV per supercell) was then fixed as limit to the numerical precision of total energy difference calculations, and only results that satisfied this precision requirement are presented. Consistently, for each calculation, the convergence of the results with respect to the number of k points was also carefully checked to ensure the same precision. Relativistic effects were included by means of the atomic zero-point energy reference. Atomic zero-point energy reference.

Electron structure and not to deviations in the Fe atom

REFERENCES AND NOTES


Acknowledgments: We thank E. Simon for fruitful discussions and E. Weckert and H. Dosch (Deutsches Elektronen-Synchrotron, Germany) for providing laboratory space. Funding: J.A.J.B. acknowledges postdoctoral fellowships from the Alexander von Humboldt Foundation and the Natural Sciences and Engineering Research Council of Canada. A.D. and A.R. were supported by the European Research Council project QSpec-NewMat (ERC-2015-AdG-694097), the Marie Skłodowska-Curie project SPINMAN (SEP-210189940), and Grupos Consolidados UPV/EHU (Universidad del País Vasco/Euskal Herriko Unibertsitatea) (IT578-13). Author contributions: S.L. conceived the experiment. S.Y., L.M., and J.A.J.B. carried out the experiments. S.Y. and S.L. performed the analysis and calculations. A.D. and A.R. carried out the DFT calculations. All authors participated in the discussion of the results and contributed to the manuscript. Competing interests: The authors declare that they have no competing interests. Data and materials availability: All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

Submitted 15 December 2016
Accepted 27 March 2017
Published 26 May 2017
10.1126/sciadv.1603137

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Sci Adv 3 (5), e1603137.
DOI: 10.1126/sciadv.1603137