

Scanning tunneling microscope measures the spin-excitation spectrum of atomic-scale magnets

A proof-of-principle demonstration sets the stage for creating designer magnets, from simple Heisenberg spin chains to exotic spin ices.

The up-or-down spin of an electron makes it a natural qubit candidate for use, someday, in a quantum computer. Circuit logic based on the flipping of spins would require less energy and suffer far less heat dissipation than that based on the switching of charge. The challenge is to prepare the spins, flip them on cue, and then reliably read out their final states within a system—all while minimizing decoherence¹ (see PHYSICS TODAY, March 2006, page 16).

Advances in the fabrication of ever-smaller structures are driving research into understanding how to meet at least part of that challenge. At atomic length scales, small numbers of spins can be coupled directly in transition-metal clusters or one-dimensional chains. Such tiny magnets are typically produced using wet chemistry or dry self-assembly. Neutron scattering, electron paramagnetic resonance, susceptibility measurements, and other diagnostics provide details about their magnetic properties, including how strongly the spins couple to each other—that is, their exchange energy J . But those standard techniques average over a huge ensemble

of clusters and miss local variations in spin interactions.

IBM researchers Cyrus Hirjibehedin, Christopher Lutz, and Andreas Heinrich now offer a more direct approach to measuring the coupling strength: Probe the interactions in a single magnet, not a macroscopic collection of them.² The researchers, who are based at IBM's campus in Almaden, California, incrementally ramp up the voltage on a scanning tunneling microscope (STM) tip positioned over an individual cluster of atoms. At some threshold, the voltage, and hence the electron's kinetic energy, becomes high enough to flip the cluster's collective spin. The energy difference in spin configurations provides a direct measure of J .

To demonstrate the approach, Heinrich's team built a simple spin chain composed of 10 manganese atoms and systematically measured how the spin excitation spectrum changed as each new atom was added. Reassuringly, different chain lengths—monomer, dimer, trimer, and so forth—exhibited the properties predicted by the Heisenberg spin model, whose Hamiltonian is sim-

ply the sum over nearest-neighbor spin interactions parameterized by J .

Spin flips

In 1993, Donald Eigler and his IBM colleagues arranged iron atoms into a circular ring—the now iconic quantum corral—and imaged the dramatic interference pattern that is an exact experimental solution of the Schrödinger equation. In 2000, his group built a similar ring—the quantum mirage—by positioning magnetic cobalt atoms in an ellipse with an additional Co atom at one of the ring's two foci. The interaction of cobalt's magnetic moment with tunneling electrons in the copper surface produced the illusion of a magnetic atom at the other focus. And two years ago, they developed a spectroscopy technique for flipping the spin of a Mn atom in a magnetic field.³

The new experiment follows naturally from those atom-manipulation projects. And it highlights the exquisite control and sensitivity an STM can bring to bear. The energy to flip an atom's spin is some 1/10 000 the energy of a single molecular bond. To spectroscopically resolve separate spin and magnetic quantum-number states, S and m_s , Heinrich and company relied on an exceedingly stable STM. The instrument sits in ultrahigh vacuum at a

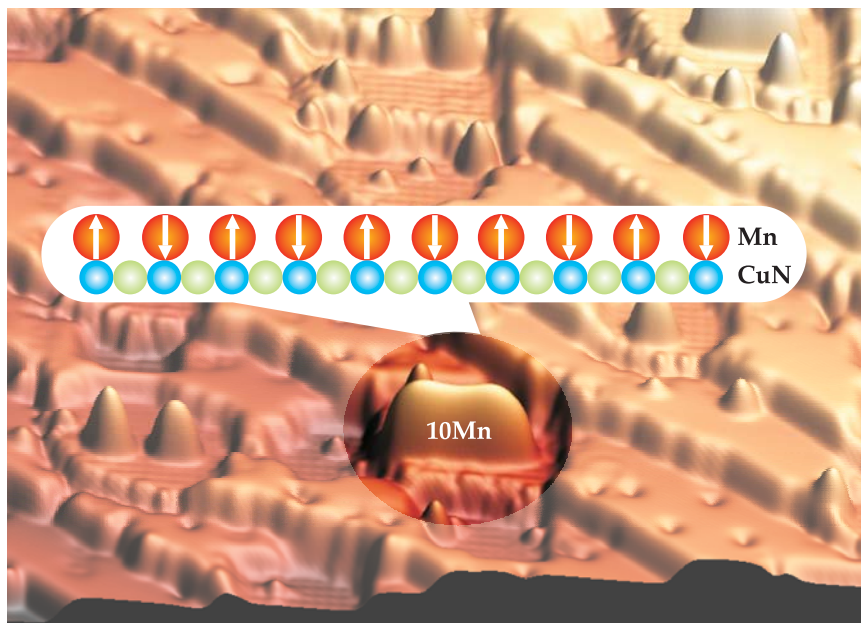


Figure 1. A scanning tunneling microscope image of manganese atoms evaporated onto a surface composed of patches of insulating copper nitride interspersed with metallic copper; the CuN appears as slightly recessed regions amid the smooth flat Cu terraces. On this terrain, the IBM group fashioned Mn spin chains 1–10 atoms long. The highlighted chain is one example. Its indistinct double-peaked structure suggests the presence of delocalized electronic states shared among the 10 atoms. The schematic (inset) pictures antiferromagnetic Mn spins, collapsed into their classical Néel state. (Courtesy of Cyrus Hirjibehedin.)

temperature of 0.6 K and in magnetic fields up to 7 T, with the tip held motionless to within a hundredth of an angstrom.

On copper, the STM tip can gently push or pull atoms into place electrostatically. But preventing the magnetic moments of atoms on the chain from mixing with copper's large number of conduction electrons requires an insulating monolayer. Bombarding the Cu with nitrogen atoms and heating the surface did the trick (see figure 1). But strong bonding precluded the team from nudging Mn atoms into place. Instead, postdoc Hirjibehedin resorted to using the STM as a crane, picking up the atoms with a voltage pulse from the tip and then depositing them in place onto the copper nitride insulator with another voltage pulse.

Manganese atoms in small chains prefer to form a quantum antiferromagnet: Odd-numbered chains display a net magnetization while even-numbered ones don't. To measure the explicit effect of parity on the coupling among spins, the researchers injected electrons into the tunnel junction formed between the tip and substrate for each length of chain. At low voltages, tip electrons tunnel elastically through the Mn chains into the Cu substrate and the differential conductance dI/dV remains predictably constant. At high enough voltages, though, tunneling injects enough energy to scatter electrons inelastically and trigger an excitation in the collective spin state of the chain. The signature of the transition is a step in the differential conductance.

Individual spins are often represented as classical objects, either up or down, but each one is more accurately a linear combination of the different configurations, with the wavefunction extending over the entire chain. The spectra in figure 2 bear out that collective behavior: The excitation energies depend on how many atoms are in the chain. In each case, the tip is positioned at the center of the chain. Significantly, though, the conductance spectra did not change when measured at different locations.

For a single atom, a spin flip excitation changes the magnetic quantum number m_s , the relative orientation of the spin, but leaves the total spin S unchanged. At zero magnetic field, where m_s is degenerate in energy, the conductance dip at zero voltage is unexpected and intriguing. Heinrich attributes the feature to the atoms' loss of rotational symmetry on a surface.

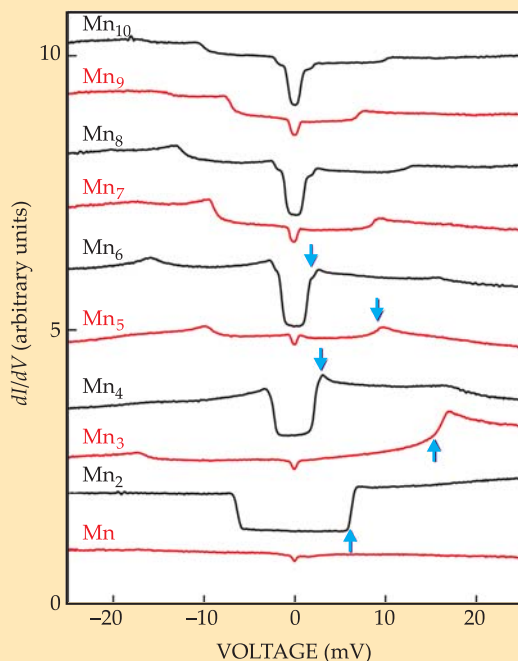


Figure 2. Electrons tunneling from the STM's tip to a chain of manganese atoms can transfer energy and angular momentum to it. Once the tip reaches a threshold voltage, electrons can tunnel not only elastically but also inelastically and prompt well-defined transitions between the magnetic ground and excited state of the Mn chain. The differential conductance dI/dV increases because the inelastic scattering opens an additional tunneling channel available to electrons. Differences between the spectra from odd- (red) and even- (black) numbered spin chains is evidence of antiferromagnetic coupling. Blue arrows mark the lowest-energy spin-changing excitation, which matches the predictions of the Heisenberg model for spin- $5/2$ atoms. (Adapted from ref. 2.)

Spectra from the trimer and longer odd-numbered chains exhibit a spin-changing excitation in addition to the dip in conductance at zero voltage from the m_s transition. Even-numbered chains, in contrast, exhibit only spin-changing excitations that take the total spin of the chain from 0 to 1. An applied magnetic field lifts the degeneracy of m_s , shifting the excitation energies by an amount proportional to the magnetic field, an indication that the excitations are indeed magnons and not, say, phonons produced from the vibrational jostling of atoms.

Simulating Hamiltonians

The energy difference between the ground and first excited state of the dimer is a measure of the exchange energy J between adjacent spins. The environment can play a crucial role: The IBM group was surprised to notice a factor of 2 difference in their measure of J depending on whether Mn atoms were positioned on Cu or N sites. In either case, armed with J , one can write down the Heisenberg Hamiltonian and diagonalize it to compare the different energies of all spin states of the system. Fitting the model to other conductance spectra—the trimer data, for instance—confirms that the total spin on Mn is $5/2$, the same as for free spins in a half-filled d shell.

For decades, simple spin models have served as tools to predict and rationalize the magnetic properties of solids. Pioneering theorists like Hans Bethe and Werner Heisenberg could scarcely have imagined that their simple model of exchanging spins would find such widespread practical applica-

tions, let alone to a laboratory system of only 10 atoms. Indeed, the focus of later workers on the thermodynamic limit boiled a lot of interesting physics out of the problem, argues Steven Bramwell of the London Center for Nanotechnology.

The IBM experiment illustrates how faithfully even simple spin Hamiltonians reproduce microscopic measurement. Future studies could realize some unusual and intriguing electronic effects. Subtle rearrangements of atoms can dramatically alter magnetic ground states, for example, and the competition between exchange and bonding interactions can produce bizarre behavior (see the article by Roderich Moessner and Arthur Ramirez in *PHYSICS TODAY*, February 2006, page 24). This past January, Peter Schiffer and his colleagues at the Pennsylvania State University explored with magnetic force microscopy the extent to which a geometrically frustrated system, an artificial “spin ice,” could be created by nanoscale fabrication techniques.⁴

Heinrich wants to explore magnetic anisotropies that exist in real systems. Among other advantages, exploiting those anisotropies might provide a mechanism to engineer atomic-scale magnetic structures into data-storage devices.

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References

1. F. Meier, J. Levy, D. Loss, *Phys. Rev. B* **68**, 134417 (2003).
2. C. F. Hirjibehedin, C. P. Lutz, A. J. Heinrich, *Science* **312**, 1021 (2006).
3. A. J. Heinrich, J. A. Gupta, C. P. Lutz, D. M. Eigler, *Science* **306**, 466 (2004).
4. R. F. Wang et al., *Nature* **439**, 303 (2006).