# Atomically assembled antiferromagnets

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Magnetism in nanostructured materials provides a unique model system to explore fundamental quantum mechanical effects experimentally. Atomic-scale magnetic structures consist of a small number of strongly coupled electron spins. This results in well-separated energy levels of the spin of these structures and its orientation is subject to quantum uncertainty. In particular antiferromagnetic spin-spin interaction produces a large variety of low-energy states even in magnetic field whereas ferromagnetic interaction typically leads to one unique ground state. Hence, antiferromagnetic coupling of atoms is at the heart of effects such as frustrated magnetic order in spin glasses or quantum critical behavior in spin ladders. Up to now most experimental and theoretical work on antiferromagnetism focusses on infinitely extended materials as it facilitates spontaneous symmetry breaking necessary to arrive at e.g. Néel order. Here we focus on nanoscale antiferromagnets and explore how stable magnetic order emerges by exchange interaction of just a few atoms.

Previously the domains in macroscopic antiferromagnets have been imaged using x-ray scattering, switched with the help of magnetoelectric antiferromagnets and their atomic spin structure has been observed by scanning probe microscopy. But when antiferromagnets shrink to nanoscale dimensions, uncompensated magnetic moments can arise at the particle's surface that often obscure the behavior of the antiferromagnet.

We use low-temperature scanning tunneling microscopy to position magnetic atoms in a regular pattern on a surface. In this way we can control the size, shape, magnetic anisotropy and spin coupling by manipulating the immediate environment of the atoms. Spin polarized imaging identifies the magnetic order of the assembled nanostructures and inelastic electron tunneling spectroscopy quantifies the magnetic anisotropy energies of the atoms. We use a monatomic layer of $Cu_2N$ on Cu (100) as substrate. This surface allows precise placement of atoms by vertical atom manipulation (Fig, 1).
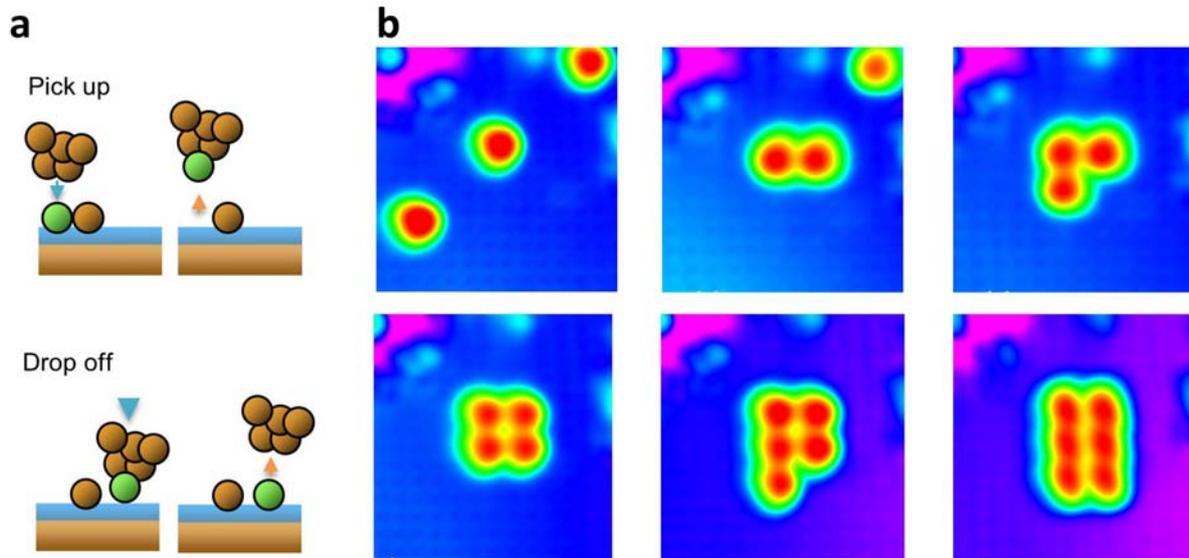


Figure 1: Atom-by-atom assembly of a nanomagnet with a scanning tunneling microscope. Fe atoms are placed into a regular array on the non-magnetic surface. (a) Manipulation sequence: Fe atoms are picked up to the tip by lowering the tip into point contact with the surface-bound atom and applying a voltage pulse, the reverse process drops the atom onto the surface at a chosen location. (b) Consecutive STM topographs recorded during the assembly of a (2x6) structure of Fe atoms on a $Cu_2N/Cu(100)$ surface (Image size: (4 x 4) $nm^2$, Color scale shows increasing height from blue/purple to yellow/red). A spacing of two unit cells (0.72 nm) was chosen to ensure antiferromagnetic exchange coupling between the atoms.

The $Cu_2N$ surface provides strong spin-spin interaction between adjacent atoms. Fe atoms placed at a distance of 0.72 nm along the N binding direction were found to interact antiferromagnetically with $J = 1.2$ meV. This favors the antiferromagnetic ground state with an energy gain, $E_J = 9.6$ meV, for each pair of Fe spins compared to ferromagnetic alignment ($E_J = 2S^2J$ with $S = 2$ for Fe). At the base temperature of the scanning tunneling microscope, T = 0.5 K, the arrays are therefore in their antiferromagnetic ground state.
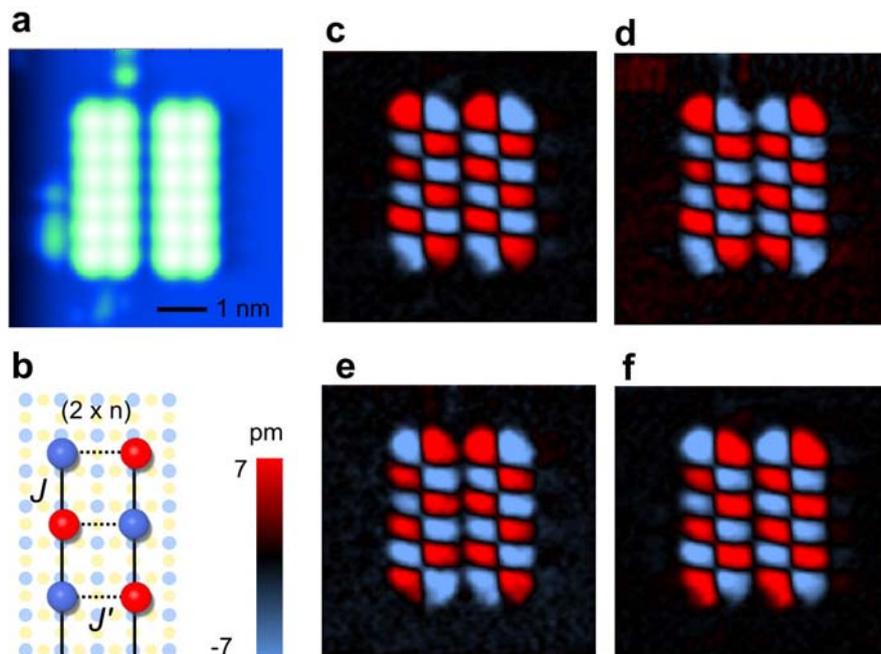


Figure 2: Antiferromagnetic order in few-atom magnetic arrays. (a) STM topograph of structure consisting of 24 Fe atoms that was assembled in two units of (2x6) arrays. Image taken with non-spin-polarized tip. (c)-(f) Spin-sensitive images of the four stable magnetic states of the array in (a). All states show antiferromagnetic order in each (2x6) subunit but changing alignment between the subunits. Magnetic contrast was obtained by repeated imaging with a spin-polarized tip at 3 T magnetic field. A color-coded difference between spin-polarized and spin-averaged topographs is shown, with red corresponding to higher tip height and blue to lower tip height in the spin-polarized image.

In assemblies of a few antiferromagnetically coupled atoms the magnetic ground state is typically a singlet ground state. This quantum superposition state is characterized by a wave function in which all spins populate opposing spin states equally. In contrast, we find that isolated antiferromagnetic (AFM) structures with as few as 6 Fe atoms exhibit stable Néel states, in which the spin orientation alternates between neighboring atoms.

Figure 2a shows a structure of 24 Fe atoms comprised of two subunits with 12 atoms each. We can resolve the magnetic order of the array by imaging it repeatedly with a normal metallic (non-spin-polarized) and a spin-polarized STM tip. The spin-polarized STM tip forms a magnetic tunnel junction in which the conductance alternates between high (parallel alignment of tip and sample spins) and low (anti-parallel alignment) as the tip passes from atom to atom. The difference image obtained by subtracting spin-average STM topograph from spin-polarized topograph clearly shows binary antiferromagnetic order in each of the (2x6) Fe arrays, Fig. 2c-f.

We find four low-energy magnetic configurations for the structure in Fig. 2.

The antiferromagnetic order in each (2x6) unit is always preserved. Two states also preserve antiferromagnetic order across the subunits (Fig. 2c,f) whereas two states have spins with equal orientation at the connecting face of the subunits (Fig. 2d,e). The spacing between the (2x6) subunits is 1.44 nm. The possibility to observe frustrated magnetic orientation across this distance indicates a much reduced magnetic interaction compared to that between adjacent Fe atoms in each subunit. This is consistent with short-ranged magnetic interaction dominated by super-exchange through the N

atoms. All states can be well described by the classical Ising model, in which the spins always point along one axis.
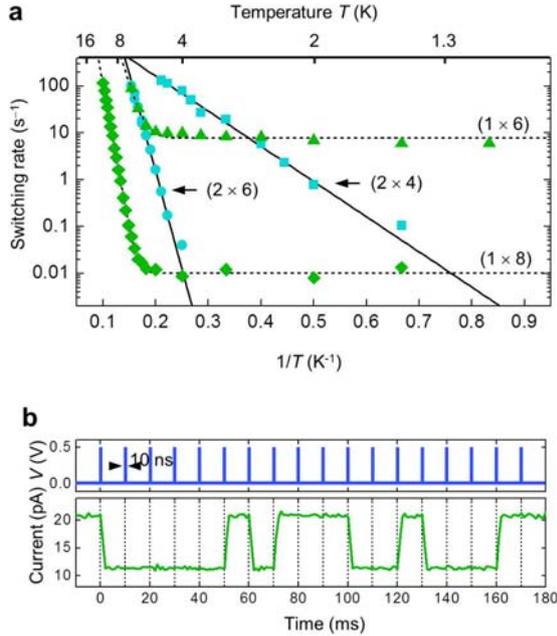


Figure 3: Switching characteristics of antiferromagnetic arrays. (a) Spontaneous switching rate for different Fe arrays plotted logarithmically as a function of temperature. At temperatures above 5 K all arrays show thermally activated spin relaxation following an Arrhenius-type temperature dependence (Linear slope in ln(t) vs. 1/T plot). All arrays stabilize when the temperature is lowered. The linear Fe chains (1x6) and (1x8) settle in a temperature-independent switching rate indicating spin relaxation via quantum tunneling of magnetization. (b) Intentional switching of a (1x8) Fe array by applying voltage pulses, $V$, with 10 ns duration and 0.5 V height (top panel). The spin-polarized tip is held constant above one end atom of the chain. The simultaneously recorded tunnel current (bottom panel) identifies the resulting magnetic state after each pulse. Note that the voltage pulses themselves are too short to register in the tunnel current.

To investigate the stability of the Néel states we examined the thermal switching rates of linear chains of Fe atoms with varying length, (1×n), and arrays of two coupled chains, (2×n) (Fig. 3a). All structures containing 8 or more atoms were found to be stable at the lowest temperature, 0.5 K. Spontaneous flipping between the two Néel states sets in with increasing temperature. Structures with more atoms remain stable to higher temperatures.

The thermal switching rates were found to be independent of magnetic field and both Néel states were occupied equal amounts of time. Hence the two observed Néel states for each (2x6) unit are fully spin compensated and there is no energy difference between them. This fits well with the expectations of two degenerate ground states of the Ising model.

Above ~5 K the switching rates of the (1×6), (1×8) and (2×6) arrays follow the Arrhenius law with comparable spin reversal barriers, $E_B \sim 7$–12 meV. This barrier is comparable to the energy needed to create a single Ising domain wall which is $E_J$. This indicates that switching proceeds by flipping one or more consecutive spins at the end of a chain and subsequent propagation of the Ising domain wall through the array [1].

Below ~5 K the switching rates of the (1×6) and (1×8) chains become independent of temperature. Such behavior is consistent with quantum tunneling of magnetization (30), which is typically observed in few-atom molecular magnets and also occurs in magnetic nanoparticles. Here it causes the AFM arrays to transition spontaneously between the two Néel states which limits their stability even though thermal switching is frozen out.

The stability of the magnetic states was not affected by imaging them using an applied voltage of <2 mV. Voltages in excess of ~7 mV caused switching. We applied a pulsed excitation scheme to determine the speed at which a switch occurs. Voltage pulses with nanosecond duration were applied to the tip in order to induce a switch. The success of the switching attempt was then monitored in a following low-voltage window. The switching rates increased drastically with increasing voltage up to switching times of ~20 ns at 0.5 V (Fig. 3b). At high speed we observe directionality of switching which is consistent with the spin-polarization of the tip. This points to an efficient reversal process of

the AFM structures by direct spin-momentum transfer with tunneling electrons similar to spin pumping of individual atoms [2].

For arrays with 12 or more atoms no spontaneous switching was observed at 0.5 K. We experimentally determined a lower limit to the stability of the (2x6) arrays of 17 hours. This high stability in conjunction with fast switching (Fig. 3b) and dense packing of multiple units (Fig. 2a) fulfills all requirements for a data storage material. A challenge to miniaturizing the bits in current ferromagnetic storage media is the interaction of neighboring bits due to their dipolar magnetic fields. This would be absent in an antiferromagnetic medium.

To demonstrate the possibility of AFM data storage we assembled eight (2x6) arrays in close proximity representing the eight bits of one Byte of information. The two different Néel states identified by the spin orientation of the first atom in each array were chosen to represent '1' and '0' respectively. We could read and write this information more than 400 times without causing damage to the atomic arrangement [3].

At the distances of less than 1 nm all atoms were magnetically coupled. But unlike with ferromagnetic interaction, antiferromagnetic coupling allowed near perfect bit-to-bit decoupling by making use of geometric magnetic frustration. This demonstrated the feasibility of information storage in antiferromagnetic nanostructures at high aerial densities (70 Tb/in$^2$), albeit at low temperature.

A central finding of this work is that a few antiferromagnetically coupled atoms can exhibit stable Néel order with completely compensated magnetic moment. It remains an intriguing open question, how these classical Néel states merge into singlet or other quantum mechanical states. Scanning tunneling microscopy now holds a completed toolkit to construct specific model spin systems with atoms on surfaces and measure their energetic and dynamic properties.

**References:**
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