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Giant magnetoresistance in FeRh: A natural magnetic multilayer

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We point out that the binary metallic compound FeRh is a naturally occurring magnetic multilayer structure that shows giant changes in electrical resistance when switched from antiferromagnetic to ferromagnetic order by an applied field. The transition from antiferromagnetic to ferromagnetic coupling and the associated drop in resistance are similar properties to those of fabricated magnetic multilayer structures. The simple crystal structure makes possible a reliable calculation of the magnetic structure of FeRh from first-principles total-energy electronic-structure calculations, which show that the ground state is type-II antiferromagnetism and that there is a metastable ferromagnetic state that is accessible by an applied magnetic field.

Large decreases in electrical resistivity are observed when a magnetic field switches the magnetic coupling from antiferromagnetic to ferromagnetic in certain composite structures made up of alternating layers of atoms with different magnetic behavior. This so-called "giant magnetoresistance effect" was first observed in a fabricated structure consisting of alternating iron and chromium layers with each layer consisting of a number of atomic planes.[1] Such magnetic multilayers are difficult to treat by first-principles quantum-mechanical calculations of electronic and magnetic structure because the unit cell must be large enough to contain the basic periodicity of the repeating unit of the supercell implied by the multilayer. Such cells contain a sufficiently large number of atoms to make great demands on the accuracy of total-energy-band calculations.

In magnetic multilayers, the coupling between successive magnetic layers implies that the total moment M of a double supercell (doubled to allow for antiferromagnetism) is either zero (antiferromagnetic coupling) or finite (ferromagnetic coupling). Thus there are two magnetic states, one being more stable than the other. Multilayer structures, which have interesting device potentials, have a stable antiferromagnetic state and a metastable ferromagnetic state.

In addition to the large number of atoms in the supercell, first-principles calculations on fabricated multilayers are made difficult by the relatively small energy separation of the two magnetic states. This energy difference is directly related to experimental switching fields. For example, for a cobalt-rhodium magnetic multilayer, Parkin[2] finds a switching field of ~ 10 kOe, which corresponds to an energy difference of $\sim 4\times 10^{-3}$ mRy/atom to switch one μ_B between the antiferromagnetic and ferromagnetic states ($1\,\text{mRy}/\mu_B = 2351\,\text{kOe}$). Such an energy difference would be very difficult to resolve with present state of the art band calculations.

In this paper, we point out that naturally occurring ordered FeRh, which consists of alternating monolayers of magnetic (iron) and nonmagnetic (rhodium) atoms stacked in the (100) direction of a CsCl unit cell, has long been known to exhibit a transition from antiferromagnetic to ferromagnetic order. Furthermore, there is an accompanying giant decrease in electrical resistance, which in fact has been used to detect the transition. In this case, the transition and the associated switching of the magnetic order between the magnetic layers can be induced by either an increase in temperature or by an applied magnetic field. That is, this system has the unusual property of being antiferromagnetic at low temperatures and of undergoing a phase transition to a ferromagnetic state at ~ 340 K. The antiferromagnetic-ferromagnetic transition can be induced at temperatures below ~ 340 K by an applied magnetic field, which goes linearly to zero as $T \rightarrow \sim 340$ K. The field is ~ 40 kOe at room temperature, and is ~ 140 Oe at a temperature 0.1 K below the transition.

This phase transition was discovered in 1938 by Fallot,[3] and has subsequently been the subject of extensive experimental and theoretical studies. In 1962, Kouvel and Hartelius[4] measured the zero-field electrical resistivity in a polycrystalline specimen as a function of temperature and found a 30% drop in the resistivity at the transition temperature. In 1974, Schinkel, Hartog, and Hochstenbach[5] confirmed the earlier result and, in addition, found that the resistivity drops by *factors* as large as 20 in fields of 100 – 200 kOe at 4.2 K. Rhodium-rich alloys show[5,6] broad transitions and smaller resistance drops than the ordered 50/50 alloy. An FeRh thin film,[7] made by evaporation of alternate layers of iron and rhodium and annealing also showed a significant resistance drop, although smaller (6%) than the bulk specimens.

An important feature of FeRh as a multilayer structure is that its magnetic behavior can be reliably calculated by first-principles total-energy-band theory. We have already reported[8] the magnetic structure of FeRh derived from such calculations, which shows good agreement with experiment. In contrast with fabricated magnetic multilayers, the coupling between iron monolayers in FeRh is sufficiently large that the energy difference between the antiferromagnetic and the ferromagnetic state is a few tenths of a mRy and can be resolved by present day electronic-structure calculations. Our FeRh calculations show that the ground state (the lowest energy state) is indeed antiferromagnetic, and moreover that it is type-II antiferromagnetism, which consists of (100) iron layers with alternating spins within the planes and rhodium layers with zero spin as shown in Fig. 1. The total energy of type-I antiferromagnetism, which consists of (100) iron layers with parallel spins within one plane that are antiparallel with the spins in the neighboring iron layer, is close to, but higher, than that of type-II antiferromagnetism. The calculations show that the type-I and type-II antiferromagnetic configurations are only slightly more stable than the ferromagnetic spin configuration and that at zero temperature[9] an applied magnetic field of ~339 kOe will switch the antiferromagnetic spin arrangement to a ferromagnetic one. For ordered FeRh, the iron local moments are $\sim 3\mu_B$ for both the antiferromagnetic and ferromagnetic configurations, while the rhodium local moments are zero for the antiferromagnetic configuration and $1\mu_B$ for the ferromagnetic configuration.

In Fig. 2 we reproduce a plot of a calculated total energy E vs total moment M (per formula unit) for FeRh at a lattice constant ~4.4% below equilibrium, showing the stable antiferromagnetic minimum at $M=0$ and the metastable ferromagnetic minimum at $M \sim 4\mu_B$. This $E(M)$ curve was obtained by utilizing a fixed-spin-moment procedure that constrains a magnetic cell to have fixed values of total magnetic moment derived from collinear spins. The magnetic cell is made sufficiently large to allow for antiferromagnetism. Such calculations allow us to study the behavior of the system in an applied magnetic field defined as $H=dE/dM$. The upper panel shows the calculated derivative or $H(M)$ curve. Note that only certain parts of the $E(M)$ curve are accessible to the system. At some critical field H_{cr}, defined by the slope of the common tangent to the two minima, the two states are in thermodynamic equilibrium. The system will be equilibrium in the ferromagnetic state for any $H > H_{\mathrm{cr}}$.

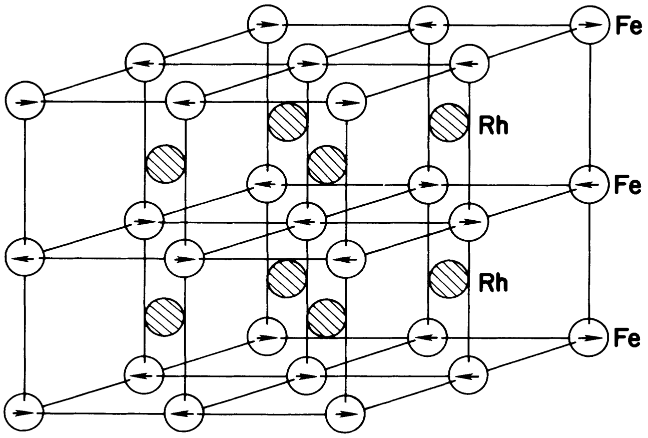

FIG. 1. Type-II antiferromagnetic spin configuration for ordered FeRh. Electronic-structure calculations yield the indicated experimental configuration with $\sim 3\mu_B$ local iron moments and zero local rhodium moments. Application of a sufficiently large field switches all iron spins to a parallel configuration and induces parallel spins of $\sim 1\mu_B$ on the rhodium atoms.

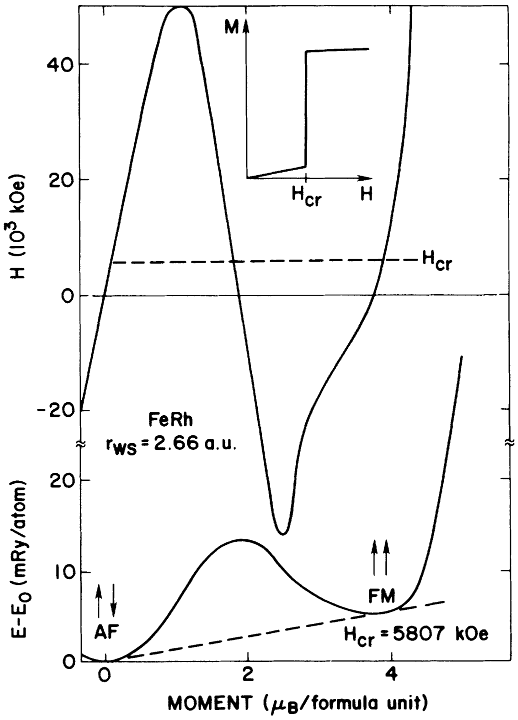

FIG. 2. Calculated $E(M)$ and $H(M)$ curves for FeRh. The antiferromagnetic (AF) minimum is the ground state, and the ferromagnetic (FM) minimum is a metastable state that can be accessed by forcing (constraining) the system to have the indicated total moments. The fields necessary to force the system to have the indicated moments are given by the derivative, $H=dE/dM$. The AF and FM states are in equilibrium at the critical (switching) field given by the slope of the common-tangent construction indicated by the dashed line.

A transposition of the ordinate and the abscissa of the accessible portions of the $H(M)$ curve yields the response of the magnetization to an applied field, or the $M(H)$ curve shown schematically in the inset. The critical field, $H_{\mathrm{cr}} \sim 5800$ kOe, required to switch FeRh from the antiferromagnetic to the ferromagnetic state shown in Fig. 2 is huge[10] in comparison with typical switching fields for fabricated magnetic multilayers. However, the results shown in Fig. 2 apply to FeRh constrained to collinear spins on a rigid lattice (i.e., no zero-point motion) at a given volume (lattice constant). The calculations show that, at the equilibrium volume, H_{cr} is reduced to 2100 kOe. A zero-point energy correction[9] leads to a further reduction to ~339 kOe at zero temperature, in general agreement with observed[11] critical fields. As noted above, H_{cr} decreases with increasing temperature and at higher temperatures goes linearly to zero as $T \rightarrow 340$ K. Schinkel, Hartog, and Hochstenbach[5] used the large drop in the electrical resistance at the transition from antiferromagnetic to ferromagnetic coupling between the iron monolayers in FeRh to determine $H_{\mathrm{cr}}(T)$.

FeRh is a unique binary metallic system with an antiferromagnetic ground state that can be switched to a ferromagnetic state with attainable fields with no change in crystallographic structure. Although there are a number of binary metallic systems[12] with stable ferromagnetic states lying closely below antiferromagnetic states, e.g., FeV and FeAl, it is much less common to find the antiferromagnetic state lower.

FeRh exists as a bulk material and exhibits properties that are similar to those of fabricated magnetic multilayers consisting of alternating layers of magnetically active and magnetically inactive atoms. In both cases, the coupling across the magnetically inactive layers can be switched from antiferromagnetic to ferromagnetic by applying a magnetic field. In both cases, antiferromagnetic coupling results in high electrical resistance and ferromagnetic coupling results in low electrical resistance. FeRh represents a magnetic multilayer in the single alternating atomic-layer limit in which the physical effects, including the magnetoresistance effects, are amplified compared to fabricated structures with thicker individual layers. The phase transition is strong and easily measurable and the electronic structure can be calculated quantitatively. FeRh should therefore provide a useful test of theories of giant magnetoresistance.

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