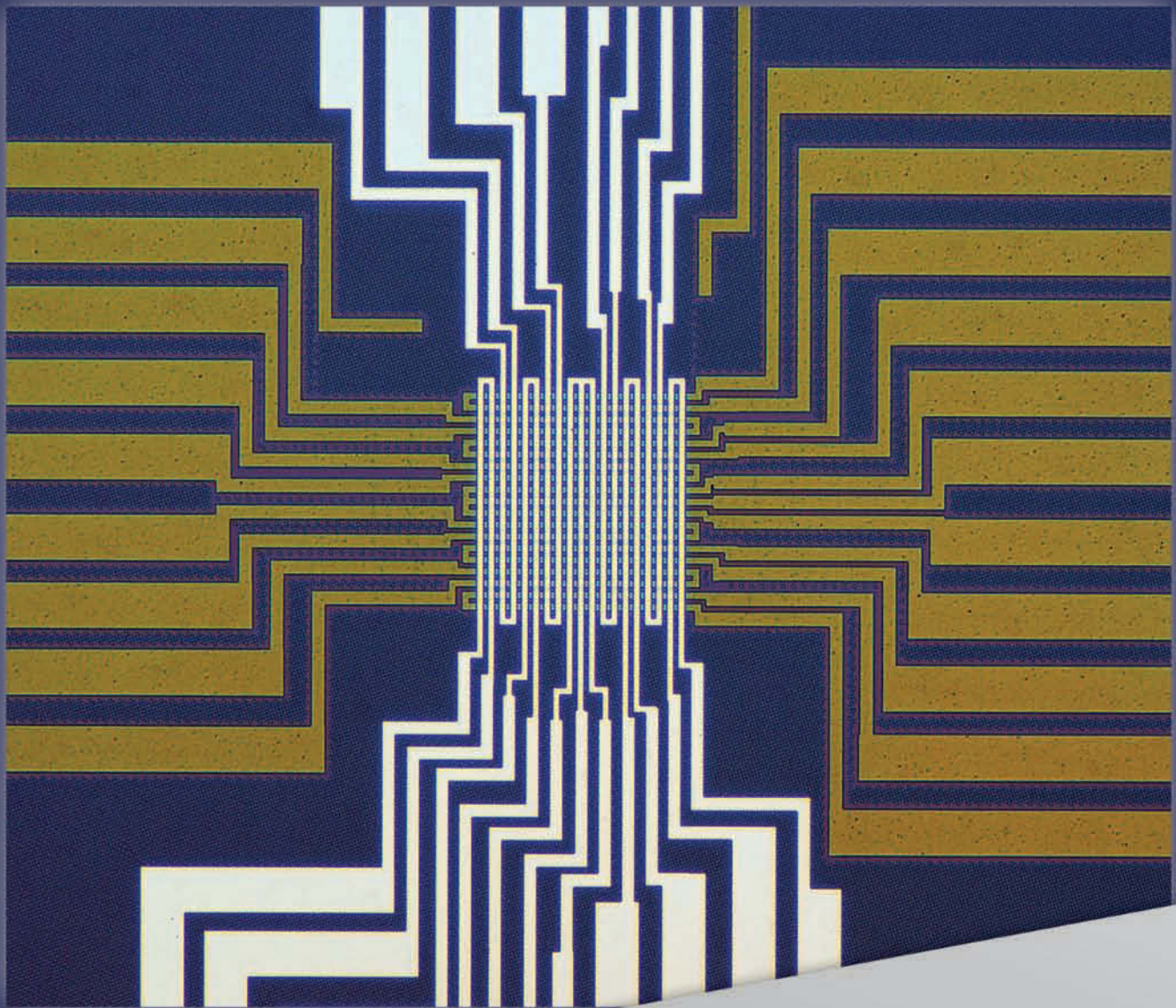# Origins of Spin Coupling across Interfaces

Michael Fitzsimmons and Sunil K. Sinha*

* University of California, San Diego

The tiny "read head" in a computer hard drive is a multilayered magnetic-thin-film structure. Essential to its operation is a ferromagnetic thin film whose magnetization is held fixed through a phenomenon known as exchange bias. Magnetic coupling of the "fixed" layer to an adjacent antiferromagnetic layer establishes the bias. The fixed layer provides a reference point for a second "free" ferromagnetic layer in the read head which enables it to discriminate between 1s and 0s magnetically stored on the hard-drive disk. We used x-rays at the Advanced Light Source and then polarized neutrons at the Los Alamos Neutron Science Center's ASTERIX spectrometer to probe the origin of exchange bias in a model ferromagnetic-antiferromagnetic thin-film system. We found an interesting interplay between loose and pinned spins near the ferromagnetic–antiferromagnetic interface, which motivated us to develop a new model of exchange bias to explain its magnitude and sign.

Currently, microelectronic devices use the electron's charge for their operation and for the most part ignore the electron's "spin" (intrinsic angular momentum), or intrinsic magnetic moment. However, new types of microelectronic devices are now emerging that use the electron's spin (or magnetic moment), together with its charge.

Compared with conventional devices, these "spintronic" devices have the potential advantages of nonvolatility, increased data-processing speed, decreased electrical-power consumption, and increased integration densities. Although a whole range of spintronic devices is being studied and developed, the read head in a computer hard drive, also called a "spin-valve head," is already widely used. Another spintronic device, magnetic random access memory (MRAM), is poised to replace the charge-based RAM now used in computers.[1] Both spin-valve heads and MRAM employ the magnetic properties of layered thin films to influence electron transport through the films and thereby read magnetically stored data.

At left is a micrograph of a magnetic random access (MRAM) circuit. MRAM is nonvolatile RAM poised to replace the electronic RAM now used in computers (reprinted courtesy of International Business Machines Corporation, Copyright 2000). **Below is a schematic of the spin-valve that reads data stored on computer hard drives. Both devices use exchange bias provided by thin antiferromagnetic layers.**
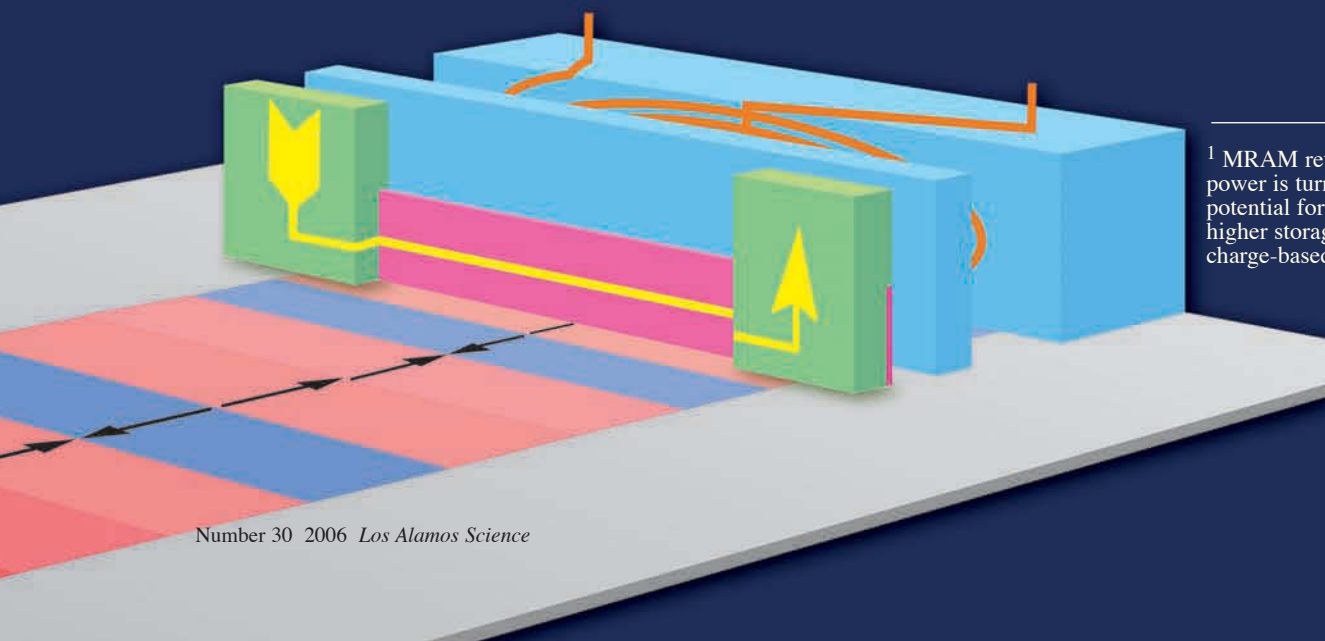
[1] MRAM retains data when the power is turned off and has the potential for higher speeds and higher storage densities than charge-based RAM.

For both computer read heads and MRAM, the thin film nanostructure essential in reading magnetically stored data consists of two closely spaced ferromagnetic thin films, one a "free" layer and the other a "fixed" layer (Figure 1). The magnetization of the free layer can be changed by the fringe fields of magnetically stored bits, whereas the magnetization of the fixed layer is engineered to remain constant. In a read head, for example, a change in the magnetization of the free layer relative to the fixed layer changes the read head's resistance, and the resulting change in voltage is used to discriminate between stored 1s and 0s [see the box "Giant Magnetoresistance (GMR) in a Hard-Drive Read Head" on page 182].

The magnetization in the fixed layer can be held constant in different ways, but the most common method is called "exchange bias." An antiferromagnetic layer placed next to the fixed layer shifts, or biases, the fixed layer's hysteresis curve (Figure 2) through magnetic, or spin exchange, coupling between the two layers. The exchange bias in the fixed layer ensures that only extremely large external magnetic fields can change that layer's magnetization, whereas relatively small external magnetic fields can only change the free layer's magnetization. Our studies have focused on the details of how exchange bias works.

How Antiferromagnets Produce Exchange Bias

The "Old" Picture. An antiferromagnet is said to consist of nominally equal numbers of atoms whose spins point in opposite directions. (An atom's spin is produced mainly by the spins of its outer electrons.) Thus, the net magnetization of an antiferromagnet is nominally zero, which means an external magnetic field does not affect
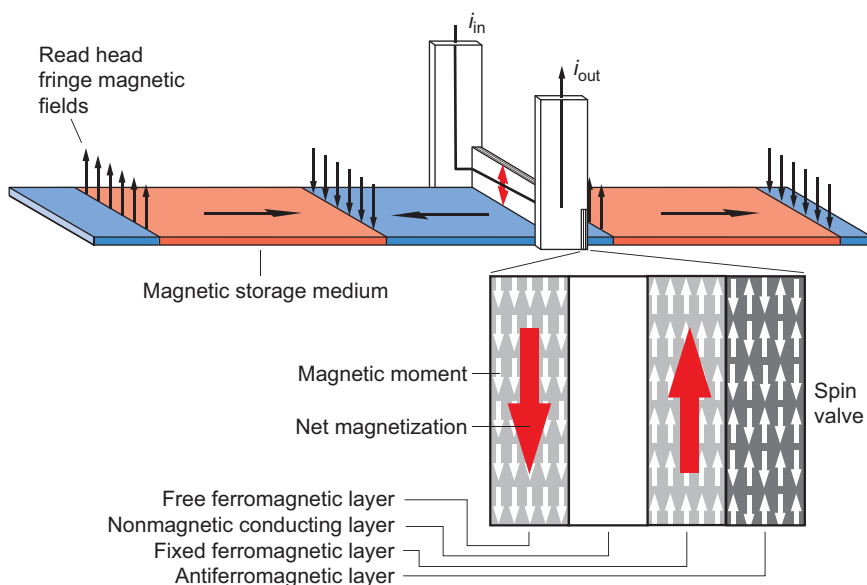
Figure 1. Operation of a Spin-Valve Read Head
A spin-valve read head consists of four conducting nanolayers: an antiferromagnetic layer, a "fixed" ferromagnetic layer, a nonmagnetic conducting layer, and a "free" ferromagnetic layer. As the spin valve passes over the magnetic bits on the hard-drive disk, for fixed current through the spin valve, the voltage across the spin valve changes because of a magnetic interaction with the net magnetic fringe fields between a 0 and a 1 or between a 1 and a 0. Specifically, those fringe fields cause the magnetization in the free layer of the spin valve to align parallel to them while the magnetization in the fixed layer of the spin valve remains in a fixed direction. A change in the free layer's magnetization causes the resistance (and thus the voltage) across the spin valve to change. The resistance is low when the magnetizations of the free and fixed layers are parallel, and it is high when they are antiparallel, as described in the box on page 182. The magnetization in the fixed layer is held constant by the exchange bias established through magnetic coupling with the adjacent antiferromagnetic layer, whose microscopic origin is still debated. The nonmagnetic conducting layer provides a conducting path between the free and fixed layers.
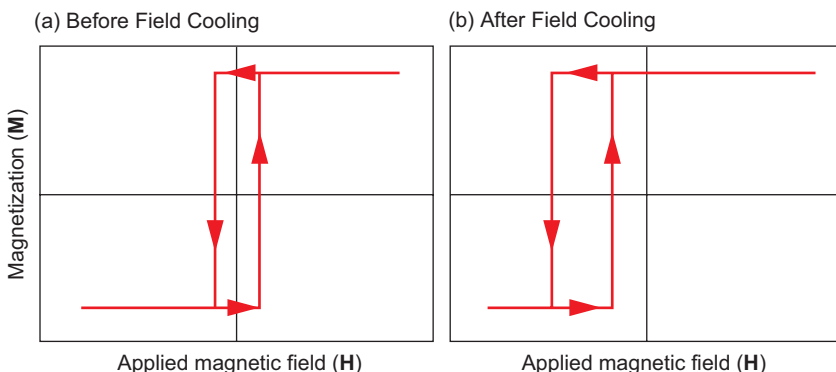
the net magnetization. However, in practice, a relatively small number of "uncompensated" spins in an antiferromagnet are not canceled by opposite spins. Strongly coupled to nearby spins in the antiferromagnetic layer, the uncompensated spins are seemingly "frozen" and do not easily respond to an external applied magnetic field. But the uncompensated spins are also coupled to the spins in the adjacent ferromagnetic layer. That coupling across the antiferromagnetic–ferromagnetic interface produces the exchange bias by inhibiting the response of the ferromagnetic

material to applied fields.

However, this model raises several questions. (1) Why is the exchange bias two to three orders of magnitude smaller than that predicted by a simple calculation assuming a perfectly smooth interface? (2) Where exactly are the uncompensated spins? (3) How do the uncompensated spins couple to the ferromagnetic spins? We performed a series of experiments to try and answer these questions.

Los Alamos Studies. In early experiments, we showed that the exchange bias produced by an anti-

(a) Before Field Cooling

(b) After Field Cooling

Magnetization (**M**)

Applied magnetic field (**H**)

Applied magnetic field (**H**)

Figure 2. Exchange Bias in a Ferromagnetic-Antiferromagnetic Two-Layer System
These plots show the hysteresis curves (magnetization vs applied magnetic field) of a ferromagnetic thin film in contact with a second film that changes from paramagnetic to antiferromagnetic when cooled below its Néel temperature. The curves are for temperatures (a) above and (b) below the Néel temperature. (The arrows indicate whether the applied field is being increased or decreased.) After cooling, the ferromagnetic layer's hysteresis curve is shifted along the magnetic field axis from being centered at 0 to being centered at a higher absolute value. "Exchange bias" refers to this effect and to the magnitude of the shift. The fixed layer in a spin-valve read head is exchange-biased to make its magnetization impervious to the small fields of the storage medium. The fixed layer thus provides a reference point that allows the nanostructure in Figure 1 to discriminate between the 1s and 0s stored on a hard drive.

We first used circularly polarized, resonant soft-x-ray reflectometry to measure the magnetization as a function of depth. The x-ray measurements provided the magnitude of the magnetization component along the direction of the incident x-ray beam in an element-specific manner (namely, that arising separately from the cobalt and iron atoms). We then used polarized-neutron reflectometry to measure the depth dependence of both the magnitude and direction of the magnetization in the sample. For both the x-ray and the neutron measurements, the spatial resolution was 1 nanometer.

ferromagnetic iron fluoride (FeF_2) nanolayer grown with minimal crystal strain is small (Fitzsimmons et al. 2002). However, dramatic increases of exchange bias using the same ferromagnetic and antiferromagnetic materials have been realized when the antiferromagnetic layer (FeF_2) is grown on magnesium fluoride (MgF_2) single crystals. Because of the large lattice mismatch between FeF_2 and MgF_2, misfit dislocations are formed in the FeF_2 (Figure 3) that partially relieve the crystal strain across the FeF_2–MgF_2 interface. These dislocations and unrelieved misfit strain are sources of uncompensated magnetization in the FeF_2 antiferromagnetic that promote large exchange bias.
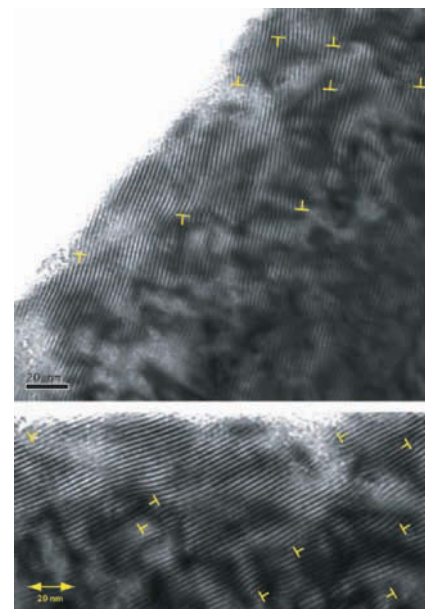
The exchange bias model system studied in our new experiments consisted of a ferromagnetic layer over an antiferromagnetic layer with the two sandwiched between two nonmagnetic

layers (Figure 4). The nonmagnetic substrate layer was a single crystal of MgF_2. The other layers, in the order in which they grew onto the nanostructure, were a 36.6-nanometer-thick, single-crystal layer of FeF_2; a 4.1-nanometer-thick layer of cobalt, which is ferromagnetic; and a 5.0-nanometer-thick layer of aluminum, which is nonmagnetic.

In all the experiments, we subjected the sample to a magnetic field parallel to the [001] direction of the FeF_2 crystal before cooling the sample well below its Néel temperature of 78.4 kelvins (the temperature below which the FeF_2 is antiferromagnetically ordered). The [001] direction is along the "easy" magnetization axis.[2]

_____

[2] The magnetic moments in an antiferromagnetic crystal prefer to align parallel or antiparallel to a crystal direction called the easy axis.

Figure 3. Transmission Electron Micrograph of the Dislocations in the FeF_2 Layer
The dislocations (yellow symbols) were produced by growing the FeF_2 layer on a MgF_2 substrate with a slightly smaller lattice spacing than the FeF_2 layer. The dislocations arise in order to partially relieve misfit strain across the FeF_2–MgF_2 interface. The dislocations and unrelieved misfit strain are sources of uncompensated magnetization in antiferromagnetic FeF_2.

Giant Magnetoresistance (GMR) in a Hard-Drive Read Head

Brian H. Fishbine

Giant magnetoresistance (GMR) was first observed independently by Baibich et al. (1988) and Binasch et al. (1989). They found the resistance of a structure of alternating nanolayers of iron and chromium decreased by as much as 50 percent when the magnetizations in all the iron nanolayers were aligned. The effect was called "giant" because it is much larger than the other effects of magnetization on the resistance of ferromagnetic materials known at the time.

Figure A shows how GMR works. Driven by a voltage produced by an electric field perpendicular to the layers, electrons travel successively through a ferromagnetic layer, a nonmagnetic conducting layer, and a second ferromagnetic layer. Each layer is nanometers thick at the most. The electrical resistance of the layered nanostructure is low when the magnetic fields in the two ferromagnetic films are parallel and high when the fields are antiparallel. The GMR effect is analogous to that observed when aligned polarizers pass light but crossed polarizers do not.

In a conducting ferromagnetic transition metal, approximately equal numbers of free electrons have spins parallel or antiparallel to a given direction. The first ferromagnetic layer allows electrons whose spins are parallel to that layer's magnetization to pass through easily. If the magnetization of the second ferromagnetic layer is parallel to that of the first layer, the same electrons also pass easily through the second ferromagnetic layer, and the resistance of the layered nanostructure will be low. However, if the magnetic fields of the two layers are antiparallel, electrons with either spin will not pass through the nanostructure easily, and the electrical resistance will be high.
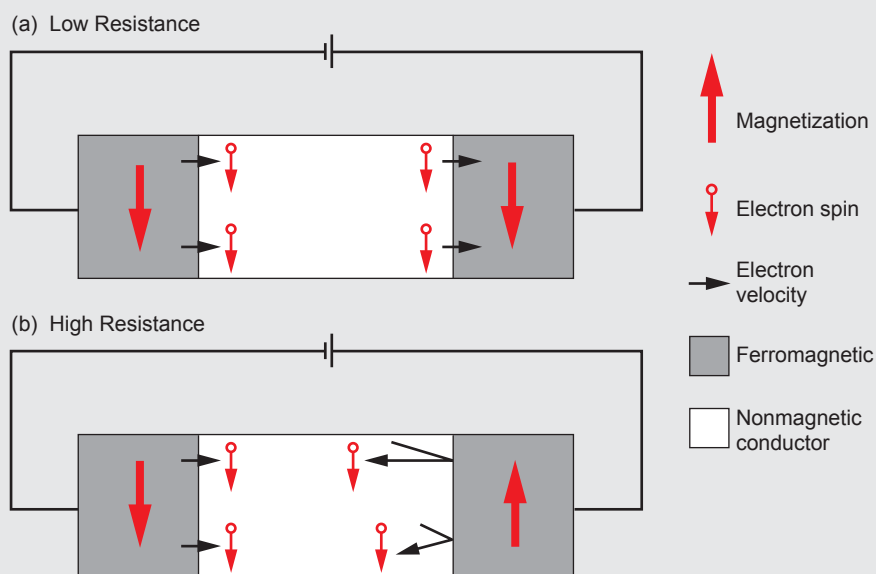

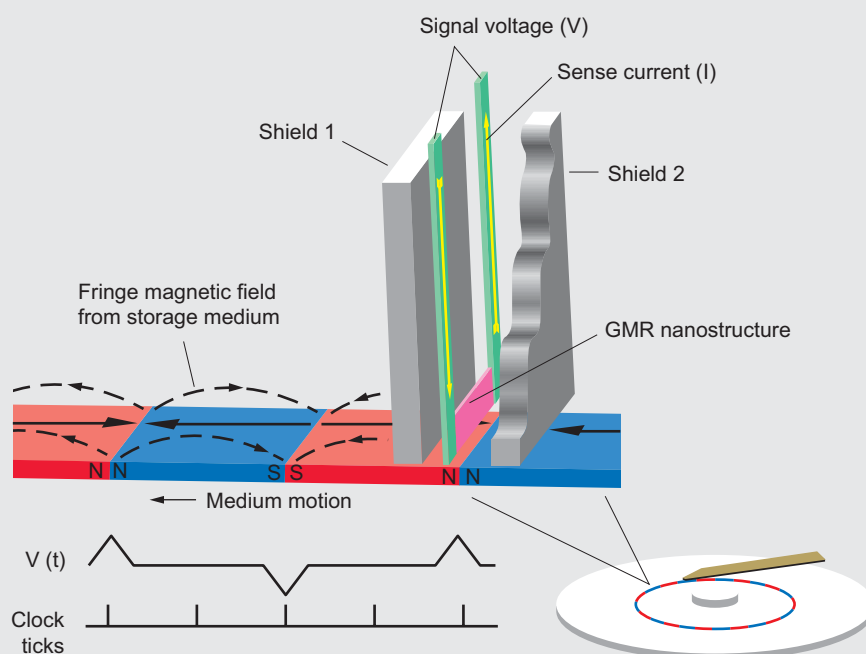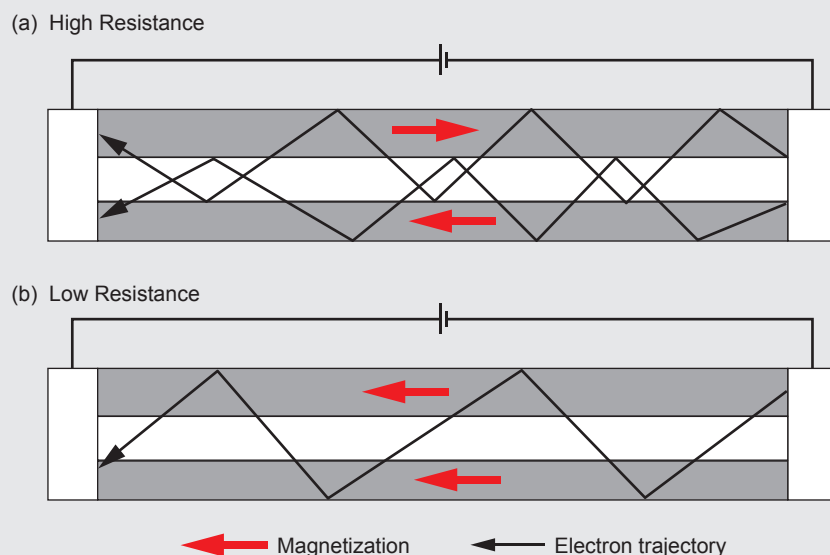Figure A. Spin-Dependent Electron Transport in a GMR Nanostructure

Figure B. The Spin-Valve Read Head in a Computer Hard Drive

(a) High Resistance

(b) Low Resistance

→ Magnetization   ← Electron trajectory

Figure C. The Geometry of the GMR Nanostructure in a Spin-Valve Read Head

Figure B shows how the GMR, or spin-valve, read head in a hard drive works. As the hard-drive disk rotates, the bits stored on it pass under the read head's GMR nanostructure at a distance of 10 nanometers or less. The bits' magnetic fields can change the magnetization of the nanostructure's "free" layer but not the magnetization of the nanostructure's "fixed" layer, which is held constant (either up or down) by the nanostructure's antiferromagnetic layer. Through the GMR effect, the resistivity of the nanostructure's layers depends on whether the magnetizations in the two ferromagnetic layers are parallel or antiparallel. Thus, the resistivity depends on the bits' magnetic fields. A constant current flows through the layers, so the voltage across them is determined by the bits' magnetic fields; data are read by measuring this voltage. The two shields ensure that the nanostructure responds only to the magnetic fields at the bit boundaries, where the magnetic fields from two adjacent bits cancel or are mainly perpendicular to the hard drive disk. The bit values are determined from the voltages measured at the clock ticks. As shown, a bit passes under

the GMR nanostructure every two clock ticks. The three layers shown in Figure C are the heart of the GMR nanostructure. A spin-valve read head has an additional layer (not shown), which stabilizes the fixed layer (see Figure 1 in the main article).

In Figure C, the current is generally parallel to the nanostructure's layers, which is the case with actual read heads but differs from the situation depicted in Figure A. As in Figure A, however, changes in resistance result from changes in how the electrons scatter from the conductor–ferromagnet interfaces as the magnetizations of the ferromagnetic layers change. We also note that the GMR effect works even if magnetizations in the ferromagnetic layers are perpendicular to the current rather than parallel to it as shown in Figure C.

GMR's high sensitivity to magnetic fields allows a read head to read small bit volumes, which translate to high storage densities.
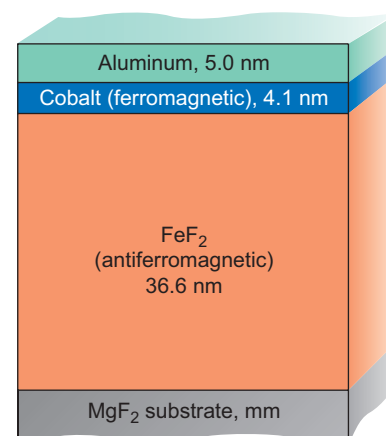
Figure 4. Model System for Studying Exchange Bias
The model system consists of a ferromagnetic cobalt layer in contact with a single-crystal FeF_2 layer. The FeF_2 layer becomes antiferromagnetic when cooled below its Néel temperature of 78.4 K.

We performed the x-ray measurements at the Advanced Light Source at Lawrence Berkeley National Laboratory. Circularly polarized x-rays tuned to the x-ray absorption L-edges of first cobalt and then iron were used. The difference in the reflected x-rays for left and right circular polarization at the L-edge of a magnetic atom is proportional to the magnetization involving that element. First, we applied a magnetic field of 796 kiloamperes per meter (kA/m) parallel to the FeF_2's [001] direction and cooled the sample to 20 kelvins. Then, we took x-ray reflectometry measurements at a single angle of incidence and reflection while gradually varying the magnitude and reversing the direction of the applied magnetic field. That procedure allows us to measure the projection of the magnetization along the direction of the incident x-ray beam as a function of applied magnetic field. Using this procedure, we measured the hysteresis curves of the ferromagnetic

(a)

(b) External magnetic field (along FeF₂ [001] direction)
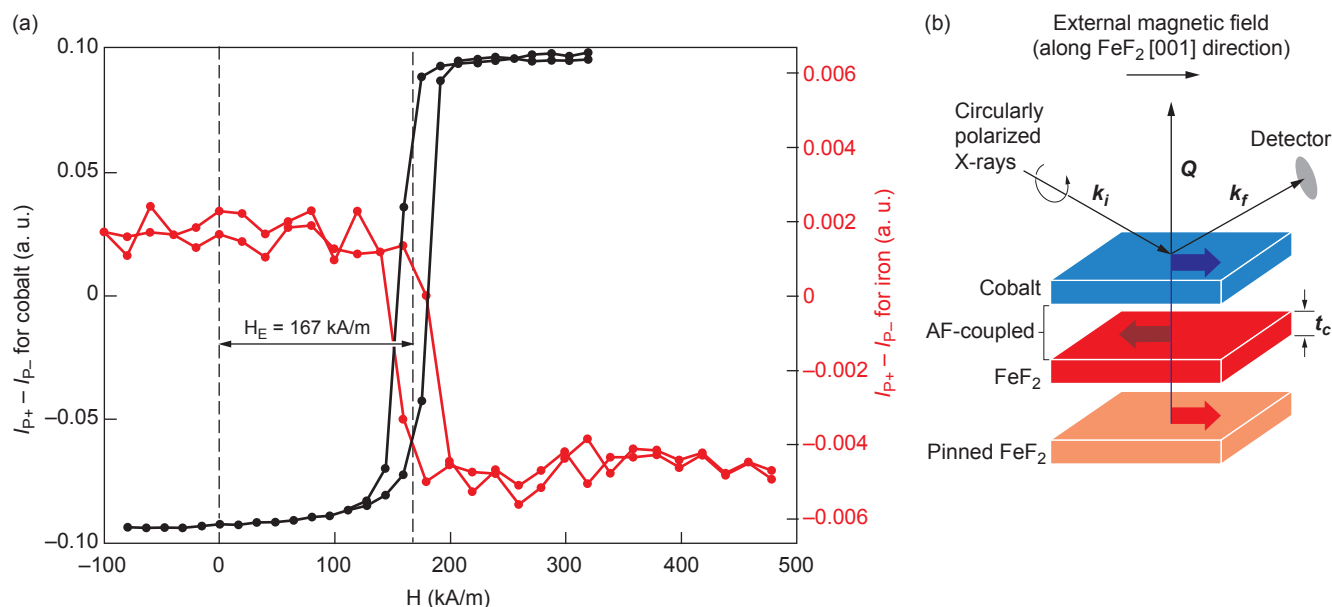
Figure 5. Hysteresis Curves for the Cobalt and Iron Spins
(a) These hysteresis curves were obtained by tuning the incident x-ray energy to the absorption edges of cobalt or iron. I_{P+} and I_{P-} are the intensities of, respectively, the left and right circularly polarized x-rays reflected from the model nanostructure. The offset of 167 kA/m is the exchange bias, a key parameter for a spin-valve read head or an MRAM cell. Note that, in this system, the exchange bias is positive, unlike the negative exchange bias in conventional systems. This result is due to the cobalt and iron spins across the interface being oppositely coupled. (b) This schematic shows the region of the sample near the cobalt–FeF₂ interface. In the cobalt layer (blue), the magnetization follows the external field. In the FeF₂ layer (red), the iron spins are antiparallel to the cobalt spins (believed to be a consequence of antiferromagnetic coupling across the interface); in the orange layer, the iron spins are pinned along the initial direction of the magnetization. In these experiments, the applied external magnetic field was always parallel or antiparallel to the FeF₂ [001] direction and varied in magnitude. The arrows indicate the projections of the magnetizations along the direction of the incident x-rays, which in (b) is shown parallel to the applied magnetic field. We measured the thickness t_c of the red slab to be 2 to 3 nm. (Reprinted with permission from S. Roy et al., *Phys. Rev. Lett.*, 95, p. 047201–2, 2005. Copyright 2005 by the American Physical Society.)

cobalt layer and the antiferromagnetic FeF₂ layer, respectively, as shown in Figure 5. An exchange bias of 167 kA/m in both layers is apparent in the figure. These measurements lead us to conclude that the cobalt and iron spins are likely to be coupled magnetically.

In a second set of x-ray measurements, we applied a field of 796 kA/m parallel and then antiparallel to the [001] direction of the FeF₂ layer and in each case measured the magnetization of the cobalt and iron spins as a function of scattering angle. From those data, we extracted the separate magnetizations of the cobalt and iron spins as functions of depth (Figure 6), from which we conclude that cobalt and unpinned iron spins are antiparal-

lel across the ferromagnetic/antiferromagnetic interface.

From these results, we concluded that a large applied magnetic field caused the cobalt spins to align with the field whereas the uncompensated iron spins near the FeF₂–cobalt interface rotated in a direction opposite to the cobalt spins because of a strong antiferromagnetic coupling across the interface between those two types of spins.

We then employed reflectometry with polarized neutrons to determine the direction, as well as the magnitude, of the magnetization in both layers. The neutron measurements were performed at the ASTERIX spectrometer at LANSCE. We first applied a magnetic field of 438 kA/m in the

[001] direction of the FeF₂ layer while cooling the sample to 10 kelvins. Then, we took neutron scattering data after rotating the sample about its surface normal until the applied field was parallel to the FeF₂'s [110] direction. This procedure intentionally twisted the magnetization in the sample. By examining where the magnetic twist occurred we could identify where the magnetization of the sample was pinned. Figure 7 shows the direction and the magnitude of the sample's magnetization as a function of depth determined from our polarized neutron measurements. While the magnetization in the cobalt layer and near the cobalt–FeF₂ interface is twisted, the net magnetization inside the nominally antiferromagnetic FeF₂
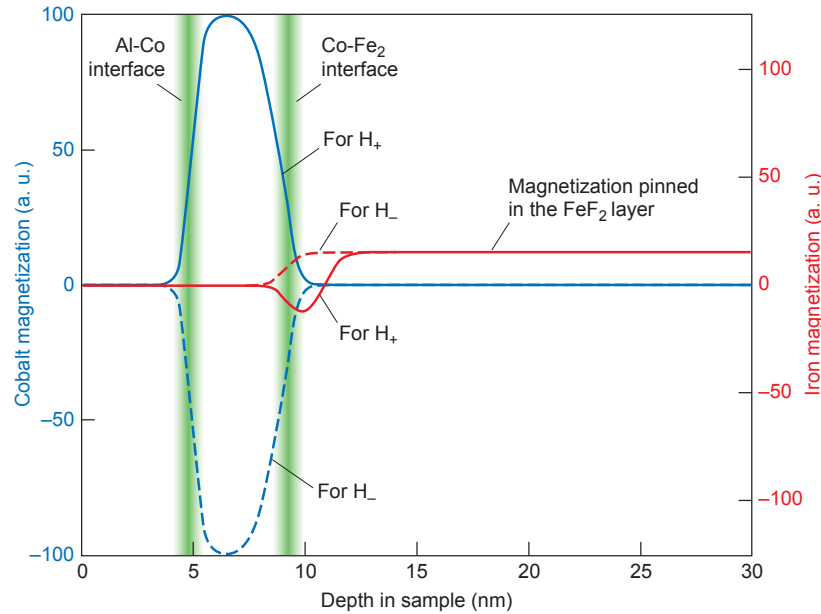
Figure 6. Magnetization Depth Profiles from Resonant-X-Ray Reflectometry
The depth profiles of the cobalt and iron magnetizations were measured for two values of applied magnetic field, H_+ = +796 kA/m and H_- = –796 kA/m. The measurements were taken after the sample had been initially magnetized in a field H_{FC} = 796 kA/m at 20 K along the [001] direction of the FeF_2 crystal. The pinned iron spins in the FeF_2 layer reflect the initial magnetization. The unpinned iron spins in the FeF_2 layer point opposite to the cobalt spins, indicating an antiferromagnetic coupling across the cobalt–FeF_2 interface. (Reprinted with permission from S. Roy et al., *Phys. Rev. Lett.*, 95, p. 047201-2, 2005. Copyright 2005 by the American Physical Society.)
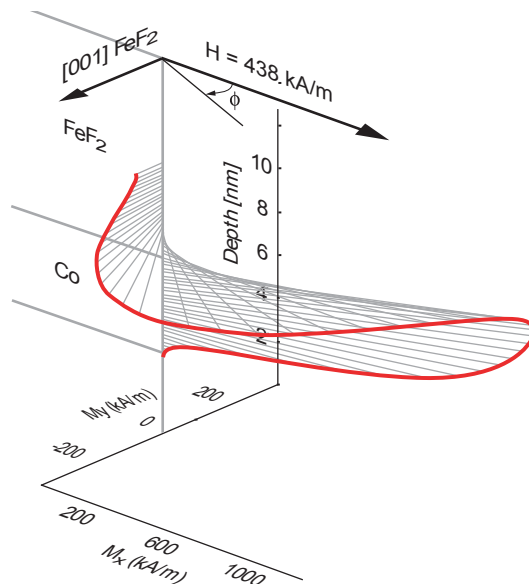
Figure 7. Magnetization Depth Profile from Polarized-Neutron Reflectometry
The depth profile of the magnetization in the cobalt-FeF_2 sample exhibited a twist with respect to the applied field of 438 kA/m because the magnetization inside the FeF_2 layer was pinned or frozen in the [001] FeF_2 direction (parallel to the direction of the field applied to the sample during cooling to 10 K) while the field applied after pinning the magnetization was perpendicular to the [001] direction. (Reprinted with permission from S. Roy et al., *Phys. Rev. Lett.*, 95, p. 047201-3, 2005. Copyright 2005 by the American Physical Society.)

layer was apparently pinned along a particular crystallographic axis, namely, that of the initial magnetization of the sample. In other words, the pinned magnetization could not be moved by the applied magnetic field. We suggest that the net magnetization is due to piezomagnetism, that is, strain-induced magnetization, in FeF_2 and to uncompensated magnetization of misfit dislocations.

The combination of our x-ray scattering study (in which the orientations of unpinned cobalt and iron spins near the interface was identified) and neutron scattering study (in which location of pinned magnetization was identified) enabled us to develop a model of the exchange bias mechanism. Because the loose and pinned iron spins prefer to point in the same direction, the pinned spins inside the antiferromagnet inhibit the response of the loose iron and cobalt spins to small applied magnetic fields. The inhibited response of cobalt spins to the magnetic field is manifested as exchange bias. These somewhat unexpected results go a long way toward yielding a clearer picture of the microscopic origins of exchange bias.

Further Reading

Baibich, M. N., J. M. Broto, A. Fert, F. N. Van Dau, and F. Petroff. 1998. Giant Magnetoresistance of (001)Fe/(001)Cr Magnetic Superlattices. *Phys. Rev. Lett.* **61**: 2472.

Binasch, G. P. Grünberg, F. Saurenback, and W. Zinn. 1989. Enhanced Magnetoresistance in Layered Magnetic Structures with Antiferromagnetic Interlayer Exchange. *Phys. Rev. B* **39**: 4828.

Fitzsimmons, M. R., C. Leighton, J. Nogués, A. Hoffmann, Kai Liu, C. F. Majkrzak, et al. 2002. Influence of In-Plane Crystalline Quality of an Antiferromagnet on Perpendicular Exchange Coupling and Exchange Bias. *Phys. Rev. B* **65**: 134436.

Roy, S., M. R. Fitzsimmons, S. Park, M. Dorn, O. Petracic, I. V Roschcin, et al. 2005. Depth Profile of Uncompensated Spins in an Exchange Bias System. *Phys. Rev. Lett.* **95**: 047201.