Exchange bias in a ferromagnetic semiconductor induced by a ferromagnetic metal: Fe/(Ga,Mn)As bilayer films studied by XMCD measurements and SQUID magnetometry

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We demonstrate an exchange bias in (Ga,Mn)As induced by antiferromagnetic coupling to a thin overlayer of Fe. Bias fields of up to 240 Oe are observed. Using element-specific x-ray magnetic circular dichroism measurements, we distinguish a strongly exchange-coupled (Ga,Mn)As interface layer in addition to the biased bulk of the (Ga,Mn)As film. The interface layer remains polarized at room temperature.

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I. INTRODUCTION

Ferromagnetic (FM) semiconductors offer the prospect of combining high-density storage and gate-controlled logic in a single material. The realization of spin-valve devices from FM semiconductors requires the controlled switching of magnetization in adjacent layers between antiferromagnetic (AFM) and FM configurations. This has motivated several theoretical investigations of interlayer coupling in all-semiconductor devices[1] and AFM coupling has recently been demonstrated in (Ga,Mn)As multilayers separated by *p*-type nonmagnetic spacers.[2] However, the Curie temperature T_C of (Ga,Mn)As is currently limited to 185 K in single layers[3] and is typically much lower for layers embedded within a heterostructure,[2] which is an obstacle to the practical implementation of semiconductor spintronics.

The development of FM metal/FM semiconductor heterostructures has the potential to bring together the benefits of metal and semiconductor based spintronics, offering access to new functionalities and physical phenomena. Recent studies of MnAs/(Ga,Mn)As and NiFe/(Ga,Mn)As bilayer films have shown FM interlayer coupling and independent magnetization behavior, respectively.[4,5] Of particular interest is the Fe/(Ga,Mn)As system since the growth of epitaxial Fe/GaAs(001) films is well established.[6] Remarkably, a recent x-ray magnetic circular dichroism (XMCD) study has shown that Fe may induce a proximity polarization in the near-surface region of (Ga,Mn)As, antiparallel to the Fe moment and persisting even above room temperature.[7] Devices incorporating Fe/(Ga,Mn)As therefore offer the prospect of obtaining nonvolatile room temperature spin polarization in a semiconductor.

Until now, no information has been revealed about the coupling of Fe to (Ga,Mn)As layers away from the near-surface region. At the surface, the (Ga,Mn)As layer may be highly nonstoichiometric and Mn rich due to its nonequilibrium nature.[8–10] Previously, Fe/(Ga,Mn)As layers were produced by a process including exposure to air followed by sputtering and annealing prior to Fe deposition, which may further disrupt the interface order. The origin of the interface magnetism then had to be inferred by comparison to a series of reference samples.[7] Demonstration of coupling between the bulk of the layers, i.e., an exchange bias effect, would provide direct evidence of the interface magnetic order. Moreover, such coupling would offer new means of manipulating the FM semiconductor spin state and utilizing the proximity polarization effect in a spintronic device.

Here, we demonstrate an antiferromagnetic coupling and exchange bias in Fe/(Ga,Mn)As bilayer films by combining element-specific XMCD measurements and bulk-sensitive superconducting quantum interference device (SQUID) magnetometry. As with previous studies of FM metal/FM semiconductor bilayers[4,5] (and in contrast to AFM coupled FM metal/FM metal exchange bias structures[11,12]) the layers are in direct contact without a nonmagnetic spacer in between. We distinguish interface and bulk (Ga,Mn)As layers that are, respectively, strongly and weakly antiferromagnetically coupled to the Fe overlayer. In agreement with Ref. 7, the interface layer remains polarized at room temperature.

II. EXPERIMENTAL DETAILS

The Fe and (Ga,Mn)As layers of the present study were both grown by molecular-beam epitaxy in the same ultrahigh-vacuum system in order to ensure a clean interface between them. The (Ga,Mn)As layer of thickness 10–50 nm was deposited on a GaAs(001) substrate at a temperature of 260 °C, using previously established methods.[3,8] Measurement of the substrate temperature was performed using a band-edge spectrometer under reflection geometry. A low Mn concentration of $x \approx 0.03$ was chosen in order to avoid the formation of compensating Mn interstitials. The substrate temperature was then reduced to ~0 °C, before depositing a 2 nm Fe layer, plus a 2 nm Al capping layer. *In situ* reflection high-energy electron diffraction and *ex situ* x-ray reflectivity and diffraction measurements confirmed that the layers are single crystalline with subnanometer interface roughness.

SQUID magnetometry measurements were performed using a Quantum Design Magnetic Property Measurement System. Mn and Fe $L_{2,3}$ x-ray absorption and XMCD measurements were performed on beamline I06 at the Diamond Light Source and on beamline 4.0.2 at the Advanced Light Source. Total-electron yield (TEY) and fluorescence yield (FY) were monitored simultaneously using the sample drain current and the photocurrent of a diode mounted at 90° to the incident beam, respectively.

III. RESULTS AND DISCUSSION

A. Magnetometry

SQUID magnetometry measurements were first performed on control Al/Fe/GaAs(001) and Al/(Ga,Mn)As/GaAs(001) samples, grown under the same conditions as the bilayers, to determine the magnetic anisotropies of the individual layers and the Curie temperature of the (Ga,Mn)As layer. The Fe film has a uniaxial magnetic anisotropy with easy axis along the [110] orientation similar to previous studies.[6] For the (Ga,Mn)As control samples, there is a competition between cubic and uniaxial magnetic anisotropies, with the former dominant at low temperatures and favoring easy axes along the in-plane ⟨100⟩ orientations and the latter dominant close to T_C (~35 K) giving an easy axis along the $[1\bar{1}0]$ orientation. Figure 1(a) shows magnetization versus temperature curves for a bilayer film containing a 20-nm-thick (Ga,Mn)As layer and for a 20 nm (Ga,Mn)As control sample. The total remnant moment of the bilayer film decreases on cooling under zero magnetic field below the T_C of the (Ga,Mn)As, indicating that this layer aligns antiparallel to the Fe magnetization at zero field. At higher fields, the (Ga,Mn)As moment aligns parallel to the magnetic field and the total magnetization increases on approaching the (Ga,Mn)As T_C from above. The absolute size of the magnetization change between $T=2$ K and $T=T_C$ at zero field and at 500 Oe are comparable, and are also similar to the low-temperature magnetization of the control sample, indicating that the (Ga,Mn)As layer is homogeneously magnetized antiparallel to the Fe layer under zero field in the bilayer sample.

Figure 1(b) shows low-temperature hysteresis loops for the same Fe/(Ga,Mn)As bilayer film and (Ga,Mn)As control sample for field oriented parallel to the [110] axis. A two-step magnetization reversal is observed, indicating different behavior of the Fe and (Ga,Mn)As layers, with the smaller loop attributed to the dilute moment (Ga,Mn)As film. The minor hysteresis loop shown in Fig. 1(b) clearly shows a shift from zero field by a bias field H_E, indicating that the Fe layer induces an exchange bias in the magnetic semiconductor. The shape and size of the minor loop is in agreement with the hysteresis loop for the control (Ga,Mn)As sample, also shown in Fig. 1(b). This again strongly indicates that the exchange bias affects the whole of the (Ga,Mn)As layer in the bilayer sample.

Similar behavior is observed for bilayer samples containing a 10 or 50 nm (Ga,Mn)As layer, with a bias field which is approximately inversely proportional to the thickness d of the ferromagnetic semiconductor layer [Fig. 1(b), inset]. This $1/d$ dependence of H_E was found previously for MnAs/(Ga,Mn)As bilayers[4] and is generally observed in exchanged-biased thin films.[13] From this dependence it is possible to describe the exchange bias in terms of an interface energy per unit area, $\Delta E = M_{FS} H_E d = 0.003$ erg/cm^2. This value is rather small compared to typical exchange bias systems,[13] reflecting the low moment density M_{FS} of the diluted FM semiconductor layer. However, the bias field for a given (Ga,Mn)As thickness is larger than is observed for MnO/(Ga,Mn)As structures[14] while the reproducibility and flexibility of the present structures is much higher due to the single-crystalline ferromagnetic nature of the Fe layer.

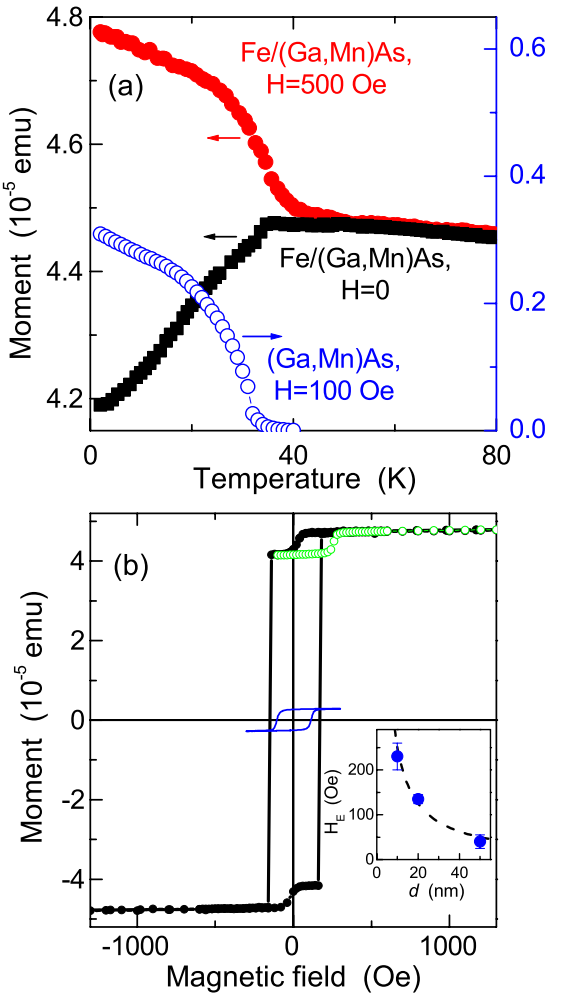

FIG. 1. (Color online) SQUID magnetometry measurements of a Fe (2 nm)/(Ga,Mn)As (20 nm) bilayer film and a (Ga,Mn)As (20 nm) control sample. (a) Moment versus temperature for the Fe/(Ga,Mn)As film at remanence (squares, left axis) and under a 500 Oe applied field (dots, left axis), and for the control sample under 100 Oe (circles, right axis). (b) Major (dots, thick line) and minor (circles) hysteresis loops at 5 K for the Fe/(Ga,Mn)As film for magnetic field applied along the [110] crystalline axis, and the hysteresis loop for the control sample (thin line) along the same axis. The inset of (b) shows the exchange bias field versus the thickness d of the (Ga,Mn)As layer in a series of bilayer films (dots) and a fit showing $1/d$ dependence (dashed line).

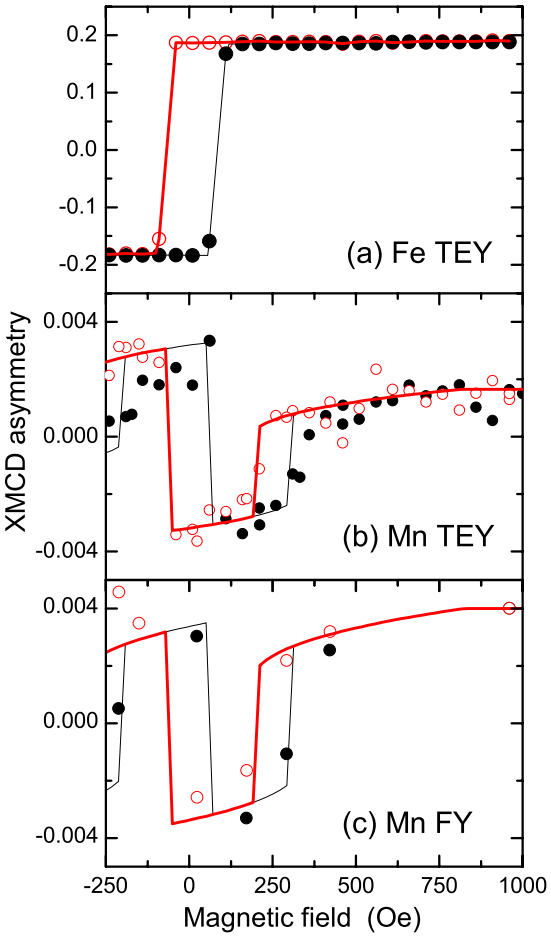

FIG. 2. (Color online) XMCD asymmetry versus applied field along the [110] axis at 2 K for a Fe (2 nm)/(Ga,Mn)As (10 nm) film. (a) Fe L_3, total electron yield; (b) Mn L_3, total electron yield; and (c) Mn L_3, fluorescent yield. Dots and circles are data for increasing and decreasing fields, respectively; lines are to guide the eye.

B. X-ray magnetic circular dichroism

To confirm the presence of AFM interlayer coupling, we performed XMCD measurements at the Mn and Fe $L_{2,3}$ absorption edges in order to determine the magnetic response of the individual elements. In $L_{2,3}$ XMCD, electrons are excited from a $2p$ core level to the unoccupied $3d$ valence states of the element of interest by circularly polarized x rays at the resonance energies of the transitions. The difference in absorption for opposite polarizations gives a direct and element-specific measurement of the projection of the $3d$ magnetic moment along the x-ray polarization vector.[15] The absorption cross section is conventionally obtained by measuring the decay products—either fluorescent x rays or electrons—of the photoexcited core hole. The type of decay product measured determines the probing depth of the technique. For Mn $L_{2,3}$ absorption, the probing depths for FY and TEY detection are $\lambda_{FY} \approx 100$ nm and $\lambda_{TEY} \approx 3$ nm. In the current experiment, the Mn XMCD measured using FY and TEY are thus sensitive to the bulk of the (Ga,Mn)As film and the near-interface layers, respectively.

Figures 2(a)–2(c) shows the magnetic field dependence of XMCD asymmetry, defined as $(I_l - I_r)/(I_l + I_r)$, where $I_{l(r)}$ is the absorption for left (right) circularly polarized x rays. This is measured at the Fe and Mn L_3 absorption peaks for a Fe (2 nm)/(Ga,Mn)As (10 nm) sample at 2 K. The external field is applied along the photon incidence direction, which is at 70° to the surface normal with an in-plane projection along the [110] axis. The XMCD data show that the Fe film displays a square hysteresis loop with a single magnetization switch, as expected for a monocrystalline Fe film with strong uniaxial magnetic anisotropy. The Mn XMCD shows a more complicated loop due to the effect of the interlayer coupling. The projected Mn moment aligns antiparallel to the Fe moment at remanence and undergoes a magnetization reversal of opposite sign to the Fe. With further increase in the external magnetic field, the Mn moment gradually rotates away from antiparallel alignment with the Fe layer and into the field direction. Qualitatively similar behavior is observed for the Fe (2 nm)/(Ga,Mn)As (20 nm) sample: the (Ga,Mn)As layer is aligned antiparallel to the Fe layer at zero field, although the bias field is lower by approximately a factor of 2 and is consistent with the data shown in Fig. 1(b).

Clear differences are observed between the Mn XMCD hysteresis loops obtained using TEY and FY detection modes. For FY the magnitude of the XMCD is similar (but of opposite sign) at remanence and at high magnetic fields, whereas for TEY at remanence it is approximately a factor of 2 larger than at 1000 Oe. The Mn $L_{2,3}$ XMCD spectra recorded at remanence and at 1000 Oe, shown in Fig. 3, confirm this result. At remanence the FY and TEY detected XMCD have similar magnitudes. However, under a large external field the XMCD is substantially smaller in TEY than in FY, confirming that the net magnetization of the Mn ions near the interface is significantly less than in the bulk of the (Ga,Mn)As film. This is the case even up to the highest field applied (20 kOe). By applying the XMCD sum rules[16] to the TEY data and by comparing the spectra to previous measurements on well-characterized (Ga,Mn)As samples,[17] the projected Mn $3d$ magnetic moments are obtained as $-1.4\mu_B$ and $+0.8\mu_B$ per ion at remanence and 1000 Oe, respectively.

The difference between these values can be understood as being due to an interface layer which is strongly antiferromagnetically coupled to the Fe layer. At zero field, both the interfacial and bulk Mn are aligned antiparallel to the Fe layer. At high fields, the bulk of the (Ga,Mn)As layer away from the interface is reoriented into the external field direction. However, the interfacial Mn remains antiparallel to the Fe layer and thus partially compensates the XMCD signal from the bulk of the (Ga,Mn)As. From the size of the remanent and 1000 Oe magnetic moments, it can be estimated that around 25–30 % of the TEY XMCD signal can be ascribed to the interfacial Mn which is strongly coupled to the Fe moments.

The interfacial Mn moments are ascribed to the proximity polarization of the (Ga,Mn)As interface by the Fe layer, such as was shown previously by XMCD as well as *ab initio* theory.[7] Evidence for this can be observed from measurement of the Mn $L_{2,3}$ XMCD signal at temperatures above the (Ga,Mn)As T_C. Similar to the previous study,[7] we observe a small but not negligible signal at room temperature (Fig. 3), with opposite sign to the Fe $L_{2,3}$ XMCD. Its spectral shape is characteristic of a localized electronic configuration close to

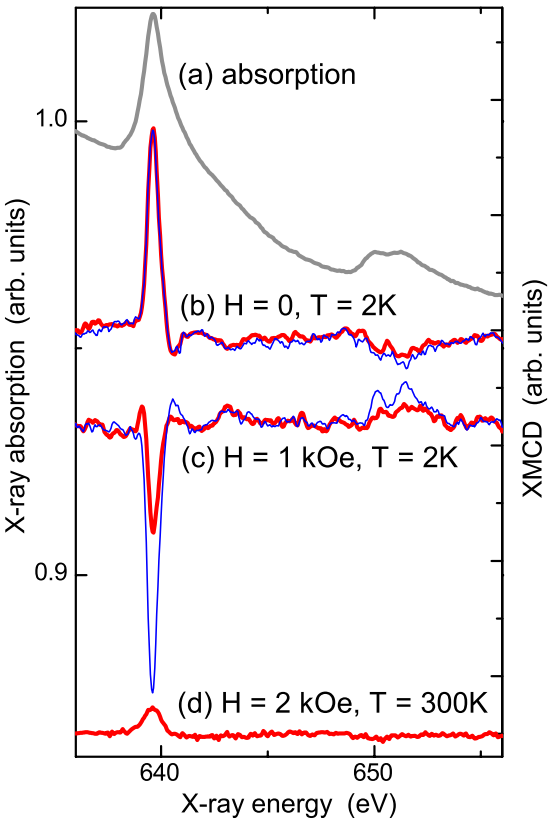

FIG. 3. (Color online) (a) Polarization-averaged Mn $L_{2,3}$ spectrum for a Fe/(Ga,Mn)As film; (b) XMCD spectra measured in remanence at 2 K; (c) XMCD spectra measured under a 1000 Oe applied field at 2 K; and (d) XMCD spectrum measured under a 2000 Oe applied field at 300 K. XMCD spectra are obtained using TEY (thick red lines) and FY (thin blue lines) detection.

d^5, similar to bulk (Ga,Mn)As (Refs. 7, 9, 10, and 17) but in contrast to Mn in more metallic environments such as Mn_xFe_{1-x} (Ref. 7) or MnAs.[18] A slight broadening is observed on the low-energy side of the Mn L_3 peak, which may be due to the different screening induced by proximity to the Fe layer. Since the measured intensity is attenuated with distance z from the surface as $I=I_0 \exp(-z/\lambda_{TEY})$, the thickness of the strongly coupled interface layer is estimated to be ~0.7 nm or 2–3 monolayers, assuming a uniform distribution of Mn ions and magnetic moments throughout the (Ga,Mn)As film.

IV. CONCLUSIONS

We have investigated the magnetic coupling between metal and semiconductor layers in a series of Fe/(Ga,Mn)As bilayer films. Bulk sensitive SQUID magnetometry measurements, as well as measurements of the magnetic response of the individual layers using XMCD, demonstrate an exchange bias effect in the (Ga,Mn)As induced by the antiferromagnetic coupling to the neighboring Fe layer. Comparison of the magnetic signal to that of a control film indicates that the bias field affects the whole of the (Ga,Mn)As layer, such that the layer is homogeneously magnetized antiparallel to the Fe layer at low magnetic fields. Furthermore, we have found that while the bulk of the layer reorients to parallel alignment when an external field large enough to overcome the bias field is applied, the Mn moments within a subnanometer interface layer remain aligned antiparallel to the Fe layer even for the largest external fields investigated. The interface Mn moments are polarized at temperatures well above the T_C of the bulk (Ga,Mn)As layer. This confirms the recently reported observation of room-temperature proximity polarization at the Fe/(Ga,Mn)As interface,[7] as well as demonstrating that the coupling at the Fe/(Ga,Mn)As interface influences the magnetization orientation of the whole of the (Ga,Mn)As layer at low temperatures. The estimated thickness of the strongly coupled interface region is around a factor of 3 smaller than in Ref. 7, which may be related to the different preparation method of the interface (*in situ* growth in the present case vs air exposure followed by sputtering and annealing in Ref. 7). It is, however, comparable to the typical depletion depth in a (Ga,Mn)As film with hole density $\sim 10^{20}$ cm^{-3}, which may be a factor underlying its relatively weak coupling to the bulk of the (Ga,Mn)As layer.

Our results shed light on the magnetic coupling in Fe/(Ga,Mn)As hybrid layers which are of potential interest for room-temperature spintronics and also offer a means of controlling the spin orientation in a FM semiconductor. The magnitude of the exchange bias is larger than what has been previously observed in (Ga,Mn)As heterostructures with an antiferromagnetic pinning layer[14] and furthermore is readily controllable by reorientation of the Fe magnetization. Such layers may find future applications in heterostructures for spin injection or the investigation of magnetoresistance effects. Future efforts should be directed at determining the underlying mechanism of interfacial coupling and proximity polarization, and their influence on spin transport in devices.

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