Magnetic structure of Fe/Cr/Fe trilayers

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Soft x-ray magnetic circular dichroism (SXMCD) is used to determine independently the magnetic behavior of each layer of Fe/Cr/Fe trilayers deposited on GaAs(001). Submonolayer coverages of Cr on Fe(001) films are found to be ferromagnetic and antialigned to the Fe moment. Subsequent Cr deposition shows a monotonically decreasing total moment, consistent with island growth of ferromagnetic Cr sheets antialigned with adjoining Cr layers. For thin Cr interlayers, the second Fe film is antialigned with the first Fe film, but a second Fe film of less than 2-ML thickness shows no SXMCD signal.

Extensive work has been ongoing on a class of magnetic multilayer systems that exhibit oscillatory coupling between ferromagnetic films separated by a nonmagnetic or antiferromagnetic spacer layer.[1-6] The first system to exhibit this coupling, and by far the most thoroughly examined, was trilayers (and superlattices) of Fe/Cr/Fe.[1,3,7,8] The observed coupling can be quite complicated, simultaneously displaying both short- and long-period oscillators[8] as well as regions of biquadratic (90°) coupling.[8-10] If these structures can be electrically isolated (by deposition on a semi-insulating substrate, e.g., GaAs), they exhibit technologically interesting magnetoresistance[3,11-13] and thermoelectric power[14] properties in addition to the unique coupling behavior.

Although a great deal is known concerning the behavior of the ferromagnetic layers, very little has been done to investigate the magnetic structure of the interlayer spacer. Prior to the interlayer coupled systems, Victora and Falicov[15] theoretically examined a monolayer of Cr/Fe(001) and determined it to be ferromagnetic with a large spin imbalance ($3.6\mu_B$), but aligned antiferromagnetically with respect to the bulk Fe. In an attempt to explain the mechanism underlying the coupling, subsequent calculations for Fe/Cr/Fe(001) trilayer structures also predicted ferromagnetic Cr layers adjoining either Fe film, which are antialigned to the Fe.[16,17] In addition, these calculations predicted that interior Cr layers were ferromagnetic and antialigned with adjoining Cr layers. There is recent experimental evidence from spin-resolved core-level photoemission of Jungblut *et al.*[18] which does indicate that the Cr is ferromagnetic and aligned opposite the Fe moment. Similar conclusions were found by spin-polarized electron-energy-loss measurements[19] and spin-resolved valence-band photoemission.[20] Still, element-resolved magnetic-structure determinations down to submonolayer Cr coverages are needed to unambiguously identify the magnetic structure of the interlayer.

In this paper we report on soft-x-ray magnetic circular dichroism (SXMCD) measurements on Fe/Cr/Fe trilayer structures deposited on GaAs(001). The SXMCD is the difference between the absorption cross section of positive and negative helicity (left and right circularly polarized) soft x rays at inner-shell absorption white lines of magnetic systems, such as the $2p \rightarrow 3d$ excitations of transition metals and the $3d \rightarrow 4f$ excitations of rare earths.[21] These strong dipole-allowed excitations directly probe the electronic states of magnetic interest and thus give element-specific magnetic information with high detection sensitivity, uniquely suitable to independently determine the magnetic orientation of the Cr and Fe layers down to submonolayer coverages.[22]

The experiments were conducted at the AT&T Bell Laboratories Dragon Beamline at the National Synchrotron Light Source (NSLS), which has recently been modified to produce simultaneously two soft-x-ray beams of opposite helicity.[23] The development and characterization of the epitaxial, single-crystal trilayer Fe/Cr/Fe structure has been described in detail elsewhere.[24] Polished 1×1 cm^2 GaAs(001), $n+$ substrates are lightly sputtered at 500 V with Ne^+ to remove surface contamination. The substrates are then heat cleaned at 580 °C to generate an oxide-free GaAs(001) surface. Fe is deposited from an e^- beam evaporator with the substrate held at 175 °C and at a vacuum of 2×10^{-10} Torr until a 150-Å Fe film is developed. The total carbon contamination is less than 0.01 ML as determined by C_{1s} x-ray photoelectron spectroscopy.

The geometry of the SXMCD measurement is shown in Fig. 1. Because the moments are anticipated to be in

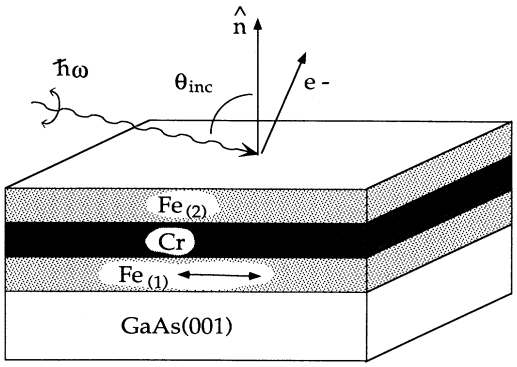

FIG. 1. Geometry of the Fe/Cr/Fe trilayer SXMCD measurement. The remanent magnetization direction is defined by the thicker Fe layer ($Fe_{(1)}$).

the film plane, we have used a glancing-angle geometry for the trilayer structures. For these measurements, the angle of incidence (θ_{inc}) is set at 65°; the degree of circular polarization is set at 90%, as independently measured by a crystal polarimeter; and the photon energy resolution is set to 0.4 eV. The films were remanently magnetized along the magnetically easy ⟨100⟩ axis[25] by an *in situ* pulse coil. The photon energy is swept through the $L_3(2p_{3/2} \rightarrow 3d)$ and L_2 $(2p_{1/2} \rightarrow 3d)$ absorption white lines of the element of interest. The absorption intensity is measured by recording both the total electron yield using a channeltron and the Auger (LMM) electron yield using a hemispherical electron-energy analyzer equipped with a multichannel detector. Measurements of the four combinations of the magnetization direction and photon helicity have been taken to rule out possible instrumental artifacts.

To investigate the magnetic structure of the trilayer system, we have made SXMCD measurements at various stages of the multilayer development. Figure 2(a) shows the room-temperature x-ray-absorption spectroscopy (XAS) and magnetic circular dichroism (MCD) of the L_3 and L_2 white lines of the as deposited first Fe film. The solid line is taken with the spin direction of the photons parallel to that of the majority electrons of the Fe film (remanent magnetization direction), while the dashed line is taken with antiparallel spin directions. The dots are the difference between the parallel and antiparallel spectra. These spectra are very similar to those of bulk Fe,[21] showing a negative MCD signal at the L_3 white line and a positive signal at the L_2 white line. After correcting for the incomplete polarization and incident angle of the soft x rays (multiplication factor of 1.23), the MCD to XAS peak height ratios are found to be 24% and 28% for the L_3 and L_2 white lines, respectively, which are the values associated with a fully magnetized single-domain Fe sample.[21] To facilitate the following discussion, we defined the remanent magnetization direction of the first Fe film as shown in the inset of Fig. 2(a).

The Cr is slowly deposited on top of the room-temperature Fe film. Our studies using auger electron spectroscopy, reflection high-energy electron diffraction, and auger electron diffraction show that the Cr does not grow on Fe/GaAs(001) films in a layer-by-layer fashion, as occurs for deposition onto single-crystal Fe whiskers,[8] but forms islands (regardless of substrate temperature). This difference is attributed to the very different starting surface for the Fe thin films compared to the Fe whiskers. Scanning tunneling microscope (STM) images of unstressed Fe whiskers show the Fe surface to consist of long, flat terraces, whereas STM images of thin Fe/GaAs(001) surfaces display an atomically rough surface.[26] These starting surface differences result in different growth modes which are then reflected in the

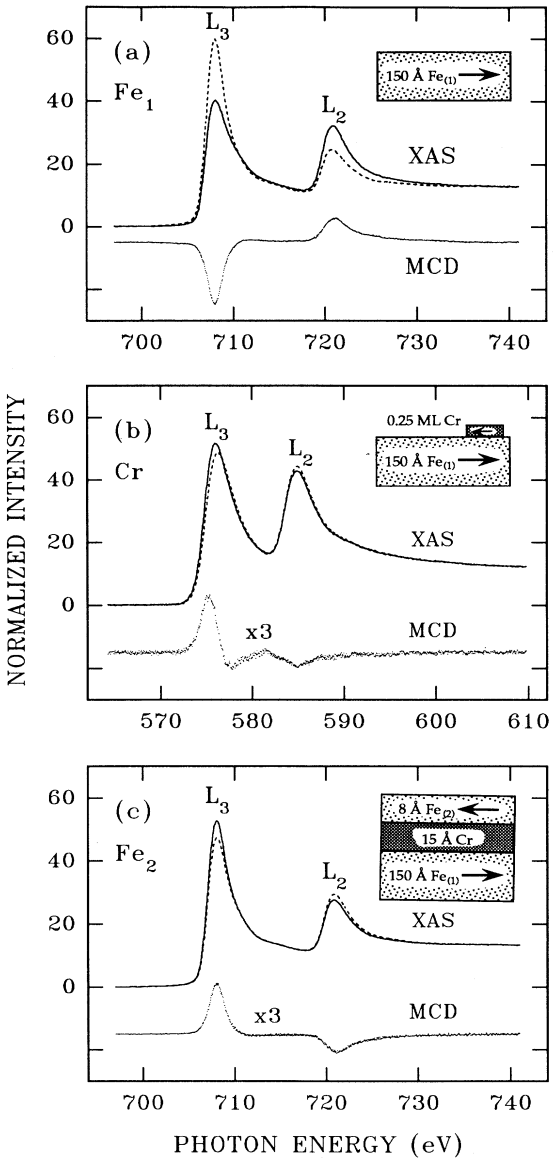

FIG. 2. X-ray-absorption and magnetic circular dichroism spectra for (a) the first Fe film 150 Å thick, (b) 0.25-ML Cr deposited atop the first Fe film, and (c) the second Fe film 8 Å thick atop a 15-Å Cr interlayer. The solid (dashed) lines are spectra taken with the spin direction of the incident photons parallel (antiparallel) to that of the majority electrons of the first Fe film. The dots are the MCD spectra obtained from the difference between the parallel and antiparallel XAS spectra. The ordinates of the MCD spectra have been arbitrarily shifted.

magnetic behavior of the coupled films. Trilayers of Fe/Cr/Fe generated on an Fe whisker show both long-period and short-period oscillations[8] in the coupling, whereas the Fe/Cr/Fe trilayers grown on GaAs(001) at these temperatures display only the long-period oscillatory coupling.

To investigate the magnetic orientation of the first monolayer of Cr, spectra for Cr coverages down to 0.25 ML were recorded. Figure 2(b) shows XAS and MCD spectra of the lowest-coverage 0.25-ML Cr film, obtained from the sum of multiple scans (45 min in total). At 0.25-ML coverage, nearly all the deposited Cr should occupy the first layer with very little in second-layer sites. Since SXMCD is element specific, it is immediately evident from the reversal of the Cr MCD intensity at both the L_3 and L_2 white lines [shown in Fig. 2(b)] in comparison to the Fe MCD spectra [shown in Fig. 2(a)], that submonolayer coverages of Cr are ferromagnetic and antialigned with the first Fe layer [as depicted in the inset of Fig. 2(b)]. In addition to the reversal of the MCD signal, the Cr data also show a strong derivative line shape at the L_3 white line, which is due to a peak-energy shift between the two XAS spectra, resembling the result of a recent atomic multiplet and crystal-field calculation.[27] Recent MCD measurements of Co/Pd multilayers[28] have used a sum-rule procedure developed by Thole *et al.*[29] to extract the orbital contribution to the moment. In addition to the orbital sum rules, sum rules for the spin contribution have also been derived by the same group.[30] Although these sum rules show promise in separating the spin and orbit contributions to the moment, difficulties with proper background subtraction have limited their use with our data. Instead, we determine the total Cr moments from a direct comparison to two different theoretical models.

For the 0.25-ML Cr data, the MCD to XAS *peak height* ratios are 7.2% and 3.5% for the L_3 and L_2 white lines, but *peak area* ratios are 2.6% and 3.6%, respectively. The L_3 to L_2 MCD peak area ratio is found to be -1.3. Utilizing these peak area ratios and comparing to the branching ratios tabulated in Ref. 27 for various d^4 and d^5 configurations for *bulk* Cr, the averaged Cr magnetic moment is estimated to be $0.6\pm0.2\mu_B$/atom. An independent estimation based on the exchange-split valence-band model,[31] using the band structure of *bulk* Cr, gives similar values. This magnetic moment is much smaller than the $3.6\mu_B$ value of Victora and Falicov for 1-ML Cr on Fe(001),[15] but in good agreement with the maximum bulk value of $0.59\mu_B$.[32] This disagreement may be from the use of bulk Cr theoretical data for our comparisons instead of data for 1 ML of Cr/Fe(001). First principles theoretical MCD calculations are needed to extract a more precise value from the data.

The additional deposition of Cr results in a continual reduction in the MCD to XAS peak height ratio, as shown in Fig. 3. This is strong evidence that subsequent Cr deposition results in an antiferromagnetic structure consisting of ferromagnetic sheets, antialigned with adjacent Cr layers. The thickness dependence of the Cr MCD signal is compared to two theoretical models for the Cr behavior. For Cr films that grow layer-by-layer

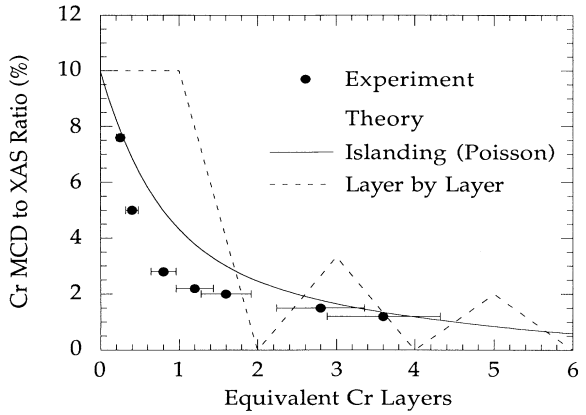

FIG. 3. Cr L_3 MCD-to-XAS peak height ratios as a function of Cr deposition (equivalent monolayers). Solid dots are experimental values, dashed line is theoretical model for antialigned ferromagnetic sheets assuming layer-by-layer growth, and solid line is a similar theoretical model but assuming island growth (as described by a Poisson distribution).

and have equal and opposite moments at adjoining Cr layers, the total averaged Cr film moment (proportional to the normalized MCD-to-XAS ratio) should oscillate between zero and a reducing value (as shown by the dashed line). If the film instead grows as islands (as described by a Poisson distribution, valid if no surface diffusion of the Cr occurs), then the total Cr moment diminishes monotonically. Our experimental data also falls monotonically (consistent with the observed island growth of the Cr on Fe/GaAs films), but falls more rapidly than the Poisson distribution allows. This suggests that although the magnetic structure of the Cr film is well represented by alternating ferromagnetic sheets antialigned with adjoining layers, the Cr film growth is more severe than a Poisson distribution. This could only occur if the Cr surface diffusion promotes three-dimensional island formation over layer-by-layer growth, resulting in a larger second- and third-layer occupancy than statistical (which results from no Cr surface diffusion). Still, the data clearly shows that the first layer of Cr is ferromagnetic. Although this magnetic structure is consistent with the behavior of bulk Cr(001) surfaces, which also consist of alternating magnetic sheets,[33,34] other mechanisms may be responsible for this decline in the Cr MCD intensity. Similar structures of alternating ferromagnetic sheets, antialigned with adjacent layers have been observed very recently for thick Cr films by scanning electron microscopy with polarization analysis[35] and spin-polarized EELS.[19]

After the growth of 15 Å of Cr onto the first Fe film, deposition of the second Fe layer is performed (also at room temperature). The XAS and MCD spectra for 8 Å of second-layer Fe is shown in Fig. 2(c). The second layer of Fe has a MCD signal, but reversed from the first Fe layer, indicating the thin second Fe layer is ferromagnetic and antiparallel with the first, thicker Fe film. The magnetic orientations, depicted in the inset of Fig. 2(c), confirms the antiferromagnetic coupling between the two Fe layers. The measured MCD is smaller than expected

because the probing depth of the auger electron measurement is larger than the interlayer Cr thickness. Therefore, the spectra of the second Fe film includes a contribution from the deeper-lying first Fe film. Because the magnetic direction of the underlying Fe film is opposite the surface Fe, the measured MCD is reduced from the actual value. This can be demonstrated by measuring the Fe MCD spectra using the total electron yield method, which has a much larger probing depth, and noting that the MCD is further reduced (actually reversing sign). For thicker Cr interlayers (35 Å), identified as being in the ferromagnetic coupling regime,[1,7,8] the MCD spectra of the second Fe film (in this case, there is no contribution of the deeper-lying first Fe film) determines the magnetic orientation of the two ferromagnetic films to be aligned. For a second-layer thickness below 2 ML, no MCD signal for the second Fe film is detected. This is probably due to a reduced Curie temperature for the very thin film and/or a perpendicular moment orientation.

In conclusion, the element specific SXMCD data for Fe/Cr/Fe trilayers clearly displays the antiferromagnetic coupling of the two Fe films. Cr deposition of Fe(001) results in ferromagnetic sheets of Cr layers antialigned to the Fe film and antialigned with adjacent Cr layers. The MCD measurements of submonolayer Cr on Fe suggests a Cr magnetic moment much smaller than the predicted value but close to that of the saturated bulk Cr value. These measurements demonstrate that SXMCD is a powerful tool for probing magnetic structures of multicomponent thin films, even at submonolayer coverages.

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