Oscillations with a Period of Two Cr Monolayers in the Antiferromagnetic Exchange Coupling in a (001) Fe/Cr/Fe Sandwich Structure

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Oscillations with a period of two Cr monolayers were measured in the antiferromagnetic exchange coupling between Fe magnetizations separated by Cr. The coupling was measured on a sample consisting of a Au(20 Å)/Fe(50 Å)/Cr(0–18 Å wedge) film deposited by a molecular-beam epitaxy on a Fe[100] single-crystal "whisker." The Cr and Fe grew monolayer by monolayer as monitored by reflection high-energy-electron diffraction. The coupling was determined from the magneto-optic Kerr effect. It was antiferromagnetic for 5–18 Å Cr with four strong peaks and a maximum value of -0.60 mJ/m^2 for eight monolayers of Cr.

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One of the most exciting recent discoveries in the field of ultrathin magnetic films is the existence of antiferromagnetic exchange coupling between Fe layers separated by Cr layers (Grünberg *et al.* [1]). It was subsequently reported by Parkin, More, and Roche [2] that the coupling shows several oscillations in magnitude as a function of the Cr thickness with a period of roughly 15–20 Å. These results were further refined [3] to show that the coupling oscillates between antiferromagnetic (AF) and ferromagnetic (F) coupling. This was done in an appealing way by growing a Fe/Cr/Fe sandwich structure in which the Cr layer thickness varied linearly from 0 to 45 Å. Using such a wedge structure the complete coupling behavior as a function of Cr thickness could be investigated on a single sample. In this Letter we report the experimental discovery of additional oscillations in the AF coupling with a shorter period equal to two Cr monolayers (ML) in a sample consisting of an epitaxial bcc Fe/Cr film of varying Cr thickness grown on the (001) face of a Fe[100] single-crystal whisker.

A simple consideration of the antiferromagnetism of bulk Cr together with direct *d-d* exchange at the Fe-Cr interface would lead one to expect the exchange coupling to oscillate between antiferromagnetic and ferromagnetic with a period of two Cr monolayers. Band-structure calculations [4] give contradictory predictions of the existence of these two-monolayer oscillations. Model calculations [5] predict a thickness dependence of the coupling consisting of two-monolayer oscillations superimposed on a longer-wavelength oscillation. The presence of interface roughness was invoked to explain the absence of two-monolayer oscillations in the existing experimental data. The motivation behind this study was to try to produce samples with sharper interfaces that might display the two-monolayer oscillations. As shown below, this was clearly achieved in our sample because of the high degree of perfection of the Fe whisker substrate and the sharpness of the Fe/Cr interfaces.

Several Fe[100] whiskers were used for growth studies and one in particular for the coupling measurements. That whisker had a length of 10 mm and a square cross section of 0.5×0.5 mm^2. The long axis was the [100] direction and the faces were {100} planes to a high degree of accuracy. The epitaxial layers were deposited in a multichamber molecular-beam-epitaxy system (VG Semicon V80M) with a base pressure of $\sim5\times10^{-11}$ mbar. The Fe whiskers were subjected to several cycles of Ar-ion sputtering at 750 °C and were then given a final anneal of 30–40 min also at 750 °C. Auger electron spectroscopy (AES) showed the presence of carbon ($<2\%$) and oxygen ($<1\%$) which are the common impurities that segregate to the surface of Fe during annealing. The depositions were monitored by reflection high-energy-electron diffraction (RHEED). The oscillations in the RHEED intensity that occur during monolayer-by-monolayer growth were measured. Low-energy-electron diffraction (LEED) and AES were done after the growths as a function of Cr thickness and on the Fe overlayer. The Cr was evaporated at ~1 Å/min from a simple evaporation furnace with the Fe substrate at both room temperature and 150 °C for growth studies while a sample grown at 150 °C was used for coupling measurements. The wedge shape was formed by slowly moving a shutter across the whisker during evaporation (wedge slope $=0.45$ ML/ mm). The position of the start of the Cr wedge was determined by AES. This also allowed a comparison of thicknesses determined by AES and the RHEED oscillations. The Fe was deposited at ~6 Å/min from an electron-beam evaporator. The sample that was later measured for coupling was rotated during the Fe deposition to ensure a uniform thickness over the sample. RHEED oscillations were also measured during several Fe depositions. The Fe/Cr film was covered with a 20-Å Au overlayer evaporated from a Knudsen cell for protection from the atmosphere. The value of the AF coupling was determined by analysis of hysteresis loops measured outside the vacuum by the longitudinal magneto-optic Kerr effect. The light source was a focused helium neon laser with ~70 μm spot size directed at 50° with respect to the surface normal. This gave a resolution of ~0.16 ML Cr. Kerr measurements were made with the plane of incidence both parallel and perpendicular to the long axis of the whisker to determine the direction of the magnetizations versus the applied magnetic field. The coupling was then determined as a function of Cr thickness at many positions of the laser spot along the whisker. The thicknesses of the overlayers were such that the Kerr effect measurement was sensitive to the overlayer and the top of the Fe whisker in a ratio of ~1:2.

The high quality of the surfaces of properly prepared Fe whiskers and their suitability as a substrate for epitaxial growths has been previously investigated by RHEED [6]. An additional important advantage in the Fe/Cr system is the very small misfit of 0.6% between the bcc Fe and Cr lattices which allows excellent epitaxial growth. In Figs. 1 and 2 we show some examples of RHEED and LEED results. Figures 1(a) and 1(b) show the RHEED pattern from a whisker before and after the deposition of six monolayers of Cr at room temperature. The bright spots correspond to the intersection of the Ewald sphere with the classic k-space rods of a two-dimensional array. The diffraction features are sharper than reported for other metallic systems and rival the best measurements on semiconductors. The RHEED pattern from the Cr overlayer also displays bright spots indicating that the long-range atomic order of the substrate is preserved. The intensity is somewhat weaker than from the uncovered Fe and there are faint long bands superimposed over the bright spots (not visible in the photos) due to an increase in disorder of the surface. A precise analysis of the surface disorder by RHEED is a difficult task and beyond the scope of this paper. However, as shown below, the surface quality is good enough to allow the observation of oscillations in the exchange coupling corresponding to two Cr monolayers. Figures 1(c) and 1(d) show photos of LEED patterns at 50 eV from the bare Fe substrate and with a 10-ML Cr overlayer deposited at 150 °C. The sharpness of the LEED spots also demonstrate the quality of the epitaxial growth.

In Fig. 2 we show RHEED oscillations measured during the deposition of Cr on Fe at room temperature and at 150 °C, and during the growth of Fe on the room-temperature-deposited Cr layer. The large oscillations of the room-temperature depositions are strong evidence of substantial monolayer-by-monolayer growth which is a necessary condition for the observation of the two-monolayer coupling oscillations. The RHEED oscillations during the Fe growth are in phase with the corresponding Cr growth, showing that the first monolayer of the Fe overlayer is a continuation of the last monolayer of the Cr. Although the existence of the RHEED oscillations shows a periodic filling of monolayers, it also implies a certain roughness caused by at least one-monolayer-high island formation at partially filled monolayers. To promote a more ideal growth from step edges we have

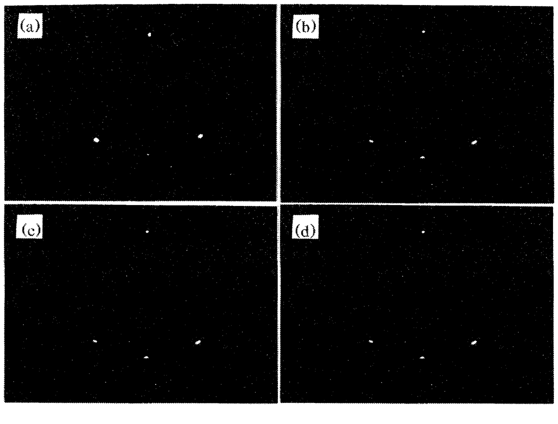

FIG. 1. RHEED patterns of the (001) surface of (a) an uncovered Fe[100] whisker, and (b) a Fe[100] whisker with a 6-ML Cr overlayer deposited at room temperature. The 30-keV electron beam was directed along the [010] azimuth with a 2° angle of incidence. The upper spot in (a) and (b) is the fraction of the straight-through beam that does not strike the sample, and the central lower spot is the specular-reflected beam. LEED patterns at 50 eV: (c) uncovered Fe whisker, and (d) whisker with a 10-ML Cr overlayer deposited at 150 °C.

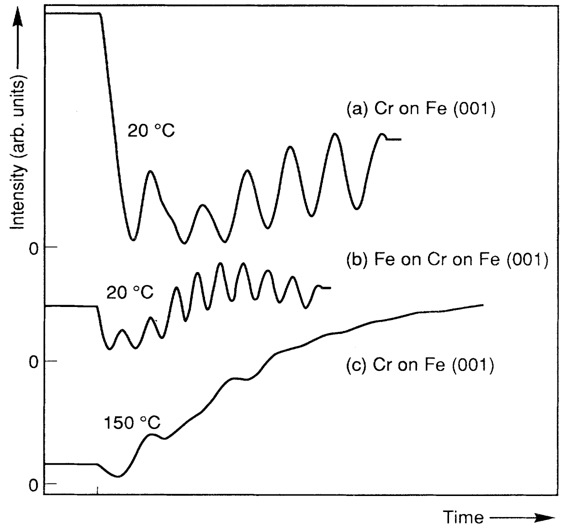

FIG. 2. RHEED oscillations in the specular-reflected beam during deposition: (a) Cr growth at room temperature, and (b) Fe growth on the Cr layer of (a). Note that the phase of the oscillations is maintained. (c) Cr growth at 150 °C. The rate was ~1 ML/min for the Cr, and ~6 ML/min for the Fe depositions.

raised the substrate temperature to 150 °C for the sample that was characterized by the Kerr effect. As seen in Fig. 2 the RHEED oscillations are weaker than at room temperature. This is in accordance with an enhanced growth from existing step edges as opposed to island nucleation on the terraces [7]. Another possible mechanism for the decrease of the amplitude of the oscillations could be interdiffusion of the Fe and Cr. However, the thicknesses determined by AES and RHEED oscillations agree very well, with the AES-determined thicknesses being ~10% higher. This rules out a large amount of interdiffusion. Thus we have produced by monolayer-by-monolayer growth a sample with good surface quality, necessary for the observation of the two Cr monolayer exchange-coupling oscillations.

In Fig. 3 we show a hysteresis loop measured by the longitudinal Kerr effect with the magnetic field and the plane of incidence of the laser beam along the long axis of the whisker for a Cr thickness of 6.4 ML at which AF coupling is present. To first order this is a measure of the magnetization parallel to the long axis of the whisker within the depth of penetration of the laser light and, as stated above, it is sensitive to both the Fe overlayer and the top of the whisker. The relative directions of the overlayer and whisker magnetizations are also shown. The hysteresis in the loop is created by the coercivity of the whisker and for most measurements was smaller than in this figure. The existence of the state where the overlayer magnetization is perpendicular to the axis of the whisker was confirmed by Kerr measurements with the plane of incidence perpendicular to the long axis of the whisker.

The loops can be characterized by two critical fields in the case of AF coupling: one at which the overlayer magnetization switches away from antiparallel to the whisker magnetization (H_1) and one at which it rotates towards parallel to the whisker magnetization (H_2). Halfway between H_1 and H_2 the overlayer magnetization is approximately perpendicular to the whisker magnetization. In Fig. 4 we plot H_1 and H_2 as a function of Cr thickness. Both H_1 and H_2 show five oscillations in magnitude with a period of two monolayers of Cr. The agreement with thicknesses from RHEED oscillations is ~6% and the uncertainty in the zero position is ~0.5 ML. The first peak appears at 4 ML of Cr and the last one at 12 ML. We have found only AF coupling above ~4 ML. The Kerr effect cannot be used to measure the coupling in the ferromagnetic region in this type of sample.

We must consider the magnetization process before we can determine the value of the coupling from the Kerr loops. The whisker and the overlayer have easy magnetocrystalline anisotropy axes in the ⟨100⟩ directions. Shape anisotropy causes the overlayer magnetization to lie in the plane and the whisker magnetization to lie along the long axis except near the ends where there are closure domains. Application of a small field along the whisker is sufficient to saturate the middle section of the whisker. With AF coupling the overlayer points antiparallel to the saturated whisker at low fields and parallel at high fields.

To quantitatively explain the hysteresis loops and in particular the fact that there is a range of field values for which the overlayer magnetization can lie approximately perpendicular to the whisker axis, it is necessary to allow for a slight rotation of the magnetization at the surface of the whisker [8]. This rotation creates a partial domain wall at the surface of the whisker and it is stabilized by the AF coupling to the overlayer. The rotation relaxes to the field direction away from the interface with the characteristic length of a domain wall (~500 Å). The energy of the wall can be analytically calculated for small rotation at the top whisker surface from simple domain-wall theory including the field energy. The total energy

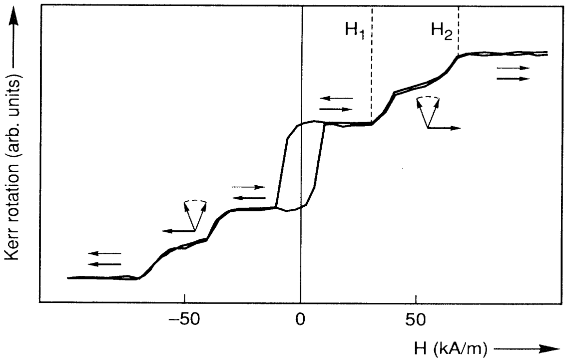

FIG. 3. Longitudinal Kerr hysteresis loop measured from a Au(20 Å)/Fe(50 Å)/Cr(0–18 Å wedge)/Fe[100]-whisker sample. The thickness of the Cr was ~6 ML at this position of the laser spot. The plane of incidence of the light was parallel to the long axis of the whisker. The thin (thick) arrow indicates the direction of the Fe overlayer (whisker) magnetization.

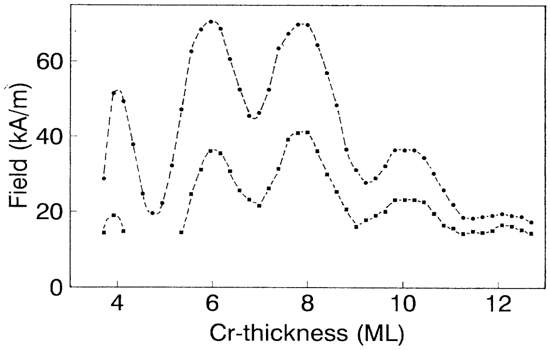

FIG. 4. The Cr thickness dependence of the critical fields, H_1 (lower curve) and H_2 (upper curve), defined in Fig. 3 at which the overlayer magnetization rotates with respect to the whisker magnetization (see text). The large oscillations in H_1 and H_2 have a period of two Cr monolayers and correspond to oscillations in the interlayer coupling. The Cr was deposited at 150 °C.

per unit area of the system assuming the bulk of the whisker is already saturated is

$$E=-HM_s d_1\cos(\theta_1)-J\cos(\theta_0-\theta_1)+\tfrac{1}{4}K_1 d_1\sin^2(2\theta_1)+[A(K_1+\tfrac{1}{2}HM_s)]^{1/2}\theta_0^2\,. \qquad (1)$$

H is the applied field, M_s is the saturation magnetization of Fe, d_1 is the thickness of the Fe overlayer, A is the interlayer coupling energy per unit area, K_1 is the fourth-order anisotropy constant assumed to be that of bulk Fe, A is the exchange constant of bulk Fe, and θ_0 and θ_1 are the angles between the field direction and the magnetizations of the top surface of the whisker and the Fe overlayer, respectively. The last term is the energy in the partial domain wall. By minimizing the energy given in Eq. (1), with J the only free parameter, we could reproduce the experimentally observed magnetization behavior including the perpendicular state of the overlayer. It can be shown [8] for small θ_0 that J can be approximated by

$$J\cong-\tfrac{1}{2}d_1(H_1+H_2)M_s\,. \qquad (2)$$

Using this expression the couplings can be easily calculated from Fig. 4, and it is clear that the coupling oscillates with the same period as H_1 and H_2. The maximum value is $J(\max)=-0.60$ mJ/m^2 at 8 ML Cr. The magnitude of the coupling is in the same range as found by other researchers [1-3].

In conclusion, we have grown a high-quality epitaxial Fe/Cr sandwich structure on a Fe[100] whisker. This sample shows oscillations with a period of two Cr monolayers in the AF exchange coupling between Fe magnetizations separated by Cr. Though more detailed growth studies need to be carried out before quantitative comparison can be made with theory, it can now be definitively stated that any theory that does not predict 2-ML oscillations in ideal samples is missing one of the essential features of the effect.

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Note added.—We have received preprints describing other observations of the 2-ML oscillations in the coupling since submission of this article [9].

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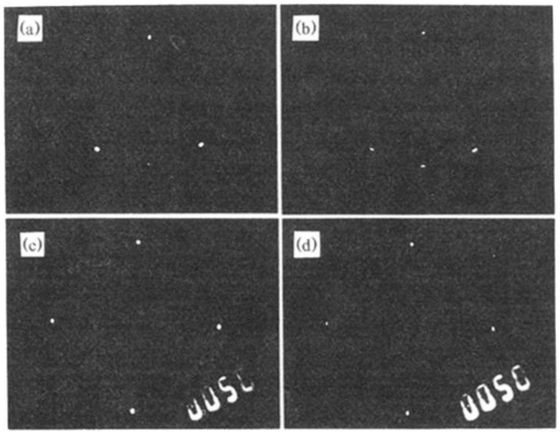

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