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Antiparallel coupling between Fe layers separated by a Cr interlayer: Dependence of the magnetization on the film thickness

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Spin-polarized low-energy electron diffraction is applied for the first time to study the coupling between ferromagnetic and antiferromagnetic ultrathin films. The remanent magnetization for the epitaxial double-layer structure $Fe_{II}/Cr/Fe_{I}/Cr(100)$ has been monitored *in situ* during growth. The two Fe slabs (Fe_{II}, Fe_{I}) separated by a thin Cr layer show in-plane antiparallel magnetic coupling along the external magnetic field direction when the two Fe-layer thicknesses differ. The measurements suggest a 90° rotation of the antiparallel magnetization for equal Fe-layer thicknesses.

The synthesis of new materials with novel properties has largely motivated current interest in superlattice and multilayer structures. Magnetic layered structure, in particular, might have wide application in the next generation of devices for magnetic recording. The production of well-characterized layered structures of magnetic materials may also provide tractable model systems to test theoretical predictions. Theoretical work[1,2] has recently been devoted especially to the magnetic properties of layered structures formed from ferromagnetic and antiferromagnetic materials. A rich variety of behavior is expected, depending on the microscopic details which determine the exchange coupling between the layers.

In this paper we report on the first application of spin-polarized low-energy electron diffraction (SPLEED) to the study of thin films and layered structures. It will be shown that by taking advantage of the surface sensitivity of this technique it has been possible to monitor the evolution of magnetic properties of ultrathin layered structures during growth, *in situ*, in real time, for incremental layer thickness as small as 0.06 monolayer (ML). The technique has been applied to the study of the remanent magnetization of Cr/Fe layered structures epitaxially grown on Cr(100). This particular system has recently attracted considerable attention since it has been found[3,4] by light scattering from spin waves that two Fe films, separated by a thin Cr layer, may order antiparallel, with their magnetizations perpendicular to a small external magnetic field.

By SPLEED we have monitored the remanent magnetization at the various stages of the multilayer structure growth: (a) $Fe_{I}/Cr(100)$ (single Fe film on Cr); (b) $Cr/Fe_{I}/Cr(100)$ (Fe sandwiched between Cr layers); (c) $Fe_{II}/Cr/Fe_{I}/Cr(100)$ (Fe double layer). In this paper we will concentrate the discussion on the results pertaining to the Fe double-layer system. A more detailed account of the data on the single Fe overlayer will be given elsewhere.[5]

Epitaxial growth has been obtained by a low rate (r) of evaporation (pressure $\leq 5\times10^{-10}$ torr for Fe at $r \simeq 0.1$ Å/min and $\leq 2\times10^{-10}$ torr for Cr at $r \simeq 1$ Å/min), on the substrate at 120 °C. The cleanliness has been checked by Auger spectroscopy. LEED shows sharp reflexes with $c(1\times1)$ symmetry of the epitaxial films. The growth mode was monitored by measuring the Auger signal of selected Fe and Cr transitions as a function of the thin-film deposition time. These plots show an exponential attenuation of the substrate signal with increasing Fe coverage. The slope of this curve shows weak kinks at thicknesses corresponding to one and two atomic monolayers, respectively. This result supports the layer-by-layer growth mode of Fe on Cr(001). The Auger results for Cr on the Fe film are less clear, suggesting either a Stranski-Krastanov growth mode or layer-by-layer growth with some Fe segration (0.4 ML).

The SPLEED measurements were performed *in situ* with the apparatus described in Refs. 6–8. The experimental setup allows measurement of the exchange asymmetry during the evaporation of the Fe and Cr thin layers. The (magnetic) exchange asymmetry A_{ex} of diffracted beams was measured in remanence, i.e., after removing the magnetizing field (60 Oe), applied along the [110] axis in the plane of the sample surface. The in-plane remanent magnetization along the [110] direction is related to the exchange asymmetry:

$$A_{ex} = \frac{I_{\uparrow\uparrow} - I_{\uparrow\downarrow}}{I_{\uparrow\downarrow} + I_{\uparrow\downarrow}} ,$$

where $I_{\uparrow\uparrow}$ ($I_{\uparrow\downarrow}$) is the diffracted intensity for incident electron polarization vector parallel (antiparallel) to the majority spin direction of the surface region probed. Note that for each point the value of A_{ex} is determined by reversing the magnetic field several times [cf. Ref. 7, Eqs. (5) and (6)]. The remanent magnetization of an Fe thin film (4 ML) covered by 1 ML of Cr was found to be the same for field in the range $30 \leq H \leq 80$ Oe. The kinetic energy of the incident electrons lies in the range $20 \leq E \leq 140$ eV, which corresponds to a probing depth $l \lesssim 5$ Å.

The exchange scattering asymmetry has been measured as a function of the primary-electron energy at various angles at selected film thickness. The exchange-asymmetry spectra of the (00) beam for Fe layers 2 to 10 ML thick deposited on Cr shows a trend towards increasing its absolute value with increasing Fe thickness. No other change in the exchange asymmetry versus primary-electron energy curves is observed for Fe films on Cr (no extra structures or shifted or distorted peaks). In order to carefully follow the onset and the development of the ferromagnetic ordering, exchange-asymmetry measurements at fixed scattering angle and fixed electron energy have been recorded *during* film growth (i.e., as a function of the incremental layer thickness). These curves, as suggested from the simple behavior of the exchange asymmetry reported above, can be directly related to the remanent magnetization of the surface of the sample along the external field direction. With an appropriate choice of the experimental parameters a sensitivity capable of detecting changes in the exchange asymmetry for incremental deposition below 0.1 ML has been achieved.

In Fig. 1 we show such a curve, collected at an electron energy of 73 eV for the $(2\bar{2})$ beam, during the sequence of growth of the layered structure Fe_{II}(18.7 ML)/Cr(10 ML)/Fe_{I}(10 ML)/Cr(100). A zero exchange asymmetry is observed, within the accuracy of the measurement (better than 0.5%), for the clean Cr(100) substrate. When the deposition of the first Fe layer (Fe_I) occurs the results clearly show no remanent magnetization along the external field direction for film thickness below 2 ML. Above this coverage a rather abrupt onset of remanent magnetization along the [110] direction occurs. At 3 ML the average value of the exchange asymmetry is close to 70% of the value for thick films. The exchange asymmetry then approaches the asymptotic value for film thickness increasing up to 10 ML. The deposition of Cr results in a decrease of the exchange asymmetry which vanishes for a Cr thickness comparable with the probing depth of the measurement. With 10 ML of Cr the asymmetry is zero.[5]

Surprisingly the growth of the second Fe layer (Fe_{II}) does not result in any measurable asymmetry up to ~ 1.1 ML thickness. This can be related to the interaction with the underlying Fe_I slab. Note that at low coverages, the sign of A_{ex} for the Fe_{II} is negative. The covergence to the asymptotic value of A_{ex} follows the same trend as for Fe_I. The slightly lower absolute value of A_{ex} for Fe_{II} with respect to Fe_I may be tentatively attributed to some roughening of the second Cr/Fe interface. When the Fe_{II} thickness approaches that of Fe_I, i.e., at $d(Fe_{II}) \simeq \frac{2}{3} d(Fe_I)$, the magnitude of the exchange asymmetry starts decreasing. When the Fe_{II} thickness is equal to that of Fe_I the exchange asymmetry is exactly zero. With increasing Fe_{II} thickness the exchange asymmetry reverses its sign again and it reaches its asymptotic value when $d(Fe_{II}) \simeq 2d(Fe_I)$.

These results give a clear indication that antiparallel coupling can be obtained between the two Fe layers. We recall that because of the surface sensitivity of the measurements the magnitude of the exchange asymmetry and its sign are essentially determined by the magnetization of the two topmost atomic layers. The sign reversal that occurs at low thickness for Fe_{II} indicates that the second Fe slab initially grows with its magnetization in opposite direction with respect to that of Fe_I and to the applied magnetic field. Such a configuration with antiparallel alignment of the magnetization has been recently predicted and discussed.[1] It should also be noted that a plausible and straightforward explanation of the results is the reduction of the demagnetization energy in the antiferromagnetic configuration. This interpretation seems to be favored by the zero exchange asymmetry observed for the Cr thin film grown on top of Fe_I. A nonzero asymmetry is expected for the case where the Cr film orders with alternating ferromagnetic (001) atomic sheets.

Another relevant feature of the curve in Fig. 1 consists of the decreasing exchange asymmetry when $d(Fe_I)$ approaches $d(Fe_{II})$. The data indicate that no remanent in-plane magnetization along the [110] direction of the outer layer of the sample is obtained after the removal of the magnetic field when the two Fe films have the same thickness. This result is consistent with recent findings by Grünberg *et al.*[3] that two equally thick Fe layers separat-

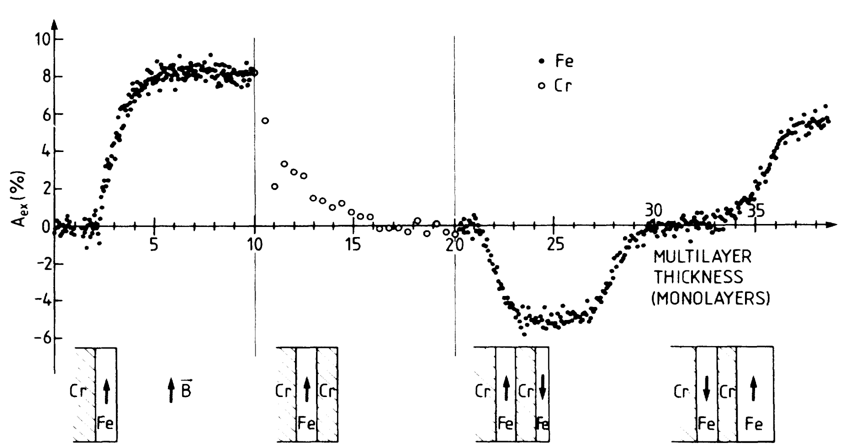

FIG. 1. Exchange asymmetry A_{ex} of the $(2\bar{2})$ beam vs incremental film thickness of the multilayer grown on Cr(001). A_{ex} reflects the remanent magnetization, along the (110) crystal axis, of a few topmost surface atomic layers of Fe (●) and Cr (○), respectively. The data were collected *during* growth of the multilayer. The angle of incidence of the polarized electron beam is $\theta = 29°$ with respect to the surface normal. The electron energy $E = 73$ eV.

ed by a thin Cr film order antiparallel with their magnetization perpendicular to the small external field. A 90° in-plane rotation of the direction of the remanent magnetization may account for the behavior of the exchange asymmetry when the Fe_{II} thickness approaches that of Fe_{I}. Also, analogously, again a 90° rotation in the same direction of the magnetization vector occurs when the thickness of Fe_{II} exceeds that of Fe_{I}, resulting finally in a magnetization along the [110] direction, with Fe_{II} now aligned with the magnetic field. In Fig. 1 we show schematically how the magnetization directions of the two layers changes with the increased thickness of Fe_{II}.

In summary, these results support and complement the recent results by Grünberg *et al.*[3] Also, our results suggest that various different static antiparallel configurations can be obtained in magnetic remanence by varying the relative thickness of the two coupled Fe layers. The stability of the antiparallel coupling perpendicular to the applied magnetic field appears, in magnetic remanence, to be confined to the region where the two Fe layers have the same thickness. Otherwise, when the thickness of two layer differs, the thicker layer aligns itself parallel to the direction of the magnetic field, and the thinner layer in the antiparallel direction.

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