Antiferromagnetic interlayer exchange coupling in sputtered Fe/Cr multilayers: Dependence on number of Fe layers

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(Received 5 November 1990; accepted for publication 6 February 1991)

The antiferromagnetic arrangement of the magnetic moments of Fe layers in sputtered Fe/Cr multilayered structures is directly demonstrated from polarized neutron reflectometry studies. Such an antiferromagnetic interlayer exchange coupling is also consistent with magnetization studies on a series of $[Fe/Cr]_N$ structures. A remanent magnetization is observed for structures containing an odd number of bilayers but no remanent moment is found for an even number of bilayers. By examining the dependence of saturation field on the number of bilayers it is shown that the antiferromagnetic coupling strength is independent of the number of bilayers and is the same for superlattice and sandwich structures.

There has been considerable interest recently in the properties of Fe/Cr multilayered structures. It was originally discovered in single-crystal Fe/Cr/Fe sandwiches that the ferromagnetic Fe layers are aligned antiparallel to one another in small magnetic fields.[1,2] Subsequently the same phenomenon of antiferromagnetic exchange coupling was found in single-crystal Fe/Cr superlattice structures.[3] In addition both the sandwich and superlattice structures display unusually large magnetoresistance effects.[3,4] Recently we showed that sputtered polycrystalline Fe/Cr multilayered films show surprisingly similar behavior to comparable single-crystal structures.[5] More importantly we discovered that both the exchange coupling and the saturation magnetoresistance oscillate as a function of Cr spacer layer thickness with a period of $\simeq 20$ Å in Fe/Cr as well as Co/Cr, Ni/Cr, and $Ni_{80}Fe_{20}$/Cr systems.[6] This means, in particular, that the exchange coupling depends strongly on the Cr layer thickness making it difficult to compare data on sandwich versus superlattice structures from different groups. In this letter we show, contrary to suggestions that the Fe layers in Fe/Cr multilayers are more strongly exchange coupled in superlattices than in sandwiches,[7,8] that the magnetic exchange coupling is independent of the number of Fe/Cr bilayers. We also directly confirm the existence of antiferromagnetic coupling in sputtered Fe/Cr superlattices from polarized neutron reflectometry studies.

The structures described in this letter were prepared by magnetron dc sputter deposition in a high vacuum system with a base pressure of $\simeq 5\times 10^{-9}$ T. The films were prepared at $\simeq 50$ °C at $\simeq 2$ Å/s in 3.25 mT argon.

Neutron reflection measurements were taken at the polarized neutron reflectometer (POSY I) at the Intense Pulsed Neutron Source at Argonne National Laboratory.[9] Polarized neutron reflectivity curves for neutrons polarized parallel or antiparallel to the magnetization of the sample were taken at a fixed angle of incidence for a range of neutron momenta. Since the experiment is limited to small values of momentum transfer, $q=4\pi \sin \theta/\lambda$, samples with relatively long superlattice periods were used. Figure 1(a) shows typical spin-dependent reflectivity curves for a sample with the structure Si(111)/Cr(100 Å)/[Fe(32 Å)/Cr(10 Å)]$_{20}$/Cr(10 Å). In this case a field of 4 kOe was applied in the plane of the sample. This field is large enough to induce a significant magnetization of the sample but not large enough to saturate the magnetization. Two broad diffraction peaks are visible. The peak at $q\simeq 0.14$ Å^{-1} corresponds to the first-order diffraction peak from the superlattice itself with a lattice spacing of 42 Å in good agreement with that expected. Note that the lattice spacing is not exactly given by $2\pi/q$ since refraction of the neutrons has to be taken into account.[9] The second peak at $q\simeq 0.08$ Å^{-1} corresponds to twice this spacing. Note that except for momenta in the region of this second peak the reflectivity is strongly spin dependent as expected for a partially magnetized sample. The observation of such a half-order peak has previously been found in a single-crystal Fe/Cr superlattice using unpolarized neutrons.[7] It is perhaps a surprising result that the degree of the antiferromagnetic coupling of the Fe layers in polycrystalline sputtered structures is similar to that found in high quality molecular beam epitaxy (MBE) grown single-crystal structures. This is an important finding since sputter deposition allow the rapid preparation of large numbers of films under very similar conditions, whereas MBE is typically a much slower and more laborious growth technique.

Spin-dependent reflectivity data were examined in detail as a function of in-plane magnetic field for a second sample. This sample had a structure of the form, GaAs(001)/Cr(100 Å)/[Fe(20 Å)/Cr(10 Å)]$_{20}$/Cr(100 Å). In an attempt to prepare very flat layers a special substrate comprising an MBE-grown epitaxial 0.6-μm-thick GaAs(001) layer on a GaAs(001) wafer capped *in situ* with a chemically passivating layer of As was used. The capping layer was removed in the sputtering system by heating the wafer in vacuum at 450 °C for 30 min. For this sample with a shorter superlattice period, the expected 30 Å diffraction peak is at too high a q to be observable.

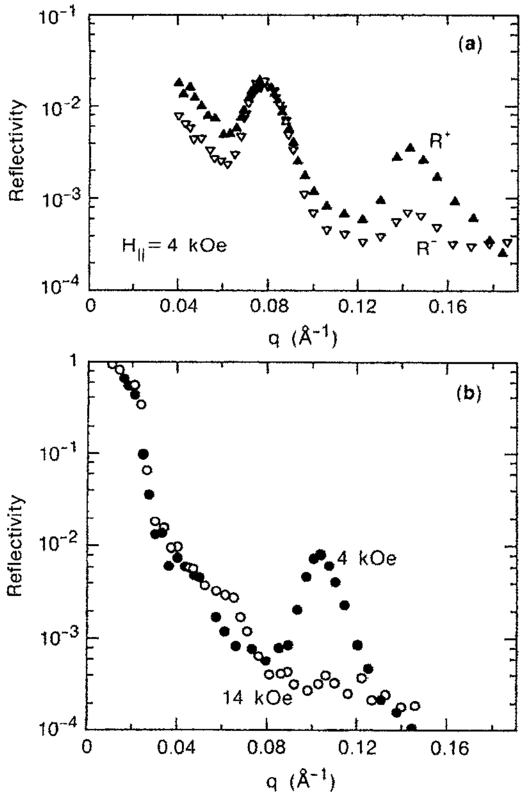

FIG. 1. (a) Spin-dependent reflectivity of a superlattice of the form, Si(111)/Cr(100 Å)/[Fe(32 Å)/Cr(10 Å)]$_{20}$/Cr(10 Å) measured at room temperature in a magnetic field of 4 kOe applied parallel to the plane of the sample. (b) Spin averaged reflectivity of a GaAs(001)/Cr(100 Å)/[Fe(20 Å)/Cr(10 Å)]$_{20}$/Cr(100 Å) superlattice for two different in-plane applied fields. At 14 kOe the sample is magnetically saturated.

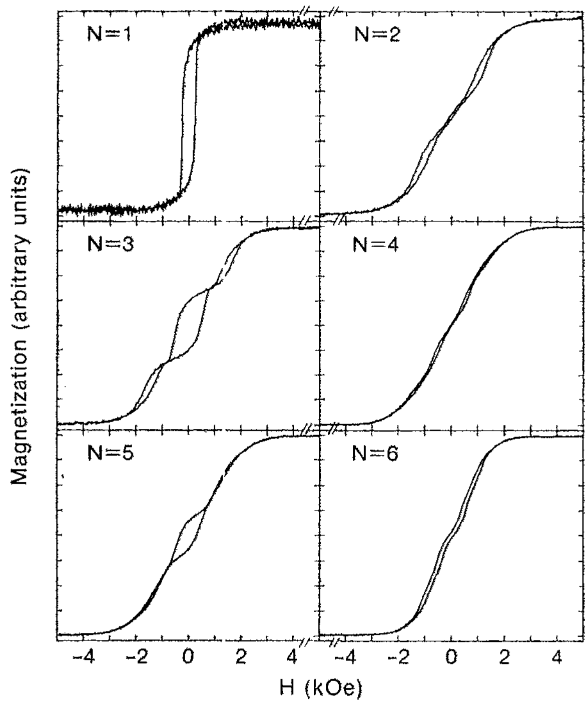

FIG. 2. Series of in-plane magnetic hysteresis loops at 300 K for six samples of the form Si(111)/Cr(115 Å)/[Fe(16 Å)/Cr(11.5 Å)]$_N$/Cr(115 Å) with varying number of bilayers, N, from 1 to 6.

However, at low magnetic fields, a broad diffraction peak is observed at $q \simeq 0.1$ Å^{-1}. As the field strength is increased the intensity of this peak decreases and for fields large enough to saturate the magnetization the peak is totally absent. Figure 1(b) shows representative data for fields of 4 and 14 kOe. These data prove that the peak at $q \simeq 0.1^{-1}$ originates from purely magnetic—rather than nuclear—scattering of the neutrons from the film. Moreover x-ray scattering experiments on the same sample confirm that the crystallographic superlattice has a spacing corresponding to exactly half that of the magnetic superlattice spacing (inferred from the position of the $q \simeq 0.1$ Å^{-1} peak). Thus in agreement with magnetization studies on these samples the magnetic spacing indicates a magnetic structure in which, at low fields, each ferromagnetic Fe layer is antiferromagnetically coupled with its immediate neighbors.

The orientation of the antiferromagnetic superlattice can be deduced from the polarization of the reflected neutron beam compared to that of the incident beam.[10] The magnetic moments of the neutrons are polarized in the direction of the applied field. If the magnetic moments in the sample are ordered along the same direction the polarization state of the neutron remains unaltered during the reflection process. If however, the moments in the sample are perpendicular to the applied magnetic field, the neutron spin precesses around such local magnetic fields and the reflected neutron will be "spin-flipped." Monitoring the polarization of the reflected neutrons with q close to 0.1 Å^{-1} at the antiferromagnetic peak at low magnetic fields shows that about 50% of the neutrons are spin-flipped. This is evidence for the presence of antiferromagnetic domains within the sample. The number of spin-flipped neutrons increases rapidly when the applied field is increased above approximately 1 kOe. This is consistent with a spin-flip transition for those portions of the polycrystalline sample with their easy axis aligned close to the applied field direction. The spin-flip transition has previously been observed in single-crystal Fe/Cr structures in light scattering[1] and ferromagnetic resonance (FMR)[8] studies. At fields just above $\simeq 1$ kOe the magnetic moments of all the Fe layers are arranged at almost 90° from the applied field. As the field is increased this angle decreases such that, for example, at 4 kOe this angle becomes $\simeq 45°$. At fields sufficiently large to completely saturate the magnetization of the sample the neutron beam depolarization attributed to the sample goes to zero.

Several series of [Fe/Cr]$_N$ multilayered structures were prepared with the number of bilayers, N, varying from 1 to 50. Typical data for six structures with bilayers varying from 1 to 6 are shown in Fig. 2. The magnetization of a single bilayer saturates in a small magnetic field and displays a considerable remanent moment in zero field. In contrast, structures with an even number of bilayers exhibit almost no remanent moment consistent with antiferromagnetic coupling of successive Fe layers. Perhaps the most interesting structures are those with an odd number of bilayers. For perfect antiferromagnetic alignment of neighboring Fe layers one would expect, in this case, a net

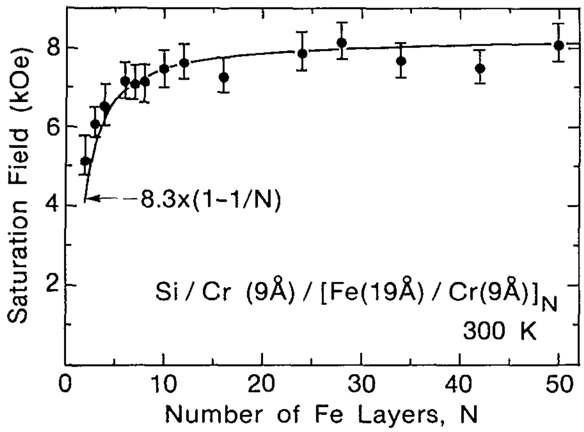

FIG. 3. Saturation field vs number of bilayers for a series of samples of the form, Si(100)/Cr(9 Å)/[Fe(19 Å)/Cr(9 Å)]$_N$. The saturation field is the field at which the magnetization has reached 85% of the saturation magnetization. The error bars represent fields within which the magnetization has reached within 82 and 88% of the saturation magnetization. The line is a fit to the data of the functional form shown in the figure.

moment equal to the moment of one of the Fe layers. This is indeed observed as is shown in Fig. 2 for the cases of three and five bilayers. Data are shown in Fig. 2 for a Cr layer thickness deliberately chosen to be just thicker than that at the first maximum in antiferromagnetic exchange coupling as a function of Cr layer thickness.[6] This illustrates the variation of the unpaired moment in the odd bilayer structures most beautifully but is not ideal for examining the dependence of coupling on N since the coupling is much more sensitive to variations in Cr layer spacing.

Data for a second series of structures in which the thickness of the Cr layer is chosen to be that for maximum antiferromagnetic exchange coupling[6] is shown in Fig. 3. The field required to saturate the magnetization of the samples is given as a function of N. The saturation field, H_S, is defined as the field at which the moment has attained 85% of the saturation moment. The figure clearly shows that H_S is decreased as N becomes small. Indeed for two bilayers the saturation field is almost exactly half that for 50 bilayers. In the simplest model for a superlattice, H_S is given by $4J/M_S d_F$, where M_S and d_F are the saturation magnetization and the thickness of the ferromagnetic layer respectively, and J is the interlayer exchange coupling.[7] For a structure containing N bilayers, H_S will be reduced from that for the superlattice by the factor $(1 - 1/N)$. A fit of this form shown in Fig. 3 describes the experimental data quite well. Thus we conclude that there is no significant difference in the magnitude of the exchange coupling in Fe/Cr superlattice structures compared to sandwich structures.

In summary we have demonstrated that sputtered polycrystalline Fe/Cr multilayered structures form macroscopic antiferromagnets for Cr layers $\simeq 10$ Å thick. The antiferromagnetic arrangement of the Fe layers is directly observed in polarized neutron reflection experiments and indirectly from magnetization studies. The field required to saturate the magnetization of a series of multilayered structures containing varying number of Fe/Cr bilayers is shown to vary as $(1 - 1/N)$. This is consistent with an interlayer magnetic coupling strength which is independent of N and is the same for sandwich and superlattice structures.

We thank K. P. Roche for technical support and D. Mauri for the use of his alternating gradient magnetometer. We thank P. D. Kirchner and R. F. Marks for providing the epitaxial GaAs films. The work at Argonne was supported by the U. S. Department of Energy, BES-Material Sciences, under contract W-31-109-Eng-38.

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