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**SPIN STRUCTURES OF Fe/Gd and Fe/Cr MULTILAYERS
DETERMINED BY POLARIZED NEUTRON REFLECTOMETRY**

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Spin Structures of Fe/Gd and Fe/Cr Multilayers
Determined by Polarized Neutron Reflectometry

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Abstract

Polarized neutron reflection was used to determine the magnetic structure of two different antiferromagnetically coupled multilayer systems, Fe/Gd and Fe/Cr. In Fe/Gd, the Fe and Gd moments are coupled antiparallel at the interface. At low temperatures a surface induced magnetic phase transition was found. In Fe/Cr, annealing at temperatures of up to 425°C, resulted in the degrading of antiferromagnetic coupling between Fe layers and in the formation of ferromagnetically coupled regions.

1 Introduction

The discovery of novel magnetic properties in artificial metallic multilayers has opened an entirely new field of investigations [1]-[3]. In these materials the individual layers, a few tens of Ångströms thick, are ferromagnetic or non-magnetic. The magnetic coupling between layers can be varied, giving rise to parallel (F) or antiparallel (AF) magnetic configurations, by changing layer thickness, chemistry, or magnetic field. Questions have arisen about the true parallelism (or antiparallelism) of the magnetization of different layers; the stability and the uniformity of the magnetic coordination; the correlation between magnetic and chemical structure. A useful tool in investigating such properties is polarized neutron diffraction (PNR). However the thickness of the layers is such that the first Bragg reflection occurs at very small angle, close to the incident beam and also close to the region of total reflection. We would like to give in this paper two examples of the application of of "stretched" polarized neutron reflectometry, or of diffraction at very small angle [4, 5].

The specular reflectivity is a function of wavevector transfer q perpendicular to the surface yielding patterns whose basic features can be described by a simple rule. Since q is a coordinate in reciprocal space, to proceed from the region of small q onward means to pass from a coarse-grained to a more detailed vision of the chemical and magnetic profile of the film. Unfortunately this can be done only at the cost of intensity, since the reflectivity decreases [6] as q^{-4} . When q reaches a value corresponding to the superlattice spacing the prominent feature of the reflectivity profile is the onset of the first Bragg diffraction peak.

The neutron measurements were taken with the polarized neutron reflectometer POSY I

at the Intense Pulsed Neutron Source of Argonne National Laboratory [7]. Neutrons of wavelengths ranging from 2.5 to 13.0 Å were polarized parallel or antiparallel to the magnetic field which was transverse to the beam and parallel to the sample's surface. The spin-dependent reflectivities (R^+ , R^-) were measured in a position-sensitive detector at an angle 2θ with the primary beam. Practically speaking, this instrument operates like a θ - 2θ diffractometer but with very high resolution ($dq = 0.0004 \text{ Å}^{-1}$) and limited range ($0.004 \text{ Å}^{-1} < q < 0.120 \text{ Å}^{-1}$).

2 Fe/Gd multilayers

Fe/Gd multilayers exhibit a strong *interface* antiferromagnetic coupling between Fe and Gd. The Fe and Gd layers themselves are simple ferromagnets with vastly different Curie temperatures and relatively weak anisotropy. For an idealized system of this kind Camley and co-workers [8, 9] suggested an interesting magnetic phase diagram. In this model the ferromagnetic Gd-Gd interaction is much weaker than the ferromagnetic Fe-Fe interaction or the antiferromagnetic one at the contact of the two materials. Thus, as the temperature is raised, the ordered moment of Gd (larger than Fe in the ground state) decreases faster than that of Fe. In the ground state the system is Gd-aligned. Raising the temperature the system becomes compensated, and then Fe-aligned. The onset of an external magnetic field creates additional phases. In weak magnetic fields the aligned state persists, and the magnetization remains constant. At a critical field a new structure, called “twisted”, appears [9]. In this structure only the Gd and Fe components perpendicular to the field are compensating each other.

The field-induced transition from the Gd-aligned to the twisted state is deeply per-

turbed at the outermost (surface and substrate) layers of the stacking. The Fe moments in the interior of the film are strongly held antiparallel to the external field by antiferromagnetic couplings to the adjacent Gd moments. In contrast, the Fe moments at the surface have Gd on one side only, and tend to turn in a magnetic field lower than the critical field for the bulk transition. If the multilayer is terminated with Gd, the opposite effect takes place: the twisting starts in the interior while twisting near the surface requires a much higher field because it is hindered by the presence of Gd at the surface. These qualitative arguments have been backed up by rigorous calculations [8]. The phase diagrams and the corresponding spin structures are shown schematically in fig. 1. Magnetization [10, 11] and Mössbauer [12] measurements on evaporated Gd/Fe multilayers indicated the presence of a phase transition similar to that predicted by Camley. However these techniques were not able to detect the twisted state at the surface. We therefore attempted to inspect it more directly by PNR.

The samples we investigated were obtained by alternately depositing Fe and Gd layers on 1" Si(111) wafers in a high-vacuum dc magnetron sputtering system. Each sample comprised a buffer layer of 56 Å Cr, followed by a total of 15 (Fe35Å/Gd50Å) bilayers and protected by another layer of 56 Å Cr to prevent oxidation of the more reactive Fe and Gd. Two samples were prepared: one with Fe facing the surface, and Gd the substrate the second with Gd facing the surface, and Fe the substrate. We present here the results obtained at 15 K, well below the compensation temperature of 130 K.

Fig. 2 shows the spin-dependent reflectivities of the sample with Fe at the surface when magnetic fields $H = 380$ Oe and 1 kOe are applied. The reflectivity curves on the left side are obtained with neutrons entering from the front side, and the curves on the right side with neutrons entering on the back side, through the silicon substrate. In low

fields the magnetic structure is basically that of the Gd-aligned state as can be deduced from the intensity ratio R^+/R^- at the Bragg reflection. Because Gd is an exceptional strong neutron absorber the measured reflectivity curves are surface sensitive. The spin dependence of the reflectivity of the front face (Fe outermost layer) changes with field much more dramatically than that of the back face (Gd outermost layer). The opposite is observed for the second, or complementary sample (not shown here). These observations give evidence that the field induced magnetic phase transition from a Gd-aligned to a twisted state starts at the Fe-terminated surface and is hindered at the Gd-terminated surface. This is in accordance with the predictions of Camley. To go further, and fully confirm the applicability of Camley's model a quantitative analysis of the results is needed. This and an extensive study of the spin-analyzed reflectivity are now in progress, and should enable us to verify not only the existence, but the detailed structure of the surface twisted state.

3 Fe/Cr Multilayers

This is the prototype and the most studied of all AF/F multilayers [1]-[3] coupled through intermediate layers. It is well known that in sputtered samples, the iron layers (typical thickness: 20 Å) are AF coupled across Cr layers up to 15 Å thick; then they are F coupled until Cr is 22 Å thick, and so on. Higher frequency AF/F oscillations appear [13] for epitaxially deposited samples. AF Fe/Cr multilayers show a giant magnetoresistance; its value is strongly dependent on the way the sample is prepared [2, 3]. We would like here to show the effect of annealing on the magnetoresistance and the concurrent changes in the magnetic configuration as revealed by PNR.

The sample preparation was similar to that described for Fe/Cd with a nominal layer thickness of 35 Å(Fe) and 10 Å(Cd). Identical prepared samples were annealed at temperatures ranging up to 425°C. Fig. 3a shows that annealing temperatures larger than 350°C dramatically reduce the magnetoresistance.

Fig. 4 shows the field-dependent diffraction patterns of a sample annealed at 350 °C. At 30 Oe the net magnetization is zero: the critical edge q_c is spin-independent and so is the first structural Bragg reflection, at $q = 0.16\text{\AA}^{-1}$. The magnetization of the sequential Fe layers is arranged as in the $+ - + -$ sequence, giving rise to the AF peak at $q = 0.08\text{\AA}^{-1}$, which is spin-independent. At 4 kOe a ferromagnetic component is present: this is made evident by the large spin dependence at q_c and at the Bragg peak. On the other hand, the intensity of the AF peak, which is always spin-independent, is severely diminished. Figs. 3b and 3c give the sublattice (AF) and net (F) magnetization for all samples as a function of the applied magnetic field as deduced from the reflectivity curves.

Let us try to interpret the results of fig. 3 in terms of a simple model. Assume that the AF component of the Fe moments lies in the film plane and perpendicular to the magnetic field: this is the spin flop state. Assume also that the F component is aligned with the applied magnetic field. The total moment per Fe atom remains approximately constant for fields larger than 1 kOe, and is similar to the moment of Fe in bulk iron. However for weaker magnetic fields this empirical rule does not hold entirely, and actually the AF contribution is seen to increase with field. In this region not all antiferromagnetic domains have spin-flopped, and the sublattice magnetization may point along the local easy direction. At these fields it is observed that the AF peak is broad and that the neutrons are partially reflected, partially scattered at 2θ angles above and below the reflected line, as if the AF domains were laterally limited [14]. Experimentally, the scattered radiation was

not fully intercepted by the position sensitive counter. The missing AF moment is indicated in fig. 3 as hatched (unannealed sample) and cross-hatched (350°C sample) areas. As the field is increased the system congeals in fewer AF domains (aligned perpendicular to the magnetic field) and the sharpened AF peak is fully accepted by the counter.

The presence of both F and AF components in a reflection spectrum can be interpreted in two ways. In an applied magnetic field, the magnetization of each layer can be canted with an AF component perpendicular to the applied field and a F component in field direction. In this picture the magnetic structure is uniform throughout the sample. In an alternate picture the sample is inhomogeneous, and composed of separate F and AF regions. Both cases give the same diffraction pattern. However, it can be noticed that while for the virgin sample F and AF components are present only in sizeable magnetic fields, for annealed samples a substantial ferromagnetic moment is present even in very weak fields. We can understand the development of F regions as resulting from the dissolution of the interfaces at annealing temperatures above 350°C. There are at least two possible ways to produce F coupling: (i) direct coupling from Fe bridges across the Cr layers and (ii) indirect coupling changing from AF to F for decreasing thickness of the dissolving Cr spacer layer.

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Figure Captions

Fig. 1 Expected phase diagram for a Gd/Fe multilayer and corresponding spin structures.

Effect of the surface on the phase diagram of Gd/Fe: if the surface layer is iron, a twisted state is induced at the surface in a field much lower than for the bulk. In contrast, Gd at the surface hinders the transition. Not shown are the disordered phase (high temperature) and the field-aligned phase (high magnetic field).

Fig. 2 Spin-dependent reflectivities of the $[\text{Fe}35\text{\AA}/\text{Gd}50\text{\AA}] \times 15$ sample with Fe at the surface and Gd facing the substrate measured at $T = 15$ K from front (pictures on the left) and backside (pictures on the right). Full circles: R^+ , open circles: R^- .

Fig. 3 Magnetoresistance, antiferromagnetic (AF) and ferromagnetic (F) moment components of the magnetization of sputtered $[\text{Fe}30\text{\AA}/\text{Cr}10\text{\AA}] \times 30$ multilayers, in the virgin and annealed states. Hatched and cross-hatched areas indicate missing AF moment of unannealed and 350°C sample, respectively.

Fig. 4 Spin-dependent reflectivities of a $[\text{Fe}30\text{\AA}/\text{Cr}10\text{\AA}] \times 30$ multilayer annealed at 350°C. Full circles: R^+ , open circles: R^- .

Fig: 1.

Fig 2

Fig 3.

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