Observation and interpretation of a partial Gd twisted spin state in an epitaxial Gd/Fe bilayer

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The interfacial coupling between Gd(0001) and Fe(110) in an epitaxial Gd/Fe bilayer has been studied by means of magnetometry and polarized neutron reflectivity with spin analysis. The results are interpreted in terms of a Gd twisted spin state in low applied fields. This original magnetic configuration was examined by theoretical calculations and found to be due to competing Gd-Gd and Gd-Fe exchange interactions with the Gd-Fe exchange interactions being long ranged and oscillatory. [S0163-1829(96)05121-1]

One of the most active areas of magnetism research over the past few years has been the study of exchange interactions between thin films. Most work has been devoted to the study of exchange coupling between magnetic slabs—either magnetic rare earth elements (R) or transition metal elements (T)—separated by nonmagnetic spacers.[1–7] Another particular aspect of magnetic interactions in thin film systems concerns exchange interactions between two magnetic layers of different nature, i.e., interfacial exchange coupling. This has attracted much less attention until now. In this Brief Report we present the results of a study of the interfacial exchange coupling in an R/T epitaxial bilayer (Gd/Fe). Although previous reports have appeared on the study of R/T bilayers and multilayers, no attempts have been made to deduce the range of the R/T exchange interactions. In addition, all previous reports have been devoted to polycrystalline systems.[8–11]

The films were grown by pulsed laser deposition. Initially a buffer layer of W(110) was deposited on Al_2O_3 (1120), as described elsewhere.[12] Fe(110) was subsequently deposited at 500 K at a rate of 0.5 Å min^{-1}. Gd was then deposited on the Fe at a deposition rate of 0.5 Å min^{-1} in a vacuum of 5×10^{-10} Torr. The first 5 Å of Gd was deposited with the substrate temperature held at 500 K; the remainder was deposited at ambient temperature. This approach enables the epitaxy to be initiated but prevents islanding. The epitaxial plane is (0001) and the epitaxial relationship for the Gd on the Fe was deduced from reflection high-energy electron diffraction to be the well known Nishiyama-Wassermann orientation.[13] The Gd was found to adopt its bulk lattice parameter ($a=3.63$ Å) from the very early stages of deposition (2 Å). The bilayer was protected by a deposition of Y with the substrate held at ambient temperature. The Y was found to epitaxy directly on the Gd. The exact thicknesses of the layers were determined by specular x-ray and neutron reflectivity and found to be Y=150±20 Å, Gd=63±5 Å, Fe=133±3 Å, and W=455±5 Å.

The field dependence of the total magnetic moment of the film was investigated by a vibrating sample magnetometer in fields up to 80 kOe and temperatures from 10 to 300 K. The in-plane low-field magnetization dependence along the [110] axes is shown in Figs. 1(a) and 1(b) for $T=10$ and 300 K, respectively. For fields greater than the coercive field (40 Oe) the approach to saturation is very slow. It is instructive to try and deduce the low field moment configuration of Fe and Gd from the above measurements. Consider, for example, the measurements along the [110] axis at 10 K. The value of the measured moment for $H=150$ Oe is 2.3×10^{-4} emu. If one assumes that the magnetization of Fe and Gd in the film at 10 K is the same as in the bulk (1740 emu cm^{-3} and 2010 emu cm^{-3}, respectively) then the total Fe moment in the film is 3.3×10^{-4} emu and the total Gd moment is 1.9×10^{-4} emu. The measured value corresponds neither to the sum nor the difference of the expected Gd and Fe moments thus indicating that the magnetic configuration is not simply collinear but more complex. The hypothesis of a complex configuration is borne out by the high field measurements as shown in Fig. 1(c) for H parallel to the [110] direction of Fe at 300 K. From fields of 1 to 20 kOe, the moment varies very slowly as the field is increased. At higher fields the susceptibility increases progressively. Complete saturation does not appear

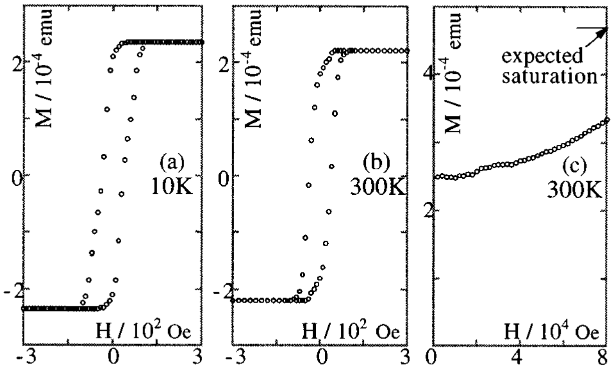

FIG. 1. Total moment measurements: (a) low field at 10 K, (b) low field at 300 K, and (c) high field at 300 K.

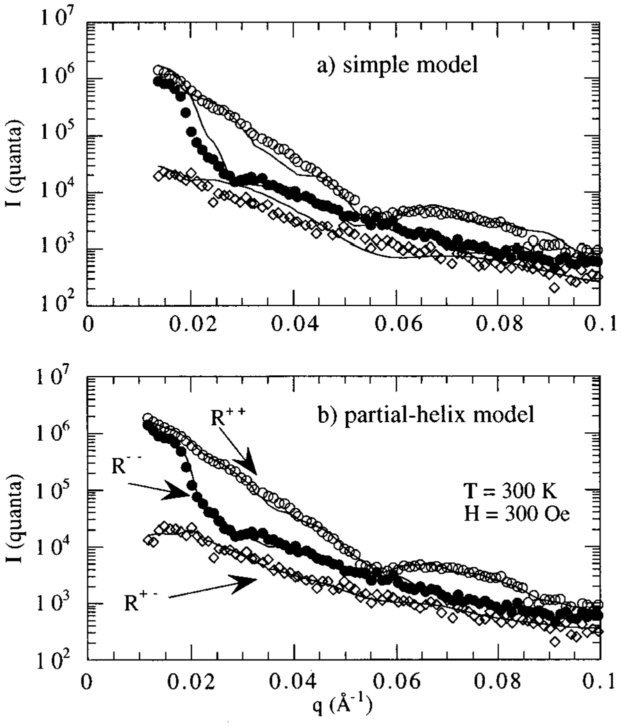

FIG. 2. Polarized neutron reflectivity measurements; experimental data are shown by data points and the best fits are shown by continuous lines.

to have occurred for fields even as high as 80 kOe.

In order to further analyze the moment configuration in the bilayer, polarized neutron reflectivity with spin analysis was performed on the G2-2 spectrometer at the Orphée reactor at Saclay, France. This spectrometer—a new facility of the Laboratoire Léon Brillouin—enables both non-spin-flip (R^{++},R^{--}) and spin-flip reflectivities (R^{+}) to be measured. During the measurements a field larger than 5 Oe is applied to ensure a good neutron polarization. For this work the neutron wavelength was 4.14 Å, the flipping ratio was 60, and the flux after analysis was 10^4 n/s^{-1} cm^{-2}. The three reflectivity profiles R^{++}, R^{--}, and R^{+-} were measured by varying the angle of incidence of the beam onto the sample at temperatures of 80, 300, and 350 K with an applied field of 300 Oe. The sample was oriented so that the in-plane Fe[110] axis (∥ to Gd[1100]) was parallel to the applied field and hence the quantization axis.

The reflectivity profiles obtained at 300 K are shown in Fig. 2. A large splitting between the $R^{++}(q)$ and $R^{--}(q)$ reflectivity profiles is observed which indicates the existence of a large component of the magnetization along the direction of the applied field. The most striking feature is however the presence of a distinctive spin-flip reflectivity (R^{+-}) which shows that the magnetization is not completely parallel nor antiparallel to the applied field.

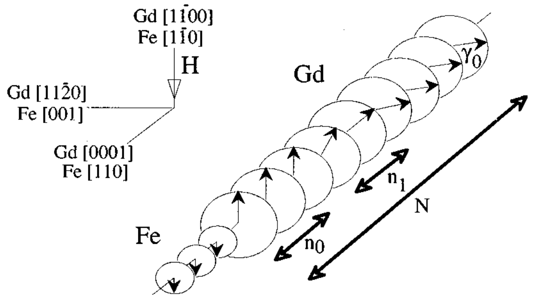

FIG. 3. Schematic representation of the moment configuration in the Gd/Fe bi-layer showing the Gd twisted spin state (see text and Table 1).

The moment configuration was *analyzed* by fitting the experimental reflectivities. The calculation procedure for the spin-flip and non-spin-flip reflectivity profiles is described in Ref. 14. Off-specular reflectivity was very small so the interface roughness was taken into account by introducing an alloy layer at each interface. The interface thicknesses then obtained are as follows: UHV/Y=20 Å; Y/Gd=14 Å; Gd/Fe=abrupt; Fe/W=10 Å. The same parameters were used to fit the data for different temperatures and different neutron spin directions. Initial attempts to fit the data to a simple model where all the Fe moments and all the Gd moments remain parallel amongst themselves with the Fe block and the Gd block canted at an angle to the applied field were unsuccessful [Fig. 2(a)]. The best fit was found for a more complex model [Fig. 2(b)] where the Fe moments stay parallel amongst themselves and where the Gd moments nearest the interface are antiparallel and then exhibit a *twisted state* further from the interface, as schematized in Fig. 3. The moments and their orientation are given in Table I. Each value, except the thickness, can be changed by ±10% and then an adjustment of the other values gives a reasonable fit. All densities were taken as bulk densities except for Fe where the average density is found to be 5% less than bulk Fe (it is minimum at the W/Fe interface and increases with the thickness).

The Gd moment decreases rapidly as the temperature increases. The mean Gd moment at 350 K is found to be 1.6 μ_B. At low temperature (70 K) the reflectivity profile is very similar to the 300 K data. It corresponds to a $2\mu_B$ moment for the Fe and a moment of about 6.5 μ_B for the Gd.

Although the observed magnetic configuration is not fully unambiguous due to the number of parameters involved in the fitting procedure, it must be stressed that it corresponds to the simplest model found to fit the experiment. In addition, this twisted Gd state can in fact be understood in the light of our understanding of magnetic interactions in bulk

TABLE I. Thickness profile of the magnetic moment and orientation for T=300 K and H=300 Oe from best fits to PNR data.

Element	Thickness (Å)	Moment (μ_B)	Orientation (angle with respect to H)
Fe	130	1.9	4.8
Gd	10	5.4	−176
Gd	10	5.2	−176
Gd	10	5.0	−158
Gd	10	4.8	−147
Gd	10	4.4	−131
Gd	10	4.0	−131
Y	14	1.0	−131
Y	158	0.0	

RT compounds.[15] The coupling between $3d$ and $4f$ spins is mediated by $5d$ electrons through $3d$–$5d$ band hybridization, which is stronger for the minority $3d$ band, and thus antiferromagnetic.[16,17] The configuration observed in the bilayer may be qualitatively understood by making the additional hypothesis that the Gd-Fe ($4f$-$3d$) interactions are long ranged and oscillatory which is reasonable as they are mediated by itinerant $5d$ electrons. Close to the Gd/Fe interface the Gd moments are antiparallel to the Fe moments as in bulk GdFe alloys. After approximately eight Gd planes from the interface the Gd-Fe interactions change sign and they then favor parallel coupling between the Fe and Gd moments. They are therefore in competition with the Gd-Gd interactions which tend to keep the Gd moments ferromagnetically coupled. The result of this competition is a progressive rotation of the Gd moments similar to that observed in an helimagnetic structure. After a further eight planes the Gd-Fe interactions become negligible and the rotation of the Gd moments stops.

These simple ideas were confirmed by calculations performed for Ruderman-Kittel-Kasuya-Yosida (RKKY) -type interactions. These were performed for the bilayer by assuming that all interactions (Fe-Fe, Gd-Fe, and Gd-Gd) are mediated by itinerant d electrons which are characterized by k_1 and k_2 which represent the Fermi momentum for d electrons in the Gd layer and the Fe layer, respectively. It is supposed that itinerant d electrons may travel in both the Fe and Gd films suffering partial reflection at the Gd-Fe interface due to the potential barrier and that they are ideally reflected at the outer interfaces of the bilayer. These itinerant d electrons interact via direct intra-atomic exchange interactions with electrons in localized Gd $4f$ shells and with d electrons in Fe. This gives rise to two kinds of oscillatory RKKY-type interactions—one between Gd magnetic moments and the other between Gd and Fe magnetic moments. It is assumed that the magnetic moments in Fe are ferromagnetically aligned due to the strong exchange coupling in metallic Fe. Let z be the coordinate perpendicular to the bilayer plane, then the expression for the generalized RKKY interaction $I(zz')$ between the total magnetic moments of the layers situated at positions z and z' is

$$\begin{aligned} I(zz') = \sum_{\kappa} \int dE \int dE' \, \frac{n(E)-n'(E')}{E-E'} \\ \times[G_{\kappa}^{+}(zz'E)-G_{\kappa}^{-}(zz'E)]J(z') \\ \times[G_{\kappa}^{+}(z'zE')-G_{\kappa}^{-}(z'zE')]J(z), \end{aligned} \tag{1}$$

where κ is the in-plane component of electron momentum, G_{κ}^{+} (retarded) and G_{κ}^{-} (advanced) are Green functions of itinerant d electrons, $J(z)$ is the intra-atomic exchange in Gd(J_0^{Gd}) or the d-d exchange in Fe (J_0^{Fe}), and $n(E)$ is the Fermi distribution function. For a free electron model these Green functions have to be a solution of the following:

$$\frac{\hbar^2}{2m}\left[k^2(z)+\frac{\partial^2}{\partial z^2}\right]G(zz') = \delta(z-z'), \tag{2}$$

where $k^2(z)={k_f}^2(z)-\kappa^2$, $k_f(z)=k_1$ if z is within the Gd film, $k_f(z)=k_2$ if z is within the Fe film, and $G(zz')=0$ for z and z' at the outer boundaries of the film. Solving Eq. (2) for Green functions and substituting its expression in (1) we may fulfill the integration in κ in an asymptotic limit and get the following expression for $I(zz')\equiv I(nn')$ for $z=a_0 n$, $z'=a_0 n'$, where n is an integer and a_0 is the interplanar distance of Gd along the [0001] direction:

$$I(nn') = J_0(n')+J_1(nn'), \tag{3}$$

$$J_0(n') = -J^{\mathrm{Gd}}J^{\mathrm{Fe}}\rho(E_f)\,\frac{\cos[2a_0k_1(n'-1)]k_2^2}{a_1k_2(k_1+k_2)^2(n'-1+\alpha_1)^2} - 2(J^{\mathrm{Gd}})^2\rho(E_f)\,\frac{k_1-k_2}{k_1+k_2}\left(\frac{\sin 2a_0k_1(n'-1)}{(n'-1+\alpha_2)^2}-\frac{\sin 2a_0Nk_1}{(N+\alpha_2)^2}\right), \tag{3a}$$

$$J_1(nn') = -\frac{(J^{\mathrm{Gd}})^2\rho(E_f)}{2}\left(\frac{1+(k_1-k_2)^2}{(k_1+k_2)^2}\right)\left(\frac{\sin 2a_0k_1(|n-n'|)}{(|n-n'|+\alpha_2)^2}-\frac{\sin[2a_0k_1\min(nn')]}{[\min(nn')+\alpha_1]^2}\right), \tag{3b}$$

where $J^{\mathrm{Gd}}=J_0^{\mathrm{Gd}}\mu^{\mathrm{Gd}}(T)/\mu^{\mathrm{Gd}}(0)$, $J^{\mathrm{Fe}}=J_0^{\mathrm{Fe}}\mu^{\mathrm{Fe}}(T)/\mu^{\mathrm{Fe}}(0)$, $\mu^{\mathrm{Gd}}(T)$ and $\mu^{\mathrm{Gd}}(T)$ are the magnetizations of Gd and Fe at temperature T, $\alpha_1=\sqrt[4]{(k_1^2-k_2^2)/k_2^2}/2a_0(k_1+k_2)$, $\alpha_2=1/\sqrt{\pi}k_1a_0$, $\rho(E_f)$ is the density of states in Gd, a_1 is the interplanar distance in Fe (=2 Å), and N is the total number of Gd atomic layers (=24). The first two terms in (3a) are usual RKKY Gd-Fe interactions which are integrated over the in-plane coordinates and over Gd-Gd interactions and the remaining terms are due to the reflections of itinerant electrons at the Gd-Fe interface and at the outer boundaries. If we suppose that $J_0^{\mathrm{Fe}}/J_0^{\mathrm{Gd}}=T_C^{\mathrm{Fe}}/T_C^{\mathrm{Gd}}=3$, then the leading term is the first term in (3a) and $J_0(n')\leqslant 0$ for $(n'-1)\leqslant \pi/4a_0k_1$ so the magnetization in several monolayers adjacent to the Gd-Fe interface has to be antiparallel to the Fe magnetization. For $(n'-1)>\pi/4a_0k_1$ the Gd magnetization may rotate away from the antiparallel configuration depending on the ratio of $J^{\mathrm{Fe}}/J^{\mathrm{Gd}}$. We therefore suppose that the angle θ_n between the direction of the magnetization in the nth Gd layer at position n with respect to the direction of the Fe magnetization is varying as

$$\begin{aligned} &\theta_n=\pi, && \text{for } 1\leqslant n\leqslant n_0, \\ &\theta_n=\pi-\frac{(\pi-\gamma_0)}{n_1}(n-n_0), && \text{for } n_0+1\leqslant n\leqslant n_0+n_1, \\ &\theta_n=\gamma_0, && \text{for } n_0+n_1+1\leqslant n\leqslant 24, \end{aligned} \tag{4}$$

where n_0 is the number of Gd layers where the magnetization of Gd is antiparallel to Fe, n_1 is the number of layers over which the magnetization rotates, and γ_0 is the angle between Gd and Fe in the $24-n_0-n_1$ remaining layers (Fig. 3). The total energy, F, of the Gd subsystem may be written as

$$F=-\sum_{1}^{24} J_0(n)\cos\theta_n-\sum_{n'\neq n}^{24} J_1(nn')\cos(\theta_n-\theta_{n'}). \quad (5)$$

The value of F has to be minimized with respect to the n_0, n_1, and γ_0 variables for given values of k_1, k_2, and $m=J^{\mathrm{Fe}}/J^{\mathrm{Gd}}$. We have done this numerically for several sets of values of k_1, k_2, and m and found that for values of $k_1=0.07$ Å^{-1}, $k_2=0.7$ Å^{-1}, and $m=3$, the obtained values of n_0, n_1, and γ_0 are 4, 8, and $\pi/2$, respectively, in semiquantitative agreement with experiment (8, 12, and $2\pi/3$, respectively). The value of $n_0\approx\pi/4a_0k_1$ and the values of n_1 and γ_0 depend essentially on the value of m. For example, for $m=10$ then $n_1=1$ and $\gamma_0=0$, and for $m\rightarrow 0$ then $\gamma_0=\pi$. These results are clear from a physical point of view: if Gd-Fe exchange is much larger than ferromagnetic Gd-Gd exchange, then the direction of the Gd magnetization is governed by the sign of the Gd-Fe exchange; on the other hand, if it is weak then the Gd stays ferromagnetic. In order to check the model, experiments at higher temperature should be made: m is expected to increase with temperature due to the faster decrease of μ^{Gd} compared with μ^{Fe}. Close to T_c, the twisted state should be replaced by a new state in which the Gd magnetization is either parallel or antiparallel to the Fe one, making a kind of antiphase arrangement.

In summary we have made experimental investigation of the moment configuration in a rare earth−transition metal epitaxial bilayer and found the Gd moments not to be collinear in zero or low applied fields. We have shown that the experimental results are consistent with a Gd twisted state in the Gd layer. We have shown that the experimental results are consistent with a Gd twisted state in the Gd layer. We have proposed an interpretation for its origin, which shows, in particular, that the interfacial Gd-Fe exchange interactions are long ranged and oscillatory. Finally, it should be noted that such a Gd twisted state is fundamentally different from the so-called twisted phases which have been observed in Fe/Gd polycrystalline multilayers.[11,18] The propagation vector which characterizes rotation of the moments in rare-earth metals is always parallel to the c axis (cf. the helimagnetic structures observed in several rare-earth metals). In polycrystalline multilayers, the direction which is perpendicular to the sample surface may correspond to various crystallographic directions. In epitaxial films, it is strictly the c direction which is perpendicular to the surface. This explains that the magnetic state described in the present paper is unique to epitaxial films.

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