Polarized neutron scattering from Gd/Fe multilayers: Twisted phase and spin-flip scattering

C. Dufour, K. Cherifi, G. Marchal, and Ph. Mangin
Laboratoire de Métallurgie Physique et Sciences des Matériaux, Université de Nancy 1, Boîte Postale 239, F-54506 Vandoeuvre CEDEX, France

M. Hennion
Laboratoire Léon Brillouin, Centre d'Etudes Nucléaires de Saclay, F-91191 Gif-sur-Yvette CEDEX, France
(Received 23 November 1992)

Low-angle polarized neutron scattering demonstrates the occurrence of aligned and twisted phases in Gd/Fe multilayers. In the aligned states, there is no spin-flip scattering. The large spin-flip scattering present at the compensation temperature shows the coherent transverse magnetic components of the twisted phase.

Because of the weak magnetocrystalline anisotropy of gadolinium, the Gd/Fe multilayered system plays a special role among the rare-earth iron multilayers. At a first attempt, the magnetic structure can be considered as the result of a competition between the strong ferromagnetic coupling between iron magnetic moments ($T_c = 1043$ K), the weak ferromagnetic coupling between gadolinium moments ($T_c = 293$ K) and the antiferromagnetic coupling between Fe and Gd magnetic moments at the interface. Calculations performed by Camley and Tilley[1,2] showed that three magnetic phases could take place: aligned Gd, aligned Fe, and twisted (Fig. 1).

The aligned-Gd phase takes place when the magnetization of gadolinium layers is larger than the magnetization of iron layers. The gadolinium moments are simply oriented in the field direction and the iron moments are antiparallel to this direction. In this phase, as in the following ones, the magnetic moments of both elements have been supposed to lie in the plane of the film as expected from demagnetization field effect. The aligned-Fe phase occurs when the magnetization of iron layers is larger than that of gadolinium ones. The iron moments are oriented in the field direction and the gadolinium moments are antiparallel. As the magnetization of gadolinium decreases more rapidly with temperature than that of iron (its Curie temperature is lower), samples in which gadolinium magnetization dominates at low temperature exhibit a compensation temperature above which iron magnetization dominates. This temperature separates at (or near) zero field the two aligned magnetic structures (see Fig. 2).

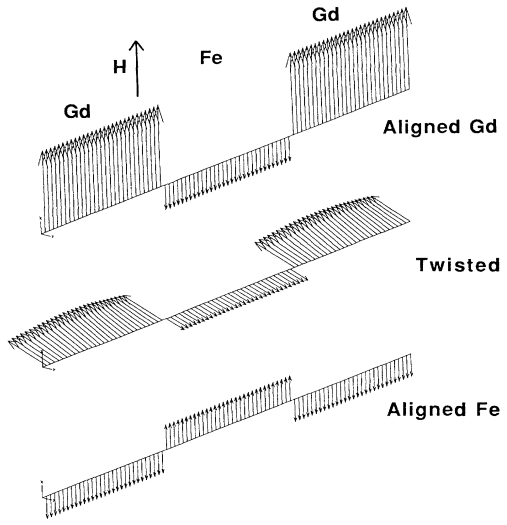

FIG. 1. Sketch of the aligned-Gd, aligned-Fe, and twisted phases.

In fact, the aligned states are unstable against a strong enough magnetic field. At a critical field H^*, the magnetic moments leave the aligned states and become rather perpendicular to the field: a twisted state with a canting of the spins inside the layers occurs. The strength of the field H^* is the result of a balance between the different contributions to the energy. From an energetical point of view, the aligned-twisted state transition can be described by two virtual stages: (i) The system undertakes a simple spin flop with both magnetizations antiparallel to one another and perpendicular to the field (perpendicular state). This stage costs an energy proportional to the field and to the difference between the magnetization of the

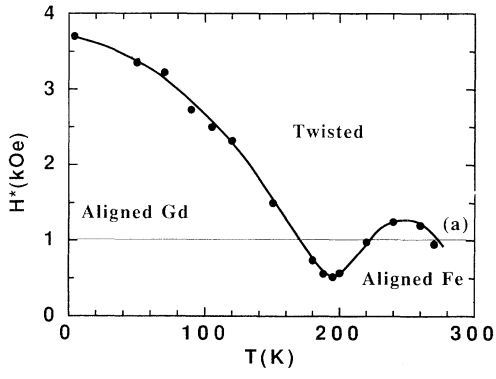

FIG. 2. Magnetic phase diagram of a Gd(84 Å)/Fe(42 Å) sample. The line separating the aligned from twisted state has been deduced from magnetization measurements (Ref. 3).

TABLE I. Expression of the total scattering amplitude densities of iron and gadolinium δ_{Fe} and δ_{Gd}, for the three scattering process $++$, $--$, and $+-$, and in the three magnetic states: aligned Gd, aligned Fe, and ideally perpendicular states.

		Aligned Gd	Perpendicular	Aligned Fe
$++$	δ_{Fe}	$\lambda^2 N_{Fe}(b_{Fe}-p_{Fe})/2\pi$	$\lambda^2 N_{Fe} b_{Fe}/2\pi$	$\lambda^2 N_{Fe}(b_{Fe}+p_{Fe})/2\pi$
	δ_{Gd}	$\lambda^2 N_{Gd}(b_{Gd}+p_{Gd})/2\pi$	$\lambda^2 N_{Gd} b_{Gd}/2\pi$	$\lambda^2 N_{Gd}(b_{Gd}-p_{Gd})/2\pi$
$--$	δ_{Fe}	$\lambda^2 N_{Fe}(b_{Fe}+p_{Fe})/2\pi$	$\lambda^2 N_{Fe} b_{Fe}/2\pi$	$\lambda^2 N_{Fe}(b_{Fe}-p_{Fe})/2\pi$
	δ_{Gd}	$\lambda^2 N_{Gd}(b_{Gd}-p_{Gd})/2\pi$	$\lambda^2 N_{Gd} b_{Gd}/2\pi$	$\lambda^2 N_{Gd}(b_{Gd}+p_{Gd})/2\pi$
$+-$	δ_{Fe}	0	$\lambda^2 N_{Fe} p_{Fe}/2\pi$	0
	δ_{Gd}	0	$\lambda^2 N_{Gd} p_{Gd}/2\pi$	0

two subnets. (ii) The system leaves the strictly perpendicular state with, inside each layer, a rotation of the spins (as in a domain wall) towards the field direction. At the interface the Fe and Gd spins are blocked antiparallel to one another and they are nearly perpendicular to the field because of the antiferromagnetic exchange. The energy gain of stage (ii) is larger than the energy loss of stage (i) if the difference between the magnetization is weak and if the field is strong. This explains why the aligned-twisted transition takes place at higher field when the temperature is shifted from the compensation temperature. At this temperature, step (i) does not cost any energy and at low field the twisted state is expected to be closer to the simple spin-flop state of the classical antiferromagnets.

The results of the calculations are consistent with magnetization measurements,[3-5] which show the occurrence of a plateaulike evolution in aligned states, and Mössbauer spectroscopy data[6] which provide the average angle between the orientation of the iron magnetic moments and the field direction. In this paper we present, from polarized neutron scattering, a strong experimental evidence for the aligned and twisted states as well as of the coherence of these magnetic structures over large distances. The data are very demonstrative and are interpreted in a direct way.

Let us at first recall the origin of the neutron intensities diffracted from a multilayer system when multiple reflections are neglected. Four scattering processes can be considered as independent: I^{++}, incident neutrons with the spin up are scattered with spin up; I^{--}, incident neutrons with spin down are scattered with spin down; I^{+-} and I^{-+}, incident neutrons are scattered with flipping of the spin.

According to the simple kinematic theory of scattering by multilayers, the different intensities are proportional to the squares of the differences between the total scattering amplitude densities of iron layers, δ_{Fe}, and of gadolinium layers, δ_{Gd}:

$$I \propto (\delta_{Fe}-\delta_{Gd})^2 .$$

When the atoms in the layers carry a moment μ, lying in the reflection plane, making an angle α with the quantification axis, the total scattering amplitudes densities for the $++$, $--$, and $+-$ processes are, respectively, written as $\delta^{++}=\lambda^2 N(b+p\cos\alpha)/2\pi$, $\delta^{--}=\lambda^2 N(b-p\cos\alpha)/2\pi$, and $\delta^{+-}=\lambda^2 Np\sin\alpha/2\pi$. N is the atomic density, b is the (complex) atomic nuclear scattering amplitude, and p is the magnetic scattering amplitude. p is known to be equal to 0.27μ, where μ is the magnetic moment (in μ_B) carried by the atom. So, for example, in the aligned-Gd state, $\alpha_{Gd}=0$, $\delta_{Gd}^{++}=\lambda^2 N_{Gd}(b_{Gd}+p_{Gd})/2\pi$, and $\alpha_{Fe}=180°$, $\delta_{Fe}^{++}=\lambda^2 N_{Fe}(b_{Fe}-p_{Fe})/2\pi$; $I^{++}\propto(\delta_{Fe}^{++}-\delta_{Gd}^{++})^2$.

The scattering amplitude densities of iron and gadolinium are reported in Table I for some specific aligned and perpendicular states. Numerical values of the intensities (in arbitrary units and normalized to $I^{--}=1$) calculated from Table I are reported in Table II. The calculations were performed by using $b_{Fe}=0.95\times10^{-12}$ cm and $b_{Gd}=(0.3+1.1i)10^{-12}$ cm.[7] The large imaginary part of gadolinium is due to the fact that the energy of neutron is close to a resonance near which the real part of b varies very rapidly.

The important point is that the non-spin-flip scattering

TABLE II. Calculated (calc.) and experimental (expt.) scattered intensites (I^{++}, I^{--}, and I^{+-}) from the aligned-Gd state at 10 K, the aligned-Fe state at 300 K, and the perpendicular twisted state at 185 K. The intensities are normalized to $I^{--}=1$ at 10 K.

Temperature		10 K	185 K	300 K
Magnetic moments (μ_B/atom)		$\mu_{Fe}=2.22$	$\mu_{Fe}=2.19$	$\mu_{Fe}=2.18$
		$\mu_{Gd}=7.55$	$\mu_{Gd}=5.3$	$\mu_{Gd}=0$
Magnetic state		Aligned Gd	Perpendicular	Aligned Fe
I^{++}	calc.	0.08	0.18	0.46
(arb. unit)	expt.	0.11	0.25	0.55
I^{--}	calc.	1	0.18	0.045
(arb. unit)	expt.	1	0.25	0.11
I^{+-}	calc.	0	0.25	0
(arb. unit)	expt.	0.02	0.14[a]	0.015

[a]This value is the experimental maximum of I^{+-} which occurs at a temperature of 180 K and is slightly shifted from the temperature at which $I^{++}=I^{--}$.

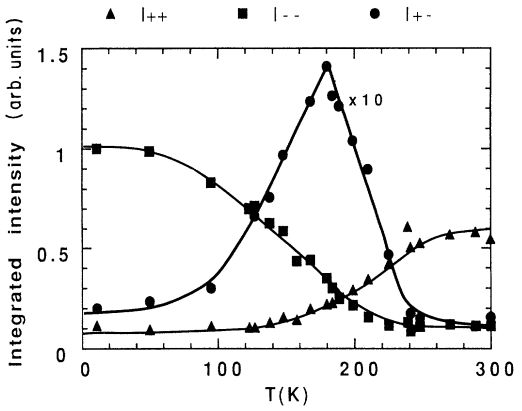

FIG. 3. Thermal variation of the low-angle first "Bragg peak" intensities I^{++}, I^{--}, and I^{+-} of the Gd(84 Å)/Fe(42 Å) multilayer. The field was kept constant and equal to 1 kOe (horizontal line of Fig. 2). The intensities are normalized to $I^{--}=1$ at 10 K.

processes are related to the nuclear scattering and to the component of the magnetization along the quantification axis (here the field direction) and that the spin-flip scattering is only due to the component of the magnetization, perpendicular to this direction.

The experimental measurements were carried out on the 4*F*1 triple-axis instrument of the "Laboratoire Léon Brillouin" in Saclay, France. The instrument is installed on the cold source and the incident wavelength was 4.48 Å. To obtain a resolution good enough to separate the peaks located at $q=0.05$ Å^{-1} from the incident beam, the instrument was set in triple-axis configuration with the analyzer in elastic position. Very thin slots collimated the incident and scattered beams. The neutrons were polarized and analyzed by two benders. A "Mezei-type" flipper was placed before the sample and a cryoflipper was set beyond the sample. The flipping ratios were about 20 in each configuration. The studied sample was similar to that whose magnetization measurements and Mössbauer spectroscopy results have been already presented.[3,6] The phase diagram deduced from magnetization is that of Fig. 2. The measurements were performed from 10 K to room temperature and with a 1 kOe field applied in the plane of the layer: we followed the line (*a*) of Fig. 2, we started in the aligned-Gd state, crossed the twisted, and entered the aligned-Fe state.

The integrated intensities, obtained from a simple Gaussian fit of the experimental peaks, are pictured in Fig. 3. The values collected at 10, 185, and 300 K are reported in Table II. The experimental results are qualitatively and, to a certain extent, quantitatively in agreement with the values calculated from ideal configurations (Table II).

At low temperatures, experimentally as well as from aligned-Gd calculations, I^{--} is larger than I^{++} by a factor 10; I^{+-}, which was expected to be zero in the purely aligned state, is found to be 0.02, which is, in fact, the lowest value which can be measured experimentally with a 20 flipping ratio.

At room temperature, the situation is reversed: iron is in the field direction, gadolinium is in the opposite one, and I^{++} is larger than I^{--} and I^{+-} is zero. As expected from the loss of gadolinium magnetization (I^{++}) 300 K is only half of (I^{--}) 10 K. Finally, the key point is that I^{+-} exhibits a peak at the compensation temperature, which means that a coherent magnetic state, with strong perpendicular components of the magnetizations takes place.

The experimental results indicate, however, that at its maximum, I^{+-} is 40% smaller than expected from a strictly perpendicular state. It proves the alignment is not perfect, as expected in the twisted phase. This is consistent with the fact that there is a temperature shift between the temperature at which I^{+-} is maximum and that for which $I^{++}=I^{--}$.

The results are very promising and bring about a proof of the model developed by Camley *et al.* and of the coherency of the magnetic structure. A complete analysis of the data, following the schema given in Refs. 8 and 9, taking into account the absorption of gadolinium and the multiple reflections has been undertaken. Starting from the microscopic model, a theoretical spin configuration is obtained. From this configuration, magnetization curves, Mössbauer spectroscopy spectra, and neutron-scattering intensities are deduced and compared to experimental data. The parameters of the model are adjusted to fit the three complementary informations. Note that some surface effect can take place: as shown theoretically by Le Page and Camley[10] and experimentally by Loewenhaupt *et al.*,[11] the last layer plays a role in the magnetic structure of some layers close to the surface and can lead to a twisted surface state.

However, beyond such refinement, the direct neutron data prove definitively the coherence of the magnetic phases over large distances, something neither the magnetization measurements not the Mössbauer spectroscopy could demonstrate.

Laboratoire de Métallurgie Physique et Sciences des Matériaux is Unité de Recherche Associée au CNRS No. 155.

[1]R. E. Camley and D. R. Tilley, Phys. Rev. B **37**, 3413 (1988).
[2]R. E. Camley, Phys. Rev. B **39**, 12 316 (1989).
[3]K. Cherifi, C. Dufour, Ph. Bauer, G. Marchal, and Ph. Mangin, Phys. Rev. B **44**, 7733 (1991).
[4]T. Morishita, Y. Togami, and K. T. Tsushima, J. Phys. Soc. Jpn. **54**, 37 (1985).
[5]H. Fujimori, Y. Kamiguchi, and Y. Hayakawa, J. Appl. Phys. **67**, 5716 (1990).
[6]Ph. Bauer, M. Sajieddine, C. Dufour, K. Cherifi, G. Marchal, and Ph. Mangin, Europhys. Lett. **16**, 307 (1991).

[7]C. F. Majkzak, J. Kwo, M. Hong, Y. Yafet, D. Gibbs, C. L. Chien, and J. Bohr, Adv. Phys. **40**, 99 (1991).
[8]G. P. Felcher, R. O. Hilleke, R. K. Crawford, J. Haumann, R. Kleb, and G. Ostrowski, Rev. Sci. Instrum. **58**, 609 (1987).
[9]S. J. Blundell and J. A. C. Bland, Phys. Rev. B **46**, 3391 (1992).
[10]J. G. Le Page and R. E. Camley, Phys. Rev. Lett. **65**, 1152 (1990).
[11]M. Loewenhaupt, W. Hahn, Y. Y. Huang, G. P. Felcher, and S. S. P. Parkin, J. Magn. Magn. Mater (to be published).