Experimental determination of the magnetic phase diagram of Gd/Fe multilayers

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By combining magnetization, Kerr effect measurements, and polarized neutron reflectometry we have determined the magnetic state of [Fe 35 Å/Gd 50 Å]×15 multilayers at different points of the H-T phase diagram. We confirm the predictions of Camley and co-workers for an idealized magnetic superlattice with antiferromagnetic interfacial coupling. The magnetic moments of the Fe and Gd layers are opposite with those of Gd becoming larger below a compensating temperature. The magnetic moments are aligned parallel to the applied magnetic field below a certain value, and then they take a twisted arrangement as the field is increased. The transition may start from the surface or the interior, depending on whether the magnetization of the surface layer is oriented in or opposite to the field direction.

I. INTRODUCTION

The controlled preparation of artificial metallic multilayers has opened an entirely new field of investigations which led, e.g., to the discovery of antiferromagnetic coupling of ferromagnetic Fe films through thin Cr spacer layers,[1] the coherent transmission of the spiral magnetic state of Dy through Y layers[2] or, quite general, the oscillatory coupling for a large class of metallic multilayers comprised of alternating thin ferromagnetic and nonferromagnetic spacer layers.[3] Camley and co-workers[4–8] investigated theoretically the magnetic phases of superlattices made of two kinds of ferromagnetic layers, in contact and with antiferromagnetic coupling at the interface. Dealing with a more complex system, Camley had to introduce some simplifying assumptions. In contrast with the superlattices of the Fe/Cr kind the interlayer interaction here is assumed to take place between the *interfacial* atoms. Specifying further the model, Camley and co-workers described what can be called an idealized Gd/Fe multilayered system. Gd and Fe layers themselves are simple ferromagnets with vastly different Curie temperatures (293 and 1024 K, respectively) and relatively weak anisotropy. The ferromagnetic interaction per atom is much weaker in the Gd layers than in the Fe layers or the antiferromagnetic interaction at the (atomic) contact between the two materials. The model makes the assumption that all spins within an atomic layer lie in the plane of the layer and all point into the same direction. Only the Zeeman interaction with the external field and an effective exchange interaction between spins in different atomic layers is taken into account.

Camley's model predicts three different phases in the H-T plane for an idealized Gd/Fe superlattice: the Gd-aligned phase where the Gd spins are aligned with the applied field and the Fe spins are antiparallel to the field; the Fe-aligned phase where the Fe spins are aligned with the field and the Gd spins are oppositely oriented; the twisted phase where the spins in each atomic layer make a different angle with respect to the applied field. A calculated phase diagram and a sketch of the magnetic structures in the different magnetic phases is shown in Fig. 1. In low magnetic fields we expect the Gd-aligned phase for $T<T_{\mathrm{comp}}$ and the Fe-aligned phase for $T_{\mathrm{comp}}<T<T_{C,\mathrm{Fe}}$. At T_{comp} the total Gd magnetization is exactly compensating the total Fe magnetization yielding an antiferromagnetic state. The compensation occurs because the ordered moment of Gd is larger than that of Fe in the ground state but decreases faster with temperature than the Fe-ordered moment. At a (temperature dependent) critical field H^* the twisted phase appears. In this structure only the Gd and Fe components perpendicular to the field are compensating each other.[4] Increasing further the magnetic field causes the moments of both gadolinium and iron to turn in the field direction.

The field-induced transition from the Fe- or Gd-aligned phase to the twisted phase is considerably modified at the outermost (surface and substrate) layers of the stacking. For instance, this is the surface effect predicted for a sample in the Gd-aligned state below T_{comp}. If the multilayer is terminated with Fe, the Fe moments in the interior of the multilayer are strongly held antiparallel to the external field by antiferromagnetic couplings to the adjacent Gd moments. In contrast, the Fe moments in the outermost layer have Gd on one side only, and tend to turn in a magnetic field lower than the critical field H^* 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.[5] A similar surface effect has been reported for Fe/Cr multilayers with an antiferromagnetic coupling of adjacent Fe layers and an even number of Fe layers.[9]

It might seem surprising to see how many of the predictions for such an idealized Fe/Gd superlattice have been partially experimentally verified on real Fe/Gd multilayers

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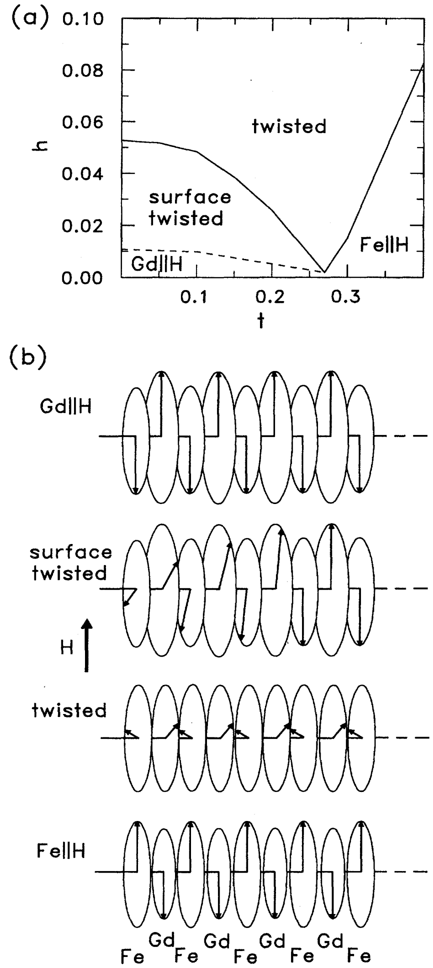

FIG. 1. (a) Calculated h-t phase diagram of a superlattice with a unit cell of 13 Fe atomic layers and 5 Gd atomic layers with the Fe film on the outside. The external field is given in dimensionless units by $h=H/JS_{Fe}$. $t=T/T_C$ is the reduced temperature where T_C refers to the Curie temperature of bulk Fe (from Ref. 5). (b) Sketch of the spin structures in the different magnetic phases; the arrows symbolize the average magnetization of one Fe or Gd layer.

as reported in the literature. The presence of a compensating temperature T_{comp} has been confirmed by magnetization measurements.[10–12] These have also inferred the presence of a transition from an Fe- or Gd-aligned phase into the twisted phase. Mössbauer effect measurements[13,14] as well as polarized neutrons[15] have shown that, in finite magnetic fields, some of the components of the sublattice magnetizations are indeed perpendicular to the applied magnetic field. It is the purpose of this paper to not only confirm the mentioned findings but also to show evidence that indeed the magnetic field-induced phase transformation originates at the surface or alternatively in the bulk depending upon the magnetization direction of the surface layer.

II. EXPERIMENT

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 [Fe 35 Å/Gd 50 Å] 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, denoted by [Fe/Gd], the second with Gd facing the surface, and Fe the substrate, denoted by [Gd/Fe].

The neutron measurements were taken with the polarized neutron reflectometer POSY I at the Intense Pulsed Neutron Source of Argonne National Laboratory.[16] 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, as a function of the neutron wavelength, in a position-sensitive detector at an angle 2θ with the primary beam. The neutron wavelengths were sorted out by the time of flight from the pulsed source to the detector. The reflectivities were determined after normalizing the reflected intensities to the incident spectrum. In this way the reflectivities R_+, R_- were measured as a function of $Q_z=4\pi\sin(\theta)/\lambda$, the component of the neutron momentum transfer perpendicular to the surface. The symbols $+$ and $-$ refer to the polarization of the incident neutrons relative to the applied magnetic field. The spin-dependent reflectivities are sufficient to define the magnetic profile of the sample, if the magnetic induction $B(z)$ is parallel to the applied magnetic field at all depths z.

As is well known,[17] if some components of the sample are magnetized along directions different from that of the applied field (which here acts as a quantization axis) the reflected neutrons are polarized in a direction different from the original one. In a second set of experiments we also analyzed the polarization of the neutron beam exiting the sample surface, by reflecting it in a polarizing mirror magnetized in the same direction as the applied field. In this way, in principle, the reflectivities R_{++}, R_{+-}, R_{-+}, and R_{--} can be obtained (the first symbol refers to the polarization state of the incident neutrons, the second to the polarization state of the reflected neutrons). Since the equipment did not include a neutron flipper after the sample, only two partial reflectivities could be obtained directly, R_{++} and R_{-+}, and the other quantities had to be derived by using the relations $R_{-+}=R_{+-}$, $R_+=R_{++}+R_{+-}$, and $R_-=R_{--}+R_{-+}$.

The samples were mounted on the cold finger of a displex-type closed-cycle refrigerator which allowed measurements between 15 and 300 K. Measurements were taken on the same sample for two configurations, with the neutron beam either reflected from the front side, or alternatively from the back side after passing through the Si substrate. If the sample were nonabsorbing the analysis of the reflectivities for both configurations would yield the same information: the magnetization profile of the entire multilayer. Gd, however, is a very strong neutron absorber and thus the reflected neutrons sense only a limited number of layers closest to the surface on which they impinge. Specifically, for our set-up the penetration depth is about 600 Å, i.e., only the first seven of the total of 15 bilayers are seen by the neutrons.

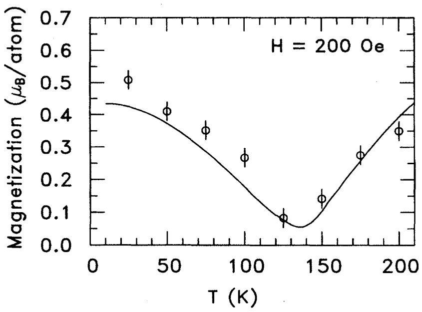

FIG. 2. Temperature-dependent magnetization of the [Gd 50 Å/Fe 35 Å]×15 multilayer measured in an external field of 200 Oe. Solid line: magnetization measured with a SQUID magnetometer, data points: magnetization deduced from the polarized neutron reflectivity measurements. The magnetization is given in μ_B/atom (for the average, all Fe and Gd atoms are taken into account).

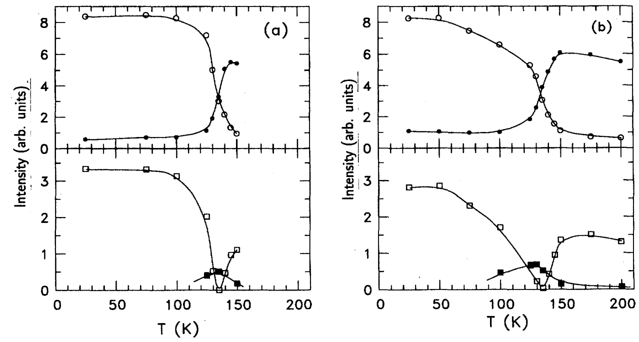

FIG. 3. Integrated Bragg intensities $I_+ = I_{++} + I_{+-}$ (full circles), $I_- = I_{--} + I_{-+}$ (open circles) and magnetic intensities parallel ($I_\parallel$, open squares) and perpendicular ($I_\perp$, full squares) to the field. The intensities were deduced from reflectivity measurements on the [Gd 50 Å/Fe 35 Å]×15 multilayer at $H = 200$ Oe. (a) measurement from the front side (Gd first layer), (b) measurement from the back side (Fe first layer) of the sample. Thin lines are guides for the eye.

Neutron measurements were taken of both front and back faces of the [Fe/Gd] as well as the [Gd/Fe] sample. The overall magnetization of both samples was measured by superconductivity quantum interference device (SQUID) magnetometry at several temperatures. With magneto-optic Kerr effect measurements (MOKE) the magnetization of the surface layers was measured. MOKE measurements were even more surface sensitive than neutrons, but they were limited to the exposed face of the multilayers.

III. MAGNETIC STRUCTURE

We will first discuss the phase transition that occurs as a function of the temperature in a low magnetic field, and then the field-dependent transition at low temperatures. In all cases we will make use of the neutron reflectivity and the magnetization measurements (SQUID and MOKE). The error of the SQUID data and the statistical error of the neutron reflection data for $R \gtrsim 10^{-3}$ are within the symbol size used. At $Q_z \to 0$ the polarized neutron reflectivity curves (see, e.g., Fig. 5) do not reach the well-defined plateau characteristic of total reflection. This is a consequence of the large absorption cross section of gadolinium. The first Bragg reflection is observed at $Q_z \approx 0.075$ Å^{-1}.

A. Temperature dependence

In Fig. 2 the magnetization of the [Gd/Fe] sample measured by a SQUID magnetometer at $H = 200$ Oe is compared with that deduced from reflectivity in the low Q_z region. The magnetic induction of a uniform sample can be obtained very simply starting from the Fresnel spin-dependent reflectivities. Writing as Q_{0+}, Q_{0-} the momentum transfer values for a certain reflectivity R_0 for the two spin states, we obtain the relation

$$cB = \bar{N}\bar{p} = \bar{N}\bar{b}\,\frac{Q_{0,+}^2 - Q_{0,-}^2}{Q_{0,+}^2 + Q_{0,-}^2} \qquad (1)$$

with $c = 2\pi\mu_n m/h^2 = 2.3\times10^{-10}$ Å^{-2}/G. $\bar{b}$ and $\bar{p}$ are the average nuclear- and magnetic-scattering amplitudes, respectively. In units of 10^{-12} cm, $\bar{p}$ is equal to 0.27 $\bar{\mu}$ with the average magnetic moment $\bar{\mu}$ of the atoms in Bohr magnetons. The layered structure of our sample is significantly more complex than a homogeneous magnetic material but we may assume that from the reflectivities close to the total reflection the magnetization can still be extracted.

To calculate the average atomic density $\bar{N}$ the effective densities of Gd and Fe atoms in the sample are taken to be the same as in the bulk materials. The nuclear scattering amplitudes are $b_{\mathrm{Fe}} = 9.54\times10^{-5}$ Å and $b_{\mathrm{Gd}} = (3.0 + i11.0)\times10^{-5}$ Å where the imaginary part of the latter one expresses the strong neutron absorption of Gd.

As seen in Fig. 2, the results from reflectivity measurements are indeed quite similar to the magnetization. Both sets of data show a minimum at $T_{\mathrm{comp}} = 135$ K, indicative of the compensation point. The same compensation point is found by looking with neutrons at the Fe terminated back face of the same sample. Very similar compensation points are also obtained for the [Fe/Gd] sample. The transition at $H \approx 0$, $T = 135$ K evidently occurs uniformly across the sample thickness. At $T = 10$ K the measured magnetization (0.45μ_B/atom) is noticeably lower than the magnetization expected for a Gd-aligned state (0.98μ_B/atom). Because it is unlikely that the Fe layers have an enhanced magnetization, the low magnetization arises probably from a reduced Gd magnetization. The measured magnetization corresponds to a Gd moment reduction of 20%. The reduced Gd magnetization was found previously by other groups.[8,14]

The upper parts of Figs. 3(a) and 3(b) show for the [Gd/Fe] sample the integrated intensities $I_\pm$ of the reflectivities at the first Bragg reflection as a function of temperature. The first Bragg reflection occurs at $Q_{z,\mathrm{Bragg}} = 2\pi/\Lambda$, where Λ is the superlattice period. The measurements were taken with the neutrons entering the sample from the frontside [Fig. 3(a)] and from the backside [Fig. 3(b)] in an external field of $H = 200$ Oe. The intensities I_+ and I_- switch as a function of temperature. Their crossing is identical to the compensation temperature and occurs at the same temperature for the Gd terminated frontside and the Fe terminated backside of the sample. This confirms the conclusion that the transition occurs uniformly across the sample thickness. However, the transition takes place in a small but finite temperature range.

The results have a transparent interpretation within the first-order Born approximation. At the first Bragg reflection the different intensities are given by[15]

$$I_+ = I_{++} + I_{+-}$$
$$= \frac{1}{C}\{[N_{\rm Fe}b_{\rm Fe} - N_{\rm Gd}b_{\rm Gd} + (N_{\rm Fe}p_{\parallel,\rm Fe} - N_{\rm Gd}p_{\parallel,\rm Gd})]^2 + [N_{\rm Fe}p_{\perp,\rm Fe} - N_{\rm Gd}p_{\perp,\rm Gd}]^2\},$$
$$I_- = I_{--} + I_{-+}$$
$$= \frac{1}{C}\{[N_{\rm Fe}b_{\rm Fe} - N_{\rm Gd}b_{\rm Gd} - (N_{\rm Fe}p_{\parallel,\rm Fe} - N_{\rm Gd}p_{\parallel,\rm Gd})]^2 + [N_{\rm Fe}p_{\perp,\rm Fe} - N_{\rm Gd}p_{\perp,\rm Gd}]^2\}. \quad (2)$$

$p_\parallel$ and $p_\perp$ are the component of the magnetic scattering amplitude in field direction and perpendicular to the direction of the external field. C is a proportionality constant. Each of the spin-analyzed components corresponds to a square bracket. The lower parts of Figs. 3(a) and 3(b) show the temperature behavior of the magnetic intensities parallel $I_\parallel$ and perpendicular $I_\perp$ to the field: while the former quantity is calculated from the splitting of I_+ and I_- ($I_\parallel = C^2[(I_+ - I_-)/(4N_{\rm Fe}b_{\rm Fe} - 4N_{\rm Gd}b_{\rm Gd})]^2$) the latter quantity is directly obtained from the spin-flip intensity $I_\perp = CI_{+-}$. To plot $I_\parallel$ and $I_\perp$ on the same scale we calculated the proportionality constant C from the low-temperature measurement where I_{+-} equals zero.

The temperature behavior of the integrated intensities indicates that at $T=25$ K the multilayer is mainly in a Gd-aligned state. In a temperature range between $T=120$ to 145 K the multilayer changes to an Fe-aligned state. At yet higher temperatures the magnitude of $I_\parallel$ decreases with the size of the Gd moment.

The interpretation of the spin configuration in the transition region is more subtle. At the crossing temperature the sublattice magnetizations of Fe and Gd are equal, and even in a very weak field should turn perpendicular to the applied field. The large $I_\perp$ intensities expected on the basis of such a model are not observed: $I_\perp$ has a maximum around $T_{\rm comp}$ but is only a fraction of the total magnetic intensity. At the spin compensation temperature large portions of the Gd and Fe magnetization are oriented parallel or antiparallel rather than perpendicular to the field direction. But at $T_{\rm comp}$, $I_+ = I_-$ and therefore the sample must have equal volumes of Gd-aligned and Fe-aligned phases. This suggests that the sample is not homogeneous, but is made of regions with slightly different compensation temperatures.

The nonhomogeneity of the sample is supported by a careful analysis of the shape of the Bragg peaks. The Bragg peaks shift position as the temperature is varied across the transition region (see Fig. 4). We were unable to explain this finding by any model, in which the magnetization is uniform along the surface plane. Instead, this is the behavior expected for a sample made of a collage of crystallites with different periodicities. Those with longer superlattice periods have

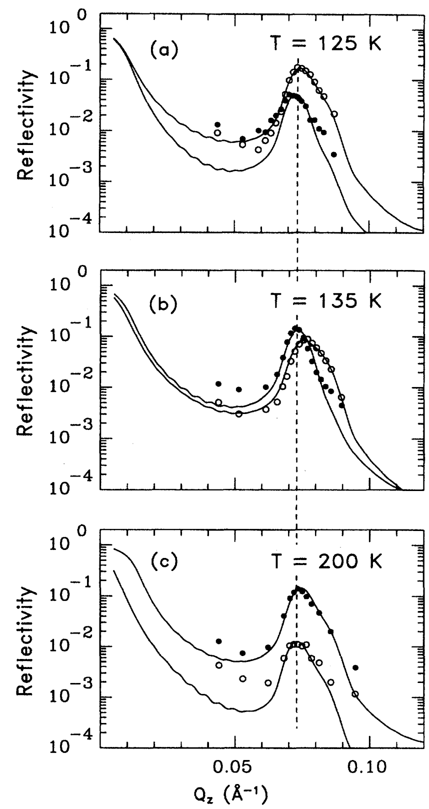

FIG. 4. Spin-dependent reflectivities of the [Gd 50 Å/Fe 35 Å]×15 multilayer measured from the back side in an external field of 200 Oe and at the temperatures given in the pictures. (●) and (○) are for neutrons polarized in and opposite to the field direction, respectively. The reflectivities are fitted (solid lines) by assuming that the sample is made of a collage of four superlattices with different periods: $L=87.6$ Å, with $T_{\rm comp}=125$ K, for 31%, $L=85.0$ Å, with $T_{\rm comp}=135$ K, for 45%, $L=80.7$ Å, with $T_{\rm comp}=145$ K, for 12%, and $L=76.3$ Å, with $T_{\rm comp}=150$ K, for 12%.

lower compensation temperature;[6] a fact verified experimentally.[12] With this idea the shift of the Bragg peaks can be explained as follows: at $T=125$ K [Fig. 4(a)] the major part of the sample is in a Gd-aligned state while the crystallites with the largest superlattice periods (and hence the lowest Q_z position of the Bragg peaks) start to change into an Fe-aligned state. Since the intensity at the first Bragg reflection I_+ is much greater for the crystallites in an Fe-aligned state than I_+ of the remaining part of the sample (see Fig. 3), the measured R_+-Bragg peak shifts to a lower Q_z position. At $T=135$ K [Fig. 4(b)] the major part of the sample is in an Fe-aligned state while only the crystallites with the shortest superlattice periods remain in a Gd-aligned state. Therefore the measured R_--Bragg peak shifts to a higher Q_z position. For example, the data in Fig. 4 can be fit with a model comprising four different layer thicknesses but with constant Fe/Gd ratio.

Implicitly, it has been assumed throughout this treatment that the size of the domains is quite large (well above the micron size), else a lateral broadening of the Bragg peaks should have been observable.[18]

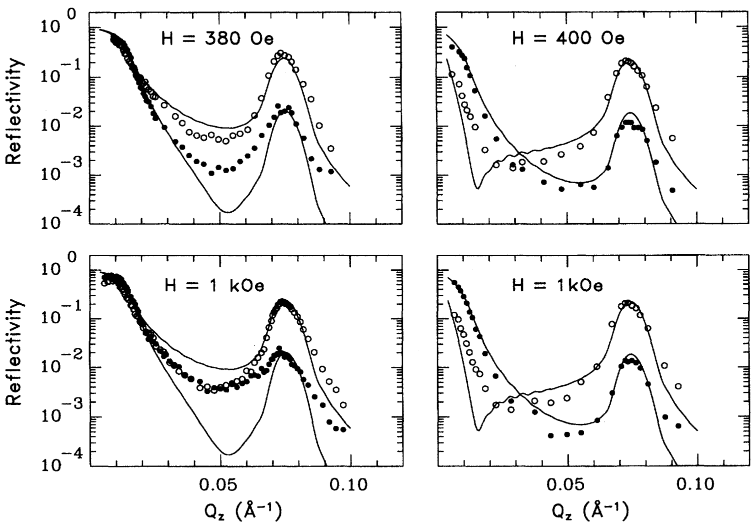

FIG. 5. Spin-dependent reflectivities of the [Fe 35 Å/Gd 50 Å]×15 sample measured at $T=15$ K from the Fe terminated front side (pictures on the left) and the Gd terminated back side (pictures on the right). Full circles: $R_{+}=R_{++}+R_{+-}$, open circles $R_{-}=R_{--}+R_{-+}$. The solid lines are reflectivities calculated for a Gd-aligned state with a Gd-moment reduced by 20% from the bulk value.

B. Field dependence (neutron reflectivity)

At magnetic fields higher than 200 Oe, the transition region vicinal to the compensation temperature becomes broader, and at the same time the spin-flip intensity which distinguishes the twisted portion of the sample becomes larger.

Since, in the Fe/Gd samples neutrons observe only the first ≈600 Å of the sample, the details of the transition region (but *not* the transition temperature) appear to be different if seen from the front and from the back face of the same sample [compare Figs. 3(a) and 3(b)]. Neutron reflectivity measurements at low temperature (15 K) show even more remarkable differences in the field behavior between the Gd terminated face and the Fe terminated face, as shown in Fig. 5 for the [Fe/Gd] sample. In the former geometry only minor changes in the reflectivities occur between 400 Oe and 1 kOe, while the changes are quite drastic for the latter over the same range of magnetic fields. The same field dependence was found for the Gd and Fe terminated face of the [Gd/Fe] sample. At the Gd terminated surface the measured reflectivities are in fairly good agreement with reflectivities calculated for a Gd-aligned state (Fig. 5). In contrast, the reflectivities of the Fe terminated surface at $H=1$ kOe are definitely not in accordance with a Gd-aligned state. This behavior of the reflectivities is expected if the transition to the twisted state was initiated at the Fe-terminated surface. (We note that the calculations were performed with a Gd moment reduced by 20%. Without the moment reduction we could not reproduce the splitting of R_{+} and R_{-} at the Bragg peak position.)

What follows is an attempt to support the explanation of the reflectivities with a surface induced phase transition by fitting some polarized neutron reflectivity measurements in terms of a model spin configuration of the surface induced phase. Figures 6(a)–6(d) show the low-temperature reflectivities of the [Fe/Gd] sample as seen from the Fe side, as well as the spin-analyzed reflectivities. At $H=380$ Oe (not shown) and $H=1$ kOe the reflectivity has only minor spin-

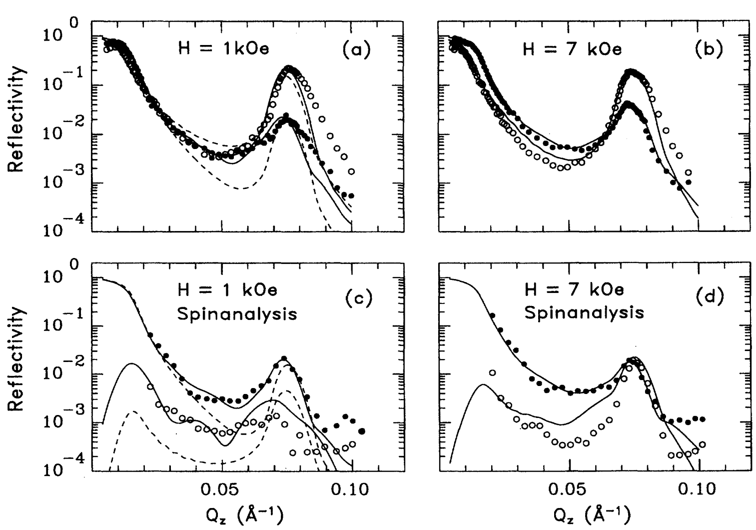

FIG. 6. Reflectivities of the [Fe 35 Å/Gd 50 Å]×15 sample at $T=15$ K and $H=1$ kOe and $H=7$ kOe measured from the front side. In (a) and (b) ● marks $R_{+}=R_{++}+R_{+-}$ and ○ marks $R_{-}=R_{--}+R_{-+}$. In (c) and (d) ● marks R_{++} and ○ marks R_{-+}. The solid lines are calculations for a model spin configuration of the surface induced phase as explained in the text. The dashed lines in (a) and (c) are calculations for a uniform model for the magnetizations with $\Theta_{Fe}=150°$ and $\Theta_{Gd}=-22°$.

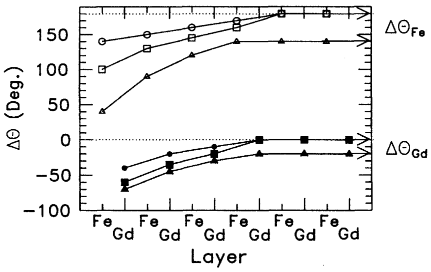

FIG. 7. Angle between the external field and the magnetizations of the individual layers used in the model calculations. Filled symbols mark the angles of the Gd layers and the open symbols the angles of the Fe layers. ○: $H=380$ Oe, □: $H=1$ kOe, △: $H=7$ kOe. The angle of the outermost layer is on the left-hand side of the picture.

flip components. Therefore the sublattice magnetizations were mainly aligned with the magnetic field. In contrast, at $H=7$ kOe the spin-flip reflectivity is strong with a peak at the Q_z position corresponding to the superlattice period as if the sample were in a coherently twisted state.

The solid lines represent reflectivities calculated for an *ad hoc* model. We assume that at low temperature the parts of the sample with different layer thicknesses are all in the same magnetic structure. Within each Fe (or Gd) layer the magnetic moments are not necessarily collinear, but we assume that this amounts only to an effective reduction of the average magnetization of this layer. For a further simplification of the model we constrain the moment reduction to be proportional to the angle between the local magnetization and the external field $M=M_{\max}-C|\Delta\Theta|$ with $\Delta\Theta=\Theta_{\text{field}}-\Theta_{\text{Gd or Fe}}$ in accordance with the model of Camley and Tilley.[5] The perpendicular components within one layer cancel out, and are not visible at low neutron momentum transfers. Reasonable fits could only be achieved with different proportionality constants for the measurements at different fields: $C_{\text{Fe}}=6\times10^{-4}$, $C_{\text{Gd}}=3\times10^{-4}$ for $H=380$ Oe, $C_{\text{Fe}}=6\times10^{-4}$, $C_{\text{Gd}}=2\times10^{-4}$ for $H=1$ kOe and $C_{\text{Fe}}=1.5\times10^{-4}$, $C_{\text{Fe}}=1\times10^{-4}$ for $H=7$ kOe. For Gd we used for the maximal magnetization $M_{\max}$, the bulk value reduced by 20%.

The angles $\Delta\Theta$ used for the calculations are shown in Fig. 7 for $H=380$, 1000, and 7000 Oe. Even in a field as low as $H=380$ Oe the magnetizations of the first Fe layers are already turned into the direction of the external field. The penetration depth of the surface induced phase transition is approximately three bilayers of Fe/Gd and appears to be largely independent of the external field.

In view of the complexity of the system and the limited observations, a number of assumptions had to be made and each of them can be individually challenged. However, we could not find any structural transition that was uniform across the sample thickness and still would fit the data. For instance, we can rule out the model of a twisted state in which both the moment reduction and the angle of magnetization relative to the field are uniform throughout the sample. In Figs. 6(a) and 6(c) the dashed lines show the best correspondence between calculated and measured reflectivi-

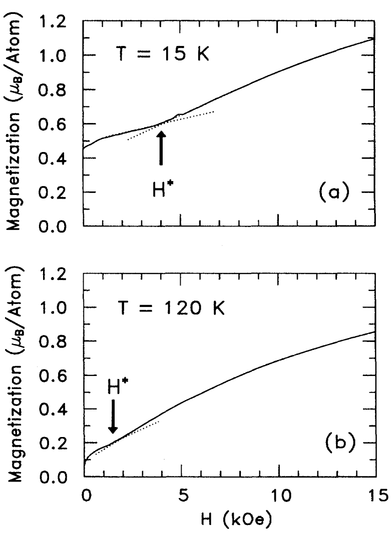

FIG. 8. Magnetization of the [Gd 50 Å/Fe 35 Å]×15 sample as a function of the magnetic field measured at the temperatures given in the pictures. H^* marks the onset of the twisted state. The magnetization is given in μ_B/atom (for the average all Fe and Gd atoms are taken into account).

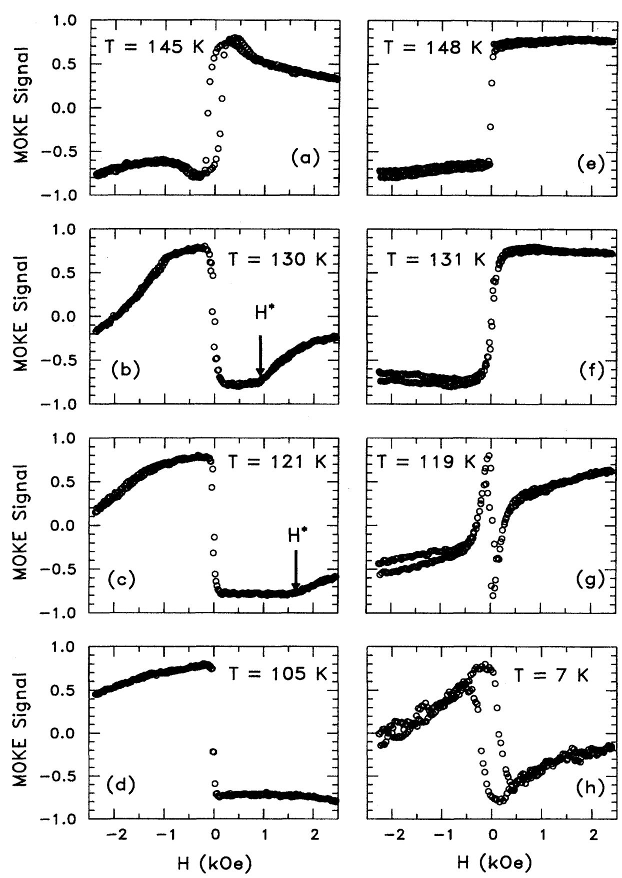

FIG. 9. MOKE patterns at different temperatures for the Gd terminated [Gd 50 Å/Fe 35 Å]×15 multilayer (left-hand side) and the Fe terminated [Fe 35 Å/Gd 50 Å]×15 multilayer (right-hand side).

ties that we could obtain with this uniform model. This model overestimates the spin flip reflectivity at the Bragg peak position. However, a fitting procedure does not exclude *a priori* all conceivable (but untested) uniform models. Thus it is worthwhile to seek confirmation to our conclusions from less sophisticated, but more direct experiments.

C. Field dependence (magnetometry)

Possibly the best measurement of the transition from the aligned to the twisted state is a magnetization measurement, which averages in an unbiased way over the magnetization of all layers. The magnetization curve of the [Gd/Fe] multilayer measured at $T=15$ K [Fig. 8(a)] and $T=120$ K [Fig. 8(b)] shows a "kink" at $H=4$ kOe and $H\approx1.7$ kOe, respectively. This is inferred[11,12] to represent the phase transition from the aligned to the twisted state in accordance with Camley's model. This model predicts a plateaulike magnetization for all fields in which the sublattice moments are in field-aligned states, followed by a steep increase of the magnetization when the moments are twisted. Similar results for the transition fields are obtained for the [Fe/Gd] sample.

In order to see the surface induced transition, we made extensive MOKE measurements at the surface of both the [Fe/Gd] and [Gd/Fe] samples. MOKE is really surface sensitive, with an effective penetration depth of the order of 200 Å, or two bilayers. In the experimental set-up, the polarization of the incident light was perpendicular to the applied magnetic field, which was always applied in the plane of the sample. In this geometry the signal (the rotation of the polarization vector in the reflected beam) is strictly proportional to the component of magnetization parallel to the applied field.[19] For the wavelength of the incident light (750 nm) the proportionality constant of Gd (Ref. 19) is one order of magnitude lower than the proportionality constant of Fe.[20] Therefore, only Fe contributes essentially to the rotation of the polarization vector of light. The signal is then proportional to the component of the magnetization of the first two Fe layers parallel (positive values) or antiparallel (negative values) to the direction of the applied field.

The results, as obtained in sweeping the magnetic field at different temperatures, are presented in Fig. 9. For the Gd terminated sample at $T=105$ K we observe [Fig. 9(d)] a step at zero field followed by an almost constant MOKE signal up to the maximum field of 2.5 kOe (the weak slope of the MOKE signal for negative fields is an artifact due to the saturation of the photodiode). This can be interpreted as the Gd-aligned phase, because the first two Fe layers are oriented opposite to the applied field. Magnetic fields up to 2.5 kOe are not sufficient to cause the twist of the moments which, in respect to the observed surface, should originate in the interior. At $T=121$ K [Fig. 9(c)] the MOKE curve shows a "kink" at a field $H^*=1.7$ kOe. The increase of the MOKE signal above H^* indicates that the magnetization of the first two Fe layers starts to turn into the field direction. Indeed, the value of H^* deduced from MOKE measurements is in accordance with that deduced from SQUID magnetization measurements [Fig. 8(b)]. Above the compensation temperature [Fig. 9(a)] the MOKE signal is reversed because the sample is Fe aligned. However, the field for which twisting occurs is not exactly symmetric to that for $T<T_{\rm comp}$ for the following reason. The surface layer is still Gd: its magnetization is now opposite to the applied magnetic field when this is weak but quickly twists when H is raised, and the vicinal Fe is dragged with it.

The interpretation given above is totally consistent with that obtained by measuring the MOKE signal of an Fe terminated sample, as presented in the second column of Fig. 9. Above the compensation temperature, at $T=148$ K [Fig. 9(e)], the twist is expected to proceed from the interior and to appear at this Fe-terminated surface at considerably higher field. At $T=119$ K the MOKE signal is negative (the sample is now Gd aligned) but a field of $\approx$50 Oe, significantly lower than the transition field $H^*=1.7$ kOe of the entire sample, is sufficient to turn the magnetization of the first Fe layer into the field direction. At $T=7$ K [Fig. 9(h)] the transition is much more gradual: the strong reduction of the MOKE signal in a field of 2.5 kOe indicates that the magnetization of the first Fe layer is approximately perpendicular to the applied field. In conclusion, the combined SQUID and MOKE measurements show very immediately and visually that in Gd/Fe multilayers the transition from an aligned to a twisted state is initiated at the surface if the minority moment is at the surface, or from the interior if at the surface is the majority moment.

IV. CONCLUSIONS

Our experimental observations on Fe/Gd sputtered multilayers confirm the applicability of Camley's model. Besides the three phases, Gd aligned, Fe aligned, and twisted, we observed that the field-dependent phase transition is surface induced as predicted by Camley.

The phase transition at the compensation temperature was found to be independent of the surface layer and occurs uniform across the samples thickness. However, the magnetic structure at the compensation temperature is much more complex than predicted by Camley's model. Presumably this is a consequence of imperfections in the layer thicknesses of our samples.

Taking at face value the interpretation of the neutron reflectivity measurements at $T=15$ K, the sublattice magnetizations are never perfectly aligned in the direction of the applied field. Even at very low fields we observe the occurrence of the surface induced phase transition. Furthermore, all attempts to describe the polarized neutron reflectivity data with the detailed magnetic structure calculated using the energy minimization algorithm given in Ref. 4 failed. Possibly, this is the point where a simple theoretical model which includes only Zeeman and exchange terms starts showing its inadequacy.

Finally the experiments indicate that the Gd magnetization is reduced by 20%. Possible reasons for the moment reduction are imperfections in the bulk or in the interfaces of the Gd layers. Two arguments suggest that the interface might be imperfect. No sample has been obtained up to now with molecular beam epitaxy. Iron and gadolinium in bulk tend to produce compounds over a range of stoichiometries. These compounds order ferrimagnetically, with the magnetic moments of the Gd and Fe sublattices oppositely oriented.

The perfection of the Gd/Fe interface, which is crucial for a contact interaction, is still to be demonstrated, possibly via x-ray diffraction or Mössbauer techniques. The results from these studies may help to resolve the observed differences between theory and experiment.

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