Magnetic X-Ray–Scattering Study of Interfacial Magnetism in a Gd-Y Superlattice

C. Vettier,[a] D. B. McWhan, E. M. Gyorgy, J. Kwo, and B. M. Buntschuh
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

and

B. W. Batterman
Cornell University, Ithaca, New York 14850
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The modulation in the magnetic moment and in the interplanar spacing (strain) in the superlattice $[Gd_{21}Y_{21}]\times 40$ has been determined by use of synchrotron and rotating-anode x-ray sources. The average moment appears to decrease in the interfacial regions, and the width of the strain interface increases with decreasing temperature.

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The technique of magnetic x-ray scattering, which was developed by Platzman and Tzoar[1] and by DeBergevin and Brunel,[2] has been used to determine the modulation in the magnetic moment in a superlattice composed of alternating regions of ferromagnetic Gd and nonmagnetic Y. Recently, the high flux and polarization of a synchrotron x-ray source has been used to gain new insight into the antiferromagnetic structure of Ho metal where the weak magnetic reflections are separated in reciprocal space from the much stronger Bragg reflections.[3] In this Letter, we use the tunability of the x-ray energy from a synchrotron source to maximize the cross term between the charge and magnetic scattering so as to measure the small changes in intensity in the Bragg reflections of a ferromagnet which occur when the direction of the spin is reversed with an external magnetic field. There is considerable interest in the properties of the interfaces between magnetic and nonmagnetic materials, and superlattices provide a means of studying an array of interfaces. Recent studies of the magnetization of Gd_nY_m superlattices show a reduced average magnetic moment,[4] and we find that the magnetic scattering data are consistent with the reduced average moment per Gd resulting from a smooth decrease in the projected moment on going from the center of a Gd region toward the interfaces. In addition, the temperature dependence of the interplanar-spacing modulation, which has a large magnetostrictive component, shows a thicker interface than that of the composition modulation.

Measurements were made on a superlattice composed of alternating regions of 21 atomic layers each of Gd and Y which were repeated 40 times, and the synthesis and bulk magnetic properties of this sample are given in Ref. 4. For scattering vectors perpendicular to the layers, the reciprocal lattice at room temperature is $Q(l,m)=2\pi(l/c+m/\Lambda)$, where $c=2\times 2.879$ Å is the average lattice constant (twice the interplanar spacing for a hexagonal-close-packed structure), $\Lambda=42\times 2.879=120.9$ Å is the modulation wavelength, and l and m are integers which denote the average Bragg reflections and their harmonics, respectively. The scattering intensity per repeat distance is given in a relativistic quantum theory for a centrosymmetric ferromagnetic structure by

$$I(l,m)=A[F'(l,m)^2+F''(l,m)^2 -(2\lambda_c/\lambda)F''(l,m)(\mathbf{k}_i\times\mathbf{k}_f)\cdot\mathbf{S}(l,m)], \tag{1}$$

where A is a constant which includes Lorentz and absorption factors, $\lambda_c=h/mc=0.02426$ Å, λ is the x-ray wavelength, and $\mathbf{k}_i$ and $\mathbf{k}_f$ are the incident and scattered wave vectors.[2] This expression is valid up to first order in λ_c/λ, and neglects the orbital contributions to the magnetic moment which are small for Gd. For simplicity we show the expression assuming complete linear polarization of the incident beam normal to the scattering plane, but the full polarization matrix is used in the calculations.[2] The structure factors for the charge, $F(l,m)$, and spin, $\mathbf{S}(l,m)$, are sums over the repeat distance of 42 atomic layers:

$$F(l,m)=F'(l,m)+iF''(l,m) =\sum_j (f_0+f_j'+if_j'')e^{iQ(l,m)z_j}, \tag{2}$$

$$\mathbf{S}(l,m)=\sum_j \tfrac{1}{2}\boldsymbol{\mu}_j f_m e^{iQ(l,m)z_j},$$

where f_0+f', if'', f_m, and $\boldsymbol{\mu}_j$ are the real and imaginary parts of the atomic form factors, the magnetic form factor, and the average magnetic moment per layer in Bohr magnetons, respectively. The relative importance of $F'(l,m)$ and $F''(l,m)$ can be varied by tuning of the x-ray energy through the L absorption edges of Gd. Near the L_{III} edge of Gd, f' and f'' change by 30 electrons relative to f_0 which is 64 electrons at $\mathbf{Q}=0$.[5] The dispersion corrections (f' and f'') used in the calculations were obtained from a composite of (1) values in Ref. 5 for Gd, (2) scaled values for the three edges of Cs,[6] and (3) calculated values.[7] The magnetic form factor is from Freeman and Desclaux.[8]

Before our determining the magnetic-moment modulation, the temperature dependence of the strain modulation is obtained in order to separate the magnetic contribution [see Eq. (1)]. Following Ref. 4, the modulations in the composition and the interplanar spacing as a function of the layer number, N, are approximated by three-parameter curves. Each modulation has an amplitude (C_0 for composition and D_0 for spacing), the number of nominal Gd layers per modulation wavelength ($N_{Gd}=21$), and an interfacial thickness for the composition, N_I^C, and for the spacing, N_I^D. The parameters were determined by minimization of the quadratic-weighted R factor[9] for 20–25 measured intensities, and the results are shown in Fig. 1 along with the resulting models for the interplanar-spacing modulation at $T=12$ and 333 K. The data for D_0 agree well with the volume magnetostriction of Gd, which varies as the square of the magnetization, as indicated by the two curves. The interfacial width of the spacing modulation increases above that for the composition below the Curie temperature, T_C. The anisotropy and the exchange are sensitive to the local strain, and the variation in N_I^D implies that the magnetic properties will vary across the Gd region from the center to the interface.

The magnetic-moment modulation was determined as illustrated in the inset of Fig. 2. An x-ray beam from the Cornell high-energy synchrotron source (CHESS) of $\sim 10^{12}$ photons/sec at the sample was obtained from a double-crystal monochromator using Si(111) crystals with the second crystal being sagitally focused. Either an ion chamber or a NaI scintillation detector was used depending on the intensity of the reflection. The magnetic contribution to the scattering varies as the projection of $\mathbf{S}(l,m)$ on the normal to the scattering plane, $S_\perp(l,m)$ [Eq. (1)]. A magnetic field of 100–150 Oe is used to reverse the direction of $\mathbf{S}(l,m)$ and consequently the sign of the magnetic contribution. (This field is greater than 2.5 times the measured coercive field for this film.) The change in intensity on field reversal is

$$\frac{\Delta I}{I}=\frac{4\lambda_c}{\lambda}\frac{F''(l,m)|S_\perp(l,m)|\sin(2\theta)}{F'(l,m)^2+F''(l,m)^2}, \tag{3}$$

where θ is the Bragg angle. The flipping ratio can be varied by tuning of the x-ray energy through the L_{II} and L_{III} absorption edges of Gd, which are shown at the top of Fig. 2. The observed flipping ratios for $Q(4,2)$ and $Q(4,3)$ at $T=200$ K are compared with the best model (solid curves in Fig. 3), and the demonstration that the flipping ratio versus energy follows the functional form of Eq. (3) is strong evidence that the observed ratios are magnetic in origin.

In order to determine the modulation in the magnetic moment, the flipping ratios for reflections $Q(l,m)$ with as wide a range of l and m as possible were determined. In all, twenty flipping ratios, which vary by a factor of 50 in $\Delta I/I$ and 10^5 in I, were measured at

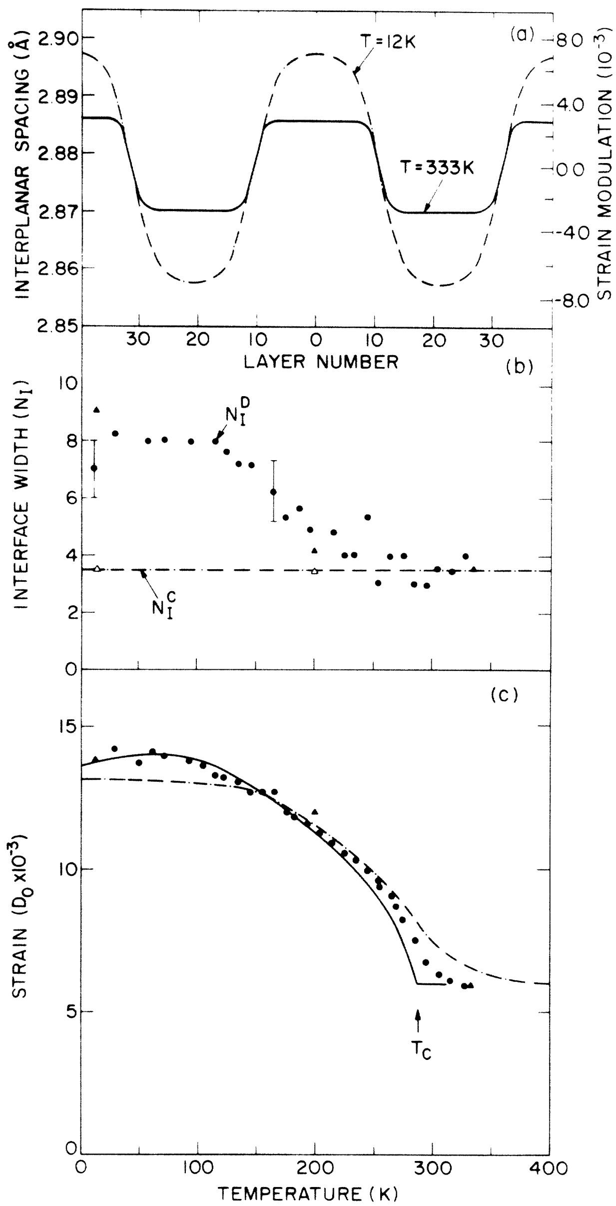

FIG. 1. (a) The interplanar-spacing modulation at $T=12$ and 333 K calculated as described in the text by use of the parameters in (b) and (c). (b) The variation with temperature of the interface thicknesses, N_I^D and N_I^C. (c) The variation with temperature of the amplitude of the spacing modulation, D_0. The dash-dotted curve is calculated by use of the thermal expansion of bulk Gd and Y (Ref. 10), and the solid curve is the square of the bulk magnetization measured in a field of 80 Oe normalized to the data at low and high temperatures. Triangles and circles are separate experiments.

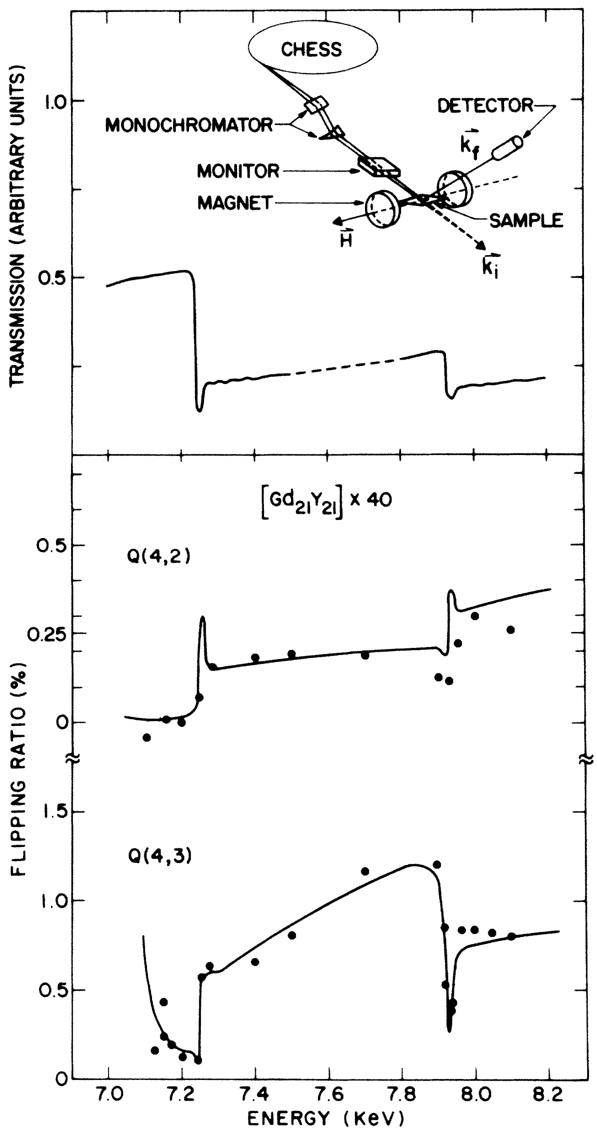

FIG. 2. Top: transmission through a Gd foil as a function of energy near the L_{II} and L_{III} absorption edges. Bottom: the observed (circles) and calculated (solid curves) flipping ratios for reflection at $Q(4,2)$ and $Q(4,3)$. The experimental arrangement is shown in the inset.

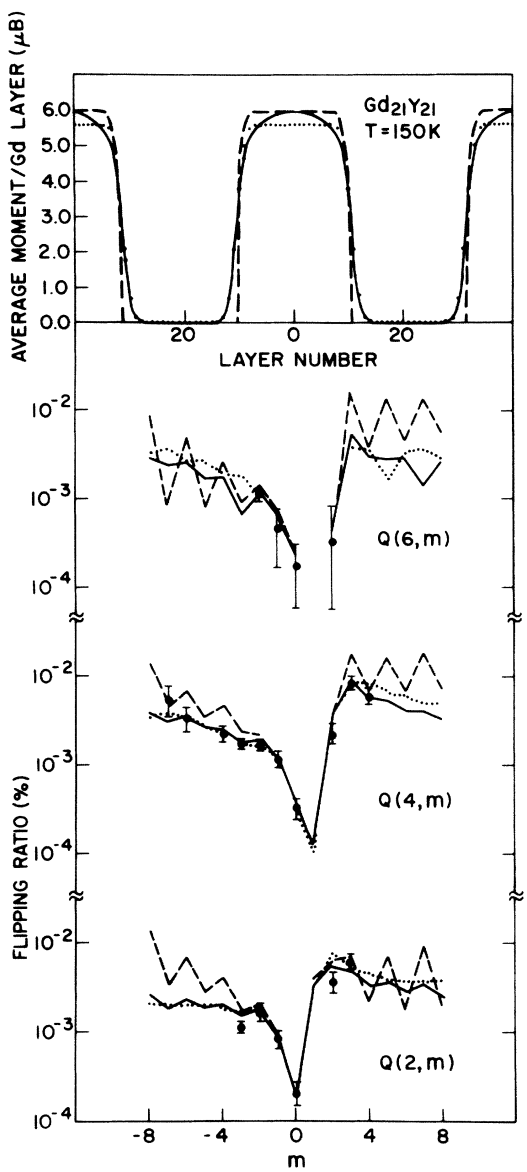

FIG. 3. Top: three models for the magnetic-moment modulation. Bottom: the calculated flipping ratio corresponding to no magnetic contribution from the two interfacial layers (dashed curve), a smooth decrease in moment (solid curve), and a uniform reduction in moment (i.e., a moment modulation that comes from the composition modulation) (dotted curve), compared with the values measured at 8.04 keV.

$T = 150$ K, and they are compared with three models for the spin structure in Fig. 3. For $Gd_{21}Y_{21}$ at $T = 150$ K an average moment per Gd of $6.0\mu_B$ is calculated from the magnetization measured in a field of 80 Oe. Our x-ray measurements are only sensitive to the local moment, which is $5.6\mu_B$ per Gd, if we assume a 7% correction for conduction-electron polarization as in bulk Gd. The magnetic moment per layer, μ_j, in Eq. (2) is the product of the Gd concentration and the moment per Gd in the layer. Thus, the magnetic-moment modulation would follow the composition modulation if the magnetic moment per Gd were reduced uniformly. The dashed curve, calculated on the assumption of no magnetic contribution from the interfacial layers, is not consistent with the measurements ($R = 0.25$); the dotted curve, which assumes a uniform reduction in the projection of the Gd moment parallel to the field, gives better agreement ($R = 0.18$); and the solid curve, which assumes the full Gd moment at the center with a smooth reduction in the projected moment as the interface is approached, gives the best fit ($R = 0.14$). The magnetic x-ray measurements are consistent with the reduced average moment in $Gd_{21}Y_{21}$ resulting from a decrease in the projected moment near the interfaces between Gd and Y.

In conclusion, we have demonstrated that the development of magnetic order in a superlattice can be determined from x-ray scattering measurements using a synchrotron source. Polarized neutron scattering is the traditional probe of magnetic structure,[11,12] but magnetic x-ray scattering is a competitive probe for superlattices because of the small sample volume and the need to measure the weak, higher-order harmonics.

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[(a)]Permanent address: Institut Laue-Langevin, Grenoble, France.

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