Observation of a Magnetic Antiphase Domain Structure with Long-Range Order in a Synthetic Gd-Y Superlattice

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A microscopic magnetic antiphase domain structure has been observed in a single-crystal Gd-Y superlattice by neutron diffraction. Furthermore, this long-range antiferromagnetic correlation is found to occur in a multibilayer, in which each bilayer consists of N_{Gd} ferromagnetic atomic planes of Gd followed by N_{Y} planes of nonmagnetic Y, for $N_{\mathrm{Y}} = N_{\mathrm{Gd}} = 10$ but not for $N_{\mathrm{Y}} = 6$ or 20. This oscillatory behavior is consistent with recent theoretical speculation that the Gd moments are coupled through the intervening Y via the Ruderman-Kittel-Kasuya-Yosida interaction.

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Thin-film multilayers have become a subject of great scientific and technical interest ever since Esaki and Tsu[1] proposed growing single-crystal multiple bilayers of two different semiconductors. Significant modification of the electronic properties has been shown to occur as a result of the artificially imposed periodicity.[2]

The interest in synthesizing such novel structures is not, however, limited to semiconductor materials. Considerable theoretical efforts have been made in recent years to describe the magnetic states of surfaces and interfaces.[3,4] Magnetic layers made up of a discrete number of atomic planes of moments can be deposited alternately with nonmagnetic layers of a given thickness as a model system for the investigation of both the interfacial magnetism and the effects of reduced dimensionality.[5]

It is also possible to study interlayer magnetic coupling in synthetic superlattices. In the case of the metallic, magnetic rare earths (RE), the magnetic moments are well localized and the indirect exchange interaction is via the conduction electrons. The long-range nature of this Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction can be expected to give rise to a modulation of the magnetic properties in an artificially layered magnetic-RE–nonmagnetic-RE structure.

We report here the results of a neutron-diffraction study of $[\mathrm{Gd}_{N_{\mathrm{Gd}}}\text{-}\mathrm{Y}_{N_{\mathrm{Y}}}]_M$ superlattices composed of M successive bilayers of N_{Gd} basal planes of hexagonal-close-packed Gd followed by N_{Y} such planes of Y. It is found that below the Curie temperature and in low fields the ferromagnetic Gd layers tend to align antiferromagnetically relative to one another for $N_{\mathrm{Gd}} = N_{\mathrm{Y}} = 10$ in a microscopic antiphase domain structure that is coherent over many superlattice periods. For $N_{\mathrm{Y}} = 6$ or 20, however, simple long-range ferromagnetic order is observed. This oscillatory dependence on N_{Y} is consistent with a theory which attributes the coupling between Gd layers to the RKKY interaction.[6] This interpretation is also supported by the recent observation of *incommensurate*, long-range spiral magnetic order in a Dy-Y superlattice.[7]

The preparation of the Gd-Y superlattices is described elsewhere.[5,8] Detailed x-ray diffraction measurements[5,9] have shown that Gd-Y superlattices have a composition modulation profile corresponding to two-atomic-plane–sharp interfaces with the modulation amplitude approaching 100%. The neutron-diffraction measurements were performed at the high-flux beam reactor at Brookhaven National Laboratory and at the high-flux isotope reactor at Oak Ridge National Laboratory. A typical spectrometer configuration is shown in the lower part of Fig. 1. Neutrons are first simultaneously monochromatized and polarized, and then scattered by the sample [a flat coil flipper (FLP) preceding the sample permits selection of either of the two neutron spin states]. The second polarizing crystal or analyzer (along with another flipper) distinguishes spin-flip from spin-nonflip scattering.

Four spin-dependent scattered intensities $I_m^{++}(Q)$, $I_m^{--}(Q)$, $I_m^{+-}(Q)$, and $I_m^{-+}(Q)$ can be measured.[10] The two superscripts denote the initial and final neu-

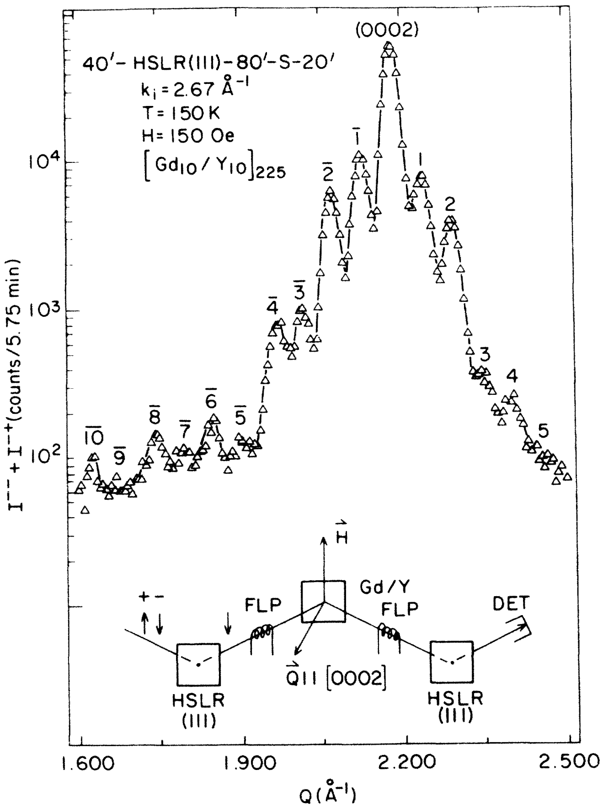

FIG. 1. Representative neutron-diffraction data for a Gd-Y superlattice as described in the text. The lower part of the figure is a diagram of the spectrometer configuration used to separate the four possible spin-dependent scattered intensities for polarized neutrons. HSLR(111) denotes the reflecting planes of the Cu$_2$MnAl Heusler polarizing crystals.

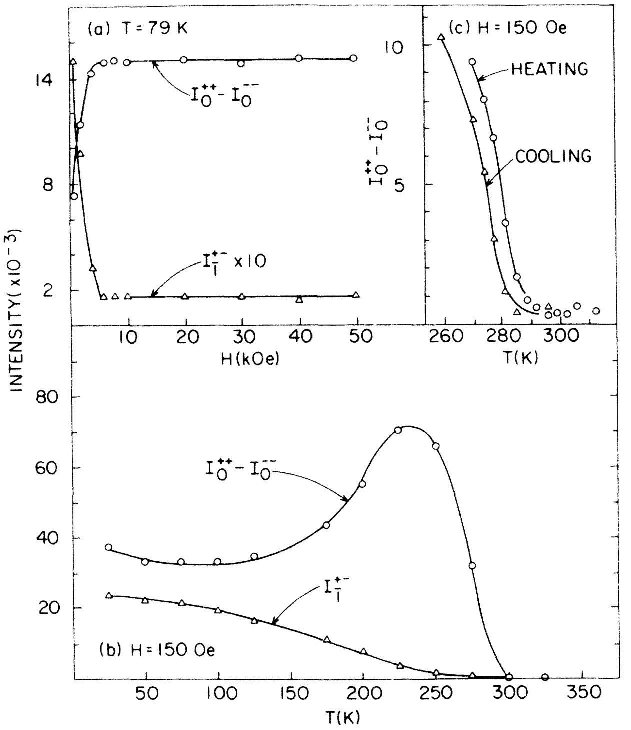

FIG. 2. (a) Magnetic field dependence and (b),(c) temperature depedence of $I_0^{++} - I_0^{--}$ ($\propto$ magnetization) and of $I_{\bar{1}}^{+-}$ for the [Gd$_{10}$-Y$_{10}$]$_{225}$ superlattice.

tron spin states, respectively, and the subscript labels the order of a satellite about a given primary Bragg reflection. $|\mathbf{Q}| \equiv |\mathbf{k}_f - \mathbf{k}_i|$, where $\mathbf{k}_f$ and $\mathbf{k}_i$ are the final and initial neutron wave vectors, respectively. Satellite intensities were measured about the (0002) and (0004) reflections as well as the forward direction with **Q** parallel to the c axis. After deconvolution from the instrumental resolution width, the natural widths of all of the observed satellite reflections were found to be approximately equivalent and to correspond to a coherence length of at least 1000 Å, or more than seventeen superlattice periods.

We will first discuss the [Gd$_{10}$-Yd$_{10}$]$_{225}$ sample in detail, for which representative data taken at 150 K in a field of 150 Oe are shown in Fig. 1. The positions Q_m of the even-number satellites measured from the primary (0002) reflection correspond to integer multiples of $2\pi/\lambda_{\mathrm{SL}}$, where $\lambda_{\mathrm{SL}} = 58.45$ Å is the chemical modulation wavelength or bilayer thickness [i.e., $Q_m = (m/2)2\pi/\lambda_{\mathrm{SL}}$]. Above T_c ($\simeq 290$ K) the intensities of the even-numbered satellites arise solely from the nuclear scattering associated with the superlattice chemical modulation so that $I_m^{++} = I_m^{--}$, whereas below T_c it is found that $I_m^{++} \neq I_m^{--}$ which indicates the presence of an additional ferromagnetic component. To within experimental accuracy, no spin-flip scattering is found to occur at the even-numbered satellite positions. The odd-numbered satellites, on the other hand, begin to appear below T_c in low fields at integer multiples of the wave vector for a *doubled* bilayer thickness [e.g., for $m = 1$, $Q_1 = (\frac{1}{2})2\pi/\lambda_{\mathrm{SL}}$; for $m = 3$, $Q_3 = (\frac{3}{2})2\pi/\lambda_{\mathrm{SL}}$, and so on] with intensities corresponding to spin-flip scattering only.

The temperature and magnetic field dependences of the difference $I_0^{++} - I_0^{--}$, which is proportional to the net sample magnetization along the direction of the applied field (perpendicular to **Q**), and of $I_{\bar{1}}^{+-}$ are plotted in Fig. 2. At 79 K a field of several thousand oersteds simultaneously saturates the magnetization and reduces the intensity of the odd-numbered satellites to within background [Fig. 2(a)]. As the temperature is lowered below T_c, the magnetization in a field of 150 Oe initially increases but eventually decreases as the intensity of the $m = -1$ satellite continues to grow [Fig. 2(b)]. Near T_c hysteresis is observed [Fig. 2(c)].

Appropriate corrections were made for the instrumental polarizing and flipping efficiencies, and a contribution to the superlattice (0002) intensity due to the

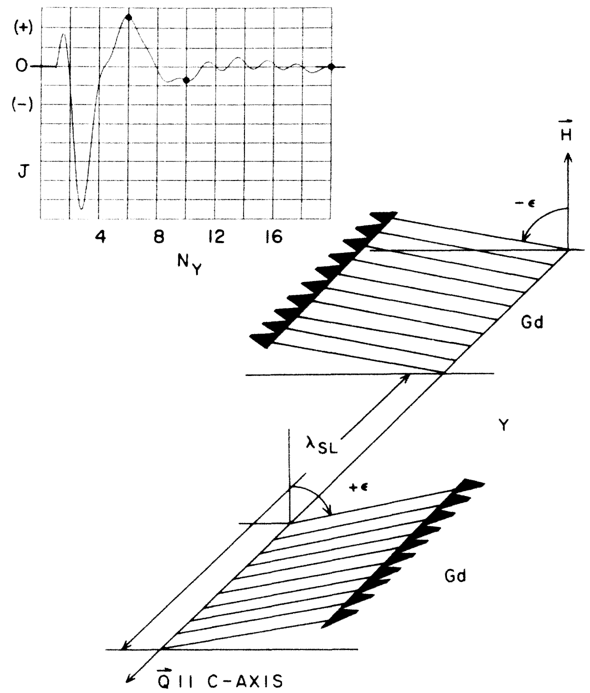

FIG. 3. Schematic representation of the antiphase domain configuration of the Gd magnetic moments in the $[Gd_{10}$-$Y_{10}]_{225}$ superlattice. The inset in the upper left-hand corner shows the results (solid curve) of the theoretical calculation of the relative strength and sign of the RKKY interaction as described in the text.

superposition of the (0002) reflection of a pure Y seed layer (amounting to about 16%) was subtracted. Further analysis of the data indicated that extinction effects were negligible. The measured intensities integrated over the entire resolution volume (with overall uncertainties of the order of 5%) were then compared to values predicted by various models for the magnetic structure. It was found that the antiphase domain model depicted in Fig. 3 (in which the atomic interplanar spacing and net magnetization per plane are uniform) is a good approximation. It is assumed that the Gd moments are confined to the basal plane, even in low fields, on the basis of the results of magnetization measurements performed on a number of samples.[11] The magnetic form factor for elemental Gd[12] was assumed and the relatively small changes in absorption and Debye-Waller factors over a limited Q range were neglected, although it was necessary to include the imaginary part of the average nuclear scattering length for the naturally occurring isotopic mixture of which the Gd layers were composed. However, better agreement is obtained with a more refined version of this basic model which includes the chemical composition and atomic-plane-spacing modulations along the c axis deduced from the x-ray measurements, as well as a reduction in the net ordered moment of the interface planes that is consistent with both magnetization[5] and magnetic x-ray scattering data.[9] The intensities calculated according to this improved model are compared to the measured values in Table I along with the corresponding "flipping ratios" $R_m^{++/--} \equiv I_m^{++}/I_m^{--}$. The calculated values are those which give the best fit and are obtained for an individual interior Gd moment magnitude of $6.3\mu_B$, in close agreement with the measured bulk value,[12] and for an angle ϵ (see Fig. 3) of 80° at 150 Oe. Models for other moment configurations including various helical arrangements gave significantly poorer agreement with the data.

The magnetic behavior of each of the other two samples, namely $[Gd_{10}$-$Y_6]_{189}$ and $[Gd_{10}$-$Y_{20}]_{100}$, is quite striking in contrast to that of the superlattice for which $N_Y = 10$. Neither develops antiferromagnetic correlations down to a temperature of 10 K in zero field. Both samples do, however, exhibit long-range ferromagnetic order below T_c.

One particularly appealing explanation to account for the observed oscillatory behavior is to attribute the coupling between two Gd layers across an intervening Y layer to the RKKY interaction. In order to estimate the strength and sign of this coupling, a calculation of the interaction between two Gd monolayers separated by pure Y (for which the calculated susceptibility func-

TABLE I. Comparison of observed intensities (normalized to $I_{\bar{2}}^{++}$) and flipping ratios for $[Gd_{10}$-$Y_{10}]_{225}$ with corresponding values calculated for the antiphase domain model described in the text.

Reflection order m $(0002)^m$	Q (Å^{-1})	T (K)	H (Oe)	I_m^{++} Obs.	I_m^{++} Calc.	I_m^{--} Obs.	I_m^{--} Calc.	I_m^{+-} Obs.	I_m^{+-} Calc.	I_m^{-+} Obs.	I_m^{-+} Calc.	$R^{++/--}$ Obs.	$R^{++/--}$ Calc.
$\bar{4}$	1.968	75	150	0.046	0.008	0.050	0.011	0	0	0	0	0.93	0.73
$\bar{3}$	2.021	75	150	0	0	0	0	0.53	0.51	0.52	0.51	···	···
$\bar{2}$	2.075	75	150	1.00	1.00	1.05	1.10	0	0	0	0	0.96	0.91
$\bar{1}$	2.129	75	150	0	0	0	0	1.70	1.77	1.69	1.77	···	···
0	2.183	75	150	4.46	4.54	3.10	3.21	0	0	0	0	1.44	1.42
1	2.236	75	150	0	0	0	0	1.22	1.33	1.25	1.33	···	···
2	2.290	75	150	0.50	0.53	0.65	0.70	0	0	0	0	0.77	0.76

tion was assumed[13]) was performed.[6] While the validity of this approximation has not been rigorously tested, it is probably the simplest physically plausible computation that can be made short of doing a superlattice band calculation. The sign and relative strength of the interaction obtained from this calculation are plotted in the inset of Fig. 3 as a function of N_Y. The points on the theoretical curve indicate those values of N_Y for which the magnetic structures have been determined and show that the experimental observations are indeed consistent. Moreover, magnetization measurements performed on the three samples studied by neutron diffraction and five additional samples with $N_Y = 9$, 11, 14, 16, and 24 (and $N_{Gd} = 10$) give relatively low remanence and high saturation magnetization values for $N_Y = 9$, 10, 11, and 16. This oscillatory dependence of the remanence and saturation magnetization on N_Y is also consistent with the proposed coupling mechanism. Superlattices for which the remanence is low and saturation magnetization high are expected to exhibit the antiphase domain structure as confirmed in the $[Gd_{10}\text{-}Y_{10}]_{225}$ sample by neutron diffraction. The calculation further predicts that a field of the order of 3 kOe would be required to bring the Gd layers of the $[Gd_{10}\text{-}Y_{10}]_{225}$ sample into parallel alignment at 0 K, which is in agreement with observation [see Fig. 2(a)].

Now that single-crystal RE superlattices can be grown with a high degree of perfection and precision, studies of fundamental phenomena including interlayer interactions and magnetic states at interfaces can be performed in a controlled and systematic manner. Novel magnetically ordered structures can be expected to occur, as has been demonstrated in the present study, and may ultimately be tailored to particular specifications.

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