Magnetic rare-earth superlattices (invited)

C. F. Majkrzak, a) Doon Gibbs, P. Böni, and Alan I. Goldman
Brookhaven National Laboratory, Upton, New York 11973
J. Kwo, M. Hong, T. C. Hsieh, R. M. Fleming, D. B. McWhan, and Y. Yafet
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
J. W. Cable
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
J. Bohr
Risø National Laboratory, DK 4000 Roskilde, Denmark
H. Grimm
IFF, Kernforschungsanlage, Jülich, West Germany
C. L. Chien
Johns Hopkins University, Baltimore, Maryland 21218

The magnetic structures of several single-crystal, magnetic rare-earth superlattice systems grown by molecular-beam epitaxy are reviewed. In particular, the results of recent neutron diffraction investigations of long-range magnetic order in Gd-Y, Dy-Y, Gd-Dy, and Ho-Y periodic superlattices are presented. In the Gd-Y system, an antiphase domain structure develops for certain Y layer spacings, whereas modified helical moment configurations are found to occur in the other systems, some of which are commensurate with the chemical superlattice wavelength. References are made to theoretical interaction mechanisms recently proposed to account for the magnetic states of these novel materials.

I. INTRODUCTION

Recent advances in molecular-beam epitaxy (MBE) techniques have made it possible to grow single-crystal, rare-earth (RE) superlattices with a high degree of perfection and limited interdiffusion.1-3 Magnetic layers consisting of a discrete number of atomic planes can be deposited alternately with nonmagnetic or other magnetic layers of a given thickness. The effects of reduced dimensionality on the magnetic ground state and corresponding critical behavior can be investigated. Because the magnetic RE moments interact through long-range, indirect exchange, a modulation of the magnetic properties in certain synthetically layered RE structures might also be expected. Furthermore, since the detailed, microscopic magnetic structures of the RE metals result from the competition between the isotropic indirect-exchange and the anisotropic crystal-field and magnetelastic interactions, the study of synthetic RE superlattices with a tailored composition and/or strain profile can, in principle, give valuable information about such phenomena as the lock-in transitions from incommensurate to commensurate phases recently described by a spin-slip model.4,5

The periodicity of the multilayer structure makes it possible to study its magnetic state by diffraction. This paper reviews research performed thus far on a number of RE superlattice systems with an emphasis on the determination of the magnetic structures. These studies have only just begun and much work remains to be done. Some of the results reported here are, therefore, of a preliminary nature. Although neutron diffraction experiments are described primarily, x-ray diffraction and magnetization measurements have been essential in achieving the present level of understanding and references to these measurements are cited at the appropriate places in the text.

II. NEUTRON DIFFRACTION BY MAGNETIC SUPERLATTICES

General discussions of the diffraction of x rays and neutrons by superlattices are given in Refs. 7, 8, and 9, respectively. Polarized neutrons are especially useful in determining the more-complicated, noncollinear magnetic structures which can occur in some of the RE superlattices. The interaction between the magnetic moment of the neutron and that of the atom depends not only on the magnitudes of the moments but also on their orientations relative to one another and the scattering vector Q, where \( Q = |k_f - k_i| \) and where \( k_f \) and \( k_i \) are the final and incident neutron wave vectors, respectively. With polarized neutrons, the squares of four structure factors can be measured.10

If the atomic moments lie in a reflecting plane normal to \( Q \) with the neutron polarization parallel to a magnetic field applied in the reflecting plane, then the structure factors are given by

\[ F(Q) = F_{\parallel}(Q) \cos \theta - F_{\perp}(Q) \sin \theta, \]

where \( F_{\parallel}(Q) \) and \( F_{\perp}(Q) \) are the parallel and perpendicular components, respectively, of the structure factor and \( \theta \) is the angle between the direction of polarization and the normal to the reflecting plane.
\[ P \pm (Q) = \sum_{j=1}^{N} (b_j \mp p_j \cos \phi_j) e^{i(Q \cdot \mathbf{r}_j)}, \]

and

\[ P \pm = \sum_{j=1}^{N} p_j \sin \phi_j e^{iQ \cdot \mathbf{r}_j}, \]

where \( b_j \) and \( p_j \) are the average coherent nuclear and magnetic scattering lengths of the atoms comprising the \( j \)th atomic plane, respectively, \( u_j \) is the position of the \( j \)th atomic plane along an axis normal to the reflecting plane, and \( \phi \) gives the orientation of the atomic moments in the plane. The magnetic scattering length \( p \) is proportional to the magnitude of the atomic magnetic moment (the magnetic form factor is included in the scattering length and is \( Q \) dependent). For the rare-earth superlattice systems described in this paper the reflecting plane is the basal plane of the hexagonal close-packed chemical structure. In this special configuration, the magnetic moment projections along the direction of the applied field produce non-spin-flip (NSF) scattering, whereas the spin-flip (SF) scattering arises from the projections of the atomic moments perpendicular to the applied-field direction.

Insofar as the magnetic structure is concerned, the unit-cell length is not necessarily the same as that of the chemical unit cell nor even commensurate with it. It is, therefore, often necessary to consider an expression for the structure factor that is a summation over \( N \times M \) atomic planes where \( N \) equals the number of atomic planes in a chemical unit cell and \( M \) equals the number of chemical unit cells or bilayers in the superlattice. Suppose, for example, that the moment arrangement in a given chemical unit cell is related to that of an adjacent bilayer by a constant phase angle \( \delta \), \( \phi_j = n\phi + m\delta \) and \( u_j = nd + m\Delta \), where \( \phi \) is a constant interplanar turn angle, \( d \) is a constant atomic interplanar spacing, \( \Delta \) is the bilayer thickness, and \( n \) and \( m \) are integers. The structure factor for spin-flip scattering can then be written as

\[ F_{SF}(Q) = \left[ 1 - e^{i(Qd + \delta)M}/(1 - e^{i(Qd + \delta)} \right] \times \left[ \frac{\sum_{n=-1}^{N} P_n e^{i(Qnd + n\phi)}}{2} \right] \left[ 1 - e^{i(Qd - \delta)M}/(1 - e^{i(Qd - \delta)} \right] \times \left[ \frac{\sum_{n=-1}^{N} P_n e^{i(Qnd - n\phi)}}{2} \right]. \]

For large \( M \), the first factor in brackets in each term of Eq. (2) approaches a delta function, peaking at values of \( Q = 2\pi n/d \pm \delta/\Delta \). Thus, satellites are displaced from positions corresponding to the chemical unit-cell period if \( \delta \) is a nonintegral multiple of \( 2\pi \). This is observed, for example, in the case of the incommensurate magnetic spiral in Dy-Y superlattices.\(^{11,12}\) The second factor in large parentheses in each term of Eq. (2) describes for relatively small \( N \) a broader function. This "envelope" function peaks at values of \( Q = 2\pi n/d \pm \delta/\Delta \) where \( j \) is an integer. The envelope in effect modulates the intensities of the relatively sharp peaks occurring at the positions \( 2\pi n/d \pm \delta/\Delta \). Upon closer examination of the envelope function, it can be seen that for \( p_n \) = const it is proportional to sin[(\( Qd \pm \phi \))/2]/sin[(\( Qd \pm \phi \))/2]. However, this simple shape can be significantly distorted (even asymmetrically about a reciprocal-lattice point\(^{13,14} \)) by any one or more of three basic modulations along the superlattice growth direction: (1) a compositional modulation giving rise to different \( p_n \), (2) a strain modulation resulting in a nonuniform \( d \) spacing, and (3) a turn-angle modulation. Although the total phase shift \( \delta \) can be determined straightforwardly from the positions of the satellites, the position of the peak of the envelope function must be deduced from an analysis of the relative intensities of the satellites. Thus, in order to extract the magnetic structure, it is necessary to know the detailed chemical modulations. These can, in principle, be determined from a combination of x-ray and neutron diffraction data taken about a number of \( (00l) \) reciprocal lattice points. The effect of \( d \)-spacing modulation, as is evident from Eq. (2), becomes more pronounced at larger values of \( Q \).

As in the case of ordinary bulk condensed-matter systems, the effects of multiple scattering and extinction on the diffraction patterns obtained for synthetic superlattices must also be properly considered. In addition, an intensity reduction and a linewidth broadening of higher-order superlattice reflections can occur depending on the nature of any distribution of bilayer thicknesses.

III. Gd-Y

A series of single-crystal \((\text{Gd}_{N_{\text{Gd}}} \cdot \text{Y}_{N_{\text{Y}}} \cdot M)\) superlattices composed of \( M \) successive bilayers of \( N_{\text{Gd}} \) basal planes of Gd followed by \( N_{\text{Y}} \) such planes of nonmagnetic Y were grown by MBE (Refs. 1 and 3) techniques. Polarized neutron-diffraction measurements\(^{15} \) were subsequently performed on a number of these superlattices with values of \( N_{\text{Gd}} = 6, 10, \) and \( 20, \) and all with a constant value of \( N_{\text{Y}} = 10 \). Below about \( 285 \text{ K}, \) the Gd atomic planes become ferromagnetically aligned within a given layer with the moments lying predominantly in the basal planes (for fields of the order of several hundred Oe applied perpendicular to \( e \)). Satellite reflections are observed in all of the samples about the \((00l)\) Bragg points at positions along the \((00l)\) direction corresponding to integer multiples of \( 2\pi/\lambda_{\text{SL}} \), where \( \lambda_{\text{SL}} \) is the bilayer thickness. Above \( T_c \), these reflections arise solely from the nuclear scattering associated with the superlattice chemical modulation, whereas below \( T_c \) an additional magnetic component contributes. To within experimental accuracy, the scattering at these satellite positions is entirely non-spin-flip. For the superlattices with \( N_{\text{Y}} = 6 \) and \( 20, \) these are, in fact, the only satellites observed and a straightforward analysis shows that the data are consistent with a simple ferromagnetic alignment of the Gd layers across the intervening Y.

The behavior of the superlattice with \( N_{\text{Y}} = 10, \) however, is markedly different. In addition to some magnetic non-spin-flip scattering at multiples of \( 2\pi/\lambda_{\text{SL}} \) about \((00l)\) below \( T_c \), another set of satellites appears at odd-integer multiples of \( 2\pi/2\lambda_{\text{SL}} \), corresponding to a doubling of the chemical bilayer periodicity, and the scattering associated with these additional satellite reflections is purely spin flip. For all of the Gd-Y superlattice samples studied by neutron diffraction (including the three described above and one for which \( N_{\text{Gd}} = 11 \) and \( N_{\text{Y}} = 9 \)), the positions of the satellites
An attempt to measure the magnetization profile as a function of temperature in a ferromagnetic [Gd$_{10}$Y$_{20}$]$_{100}$ sample by neutron diffraction was also made in order to study the effect of reduced dimensionality and interface anisotropy on the magnetic critical behavior.\textsuperscript{19} As discussed in Sec. II above, it is necessary to accurately determine the chemical compositional and strain modulation profiles before any unambiguous conclusions regarding the magnetization profile can be drawn. In this particular case the problem is complicated by a strongly temperature-dependent strain profile and more-comprehensive measurements must be performed.\textsuperscript{20}

Interlayer interaction studies of the Gd-Y superlattice system involving more-exotic layered structures, specifically arrangements where two different Y layer thicknesses corresponding to ferromagnetic and antiferromagnetic interactions between Gd layers are propagated in a Fibonacci sequence, are also in progress.\textsuperscript{21} The expectation is that something can be learned about the long-range order, or the lack thereof, in such a quasiperiodic heterostructure with competing interactions.

IV. Dy-Y

Bulk Dy displays a simple spiral structure below its Néel point (\(\approx 180\) K) with a spiral wave vector which varies continuously down to its Curie point (\(\approx 90\) K) where an abrupt transition to a ferromagnetic state occurs.\textsuperscript{22} Superlattices of Dy and Y have been extensively investigated by more than one group, and neutron diffraction has shown that the basal-plane helimagnetic ordering of the Dy moments is propagated through the intervening Y layers coherently.\textsuperscript{11,12,23-26} It has been suggested that the mechanism for the long-range interlayer coupling is a conduction-band spin-density wave in both the Y and Dy.\textsuperscript{11} An RKKY interlayer interaction coupling scheme has also been discussed by Yafet for which simplified model calculations pertaining to both Gd-Y and Dy-Y superlattices have actually been made.\textsuperscript{17,18} Although the spiral wave vector changes continuously with temperature between \(T_N\) and \(T_C\) in bulk Dy, it locks to a constant value at low temperatures in Dy-Y superlattices.\textsuperscript{12,26} A systematic treatment connecting these wave vectors to their corresponding chemical superlattice structures has not yet been given. A detailed account of much of the work which has been done on Dy-Y superlattices is given by Rhyne et al.\textsuperscript{11}

V. Ho-Y

Like Dy, bulk holmium displays a simple spiral magnetic structure below its Néel temperature (\(\approx 130\) K). Below \(\approx 70\) K additional fifth and seventh harmonics are observed, indicating a distortion of the basal-plane magnetic spiral. For temperatures \(<\approx 20\) K the magnetic structure is conical with the wave vector locked to \(\frac{1}{2}\). The existence of fifth and seventh harmonics in the \(\frac{1}{2}\) phase has been shown to originate in "bunching" of the moments about the six easy directions of the basal-plane crystal field.\textsuperscript{27} More recently, high-resolution x-ray and neutron scattering studies of the temperature dependence of the wave vector for different
bulk and thin-film holmium samples have revealed the following: (1) a sequence of lock-in transitions to commensurate wave vectors and (2) the existence of lattice modulations accompanying magnetic ordering. A spin-slip description of the magnetic structure of bulk holmium has been suggested, which extends the concept of bunching to wave vectors greater than \( \frac{1}{2} \). Neutron-diffraction measurements have confirmed the existence of higher harmonics at low temperatures in Ho-Y superlattices. Further, it has been found that the magnetic wave vector locks at low temperatures, in a manner similar to Dy-Y superlattices. Data for the total phase shift per bilayer of Ho\( _{17} \)Y, Ho\( _{15} \)Y\(_4\), and Ho\( _{10} \)Y\(_{12}\) superlattices (nominal compositions) are shown in Fig. 2. Accurate determination of the low-temperature holmium wave vectors and relative fifth to seventh harmonic intensities (which will be essential in identifying possible spin-slip structures) depend on detailed fitting of the compositional and strain modulations present in the superlattice. Work to that end is now in progress.

**VI. Gd-Dy**

Neutron-diffraction studies of a number of different Gd-Dy superlattice systems with various combinations of Gd and Dy layer thicknesses have only just begun, but the results obtained so far are very interesting. The superlattice growth direction is along the c axis as in the case of the other RE superlattices described above. Figure 3 illustrates how part of the (unpolarized) neutron-diffraction profile (Q \( \parallel \) c\(^*\)) for a Gd\(_{15}\)-Dy\(_{10}\) (nominal composition) superlattice evolves with temperature. The peak at the right-hand side of each plot, centered at about 2.21 Å\(^{-1}\), is the (002) reflection which appears as a double peak at higher temperatures because the Y (002) seed layer peak is resolved. At 300 K, only the first-order chemical modulation satellite [to the left of the (002) reflection] is observed in the range of Q included in the figure. For temperatures between approximately 200 and 130 K, a relatively weak incommensurate peak with a temperature-dependent wave vector coexists with commensurate magnetic reflections at multiples of one-half the chemical modulation wave vector (indicative of a doubled magnetic unit cell). At the onset of long-range magnetic order, the wave vector of the incommensurate phase is that which would be expected for a "superspiral" consisting of ferromagnetic Gd layers coherent with helimagnetic Dy layers possessing an interplanar turn angle equivalent to that found in the bulk. Below about 130 K, the magnetic structure appears to be entirely commensurate. Four other Gd-Dy superlattices with different layer thicknesses are also found to have diffraction profiles which indicate predominantly commensurate magnetic structures with a magnetic unit cell that is either the same as or double that of the chemical cell. Magnetization measurements performed at a similar applied field display a pronounced temperature dependence with minima in the magnetization showing some correspondence to neutron-diffraction profiles characteristic of a doubled magnetic unit cell.

Polarized neutron-diffraction profiles for Gd\(_{15}\)-Dy\(_{10}\) about (002) in a field of about 100 Oe are shown in Figs. 4(b), 4(c), and 4(d) at temperatures of 80 and 120 K. Q is again parallel to the c axis and the magnetic field is applied perpendicular to both the [001] and [h 00] directions which...
define the scattering plane. Although the profiles plotted in Fig. 4(a), which are equivalent to what would be observed for an unpolarized beam, are qualitatively similar for the two temperatures, the individual spin-flip [Fig. 4(d)] and non-spin-flip [Figs. 4(b) and 4(c)] cross sections are clearly different and indicate that a significant change in the magnetic structure has occurred. Consider now what the magnetic structure at a given temperature, say 80 K, might be. First of all, we know that the magnetic structure has long-range order from the narrow widths of the satellite reflections and that the magnetic unit cell is twice that of the chemical unit cell. Furthermore, the polarized beam data tells us that not only does the configuration of spin components normal to the applied field (spin-flip scattering) have a doubled unit cell, but so does the arrangement of moment projections parallel to the field (non-spin-flip scattering). These qualitative observations form constraints for any moment model which might be considered. Assuming that the Gd layers consist of ferromagnetically aligned planes and the Dy layers of ferromagnetic planes arranged in some modified helical array, a systematic variation of several parameters (such as Dy moment turn angle and Gd layer moment orientation) was performed in calculating the model structure factors. For the range of parameter variations considered, the configuration found to give the best statistical fit (minimum $\chi^2$) is depicted in Fig. 5(c) along with some of the other RE superlattice magnetic structures discussed above. The moments of adjacent ferromagnetic layers of Gd are approximately at right angles to one another with one layer aligned along the direction of the applied field (two possible “domain” configurations are possible; 90° clockwise or counterclockwise from the field direction). The Dy moments “fan” out (as do the moments in bulk Dy in a sufficiently large applied field) from more nearly ferromagnetic alignment with the Gd layers at the interface to a maximum interplanar turn angle at the center of the Dy layer which is close to that observed in the bulk. The moments of the Gd and Dy were found to be $g_{\mu_B}$ and $g_{\mu_B}$, respectively, with $c$-axis projections corresponding to 40° from the $c$ axis in the Gd to 90° at the center of the Dy. A $c$-axis component (which is present in bulk Gd) was included in the model because the satellite profile about (102), which is sensitive to moments out of the basal plane, was found to be dissimilar from that about (002) [a disordered in-plane component modulated along the $c$ axis could, however, have the same effect]. The observed intensities (accurate to about 10% or better) were measured at 80 K and at 150 Oe after cycling down from approximately 8 kOe. The calculated and observed intensities are given in Table I. In performing the model calculations, it was assumed that the chemical modulations closely resemble those observed for the Gd-Y system. In the final analysis it will of course be necessary to determine the actual atomic plane spacing and compositional modulations for the Gd-Dy superlattices. In measuring the field dependence at 80 K, it was found that a field of about 6 kOe was required to completely align all Gd and Dy moments along the field direction (again perpendicular to the [001] and [h 00] direc-
TABLE I. Comparison between observed (Obs.) structure factors and those calculated (Calc.) for the model described in the text. $H = 150$ Oe and $T = 80$ K.

| $Q$ (Å$^{-1}$) | $|F^{+}\rangle^2$ | $|F^{-}\rangle^2$ | $|F^{0}\rangle^2$ |
|--------------|-----------------|-----------------|-----------------|
| 2.040        | 0.027 0.052     | 0.017 0.025     | 0.021 0.005     |
| 2.122        | 0.085 0.092     | 0.087 0.092     | 0.051 0.065     |
| 2.200        | 1.000 1.000     | 0.247 0.168     | 0.124 0.137     |
| 2.286        | 0.059 0.067     | 0.056 0.067     | 0.032 0.047     |
| 2.366        | 0.044 0.086     | 0.013 0.027     | 0.021 0.010     |

ACKNOWLEDGMENTS

We would like to acknowledge valuable discussions with R. W. Erwin, C. P. Flynn, J. J. Rhyne, M. B. Salamon, and other workers in this field. Work at Brookhaven National Laboratory and Oak Ridge National Laboratory was supported by the U.S. Department of Energy, Contract Nos. DE-AC02-76CH00016 and DEAC05-840421400, respectively.


20. C. F. Majkrzak et al. (unpublished).


29. C. F. Majkrzak et al. (unpublished).


