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## MAGNETIC SUPERLATTICES

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**Abstract.** – Single crystal magnetic rare earth superlattices were synthesized by molecular beam epitaxy. The studies include four rare earth systems: Gd-Y, Dy-Y, Ho-Y, and Gd-Dy. The magnetic properties and the long-range spin order are reviewed in terms of the interfacial behavior, and the interlayer exchange coupling across Y medium.

### 1. Introduction

Significant progress has been made in the past decade in the studies of metallic multilayers due to the advance of thin film deposition techniques [1]. The original concept of "artificial superlattice" suggested by Esaki and Tsu for III-V semiconductors was extended to magnetic multilayers which consist of alternating magnetic layer with another nonmagnetic layer along the growth direction [2]. By varying the thickness of the magnetic region to the nonmagnetic region, magnetic superlattices can be used as a model system to investigate the interfacial magnetic behavior, two dimensional magnetism, and interlayer exchange coupling effects. The first two aspects have been addressed in the previous studies on Cu-Ni superlattices by Gyorgy *et al.* [3], and on MnSb/Sb superlattices by Shinjo *et al.* [4].

One important goal of our studies is the modulation effect on the magnetic properties of the superlattice as a whole due to the long-range interlayer exchange coupling. The magnetic rare earth system was chosen because the magnetic rare earth moments are well localized. The indirect exchange or the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction is *via* the conduction electrons, and thus of longer range. It is highly probable that an effective magnetic coupling between separate magnetic layers can be formed across the intervening nonmagnetic medium. Furthermore, elemental magnetic rare earths exhibit long-range magnetic order modulated along the *c*-axis on a length scale compatible with typical superlattice wavelengths [5]. This raises interesting possibilities of tailoring the natural spin structures with the artificially imposed periodicities.

This article reviews the structural and magnetic properties of the single-crystal, magnetic rare earth superlattices. The content includes the molecular beam epitaxy (MBE) growth, structural characterization, magnetic properties and magnetically ordered spin structures of Gd-Y, Dy-Y, Ho-Y and Gd-Dy superlattices, and theoretical basis for the magnetic exchange coupling in the superlattices. Most of the results presented here are of a "summary" nature, and the references to the previous publications of more complete studies are cited in the text below.

### 2. Superlattice growth

All the samples were prepared by the metal molecular beam epitaxy technique using a combination of electron beam and effusion cell evaporation [6]. Direct depositions of rare earths onto Si, sapphire, or other commercially available substrates led to severe chemical reactions between the rare-earth films and the substrates. The chemical interactions was eliminated by intervening a single crystal buffer layer. A Nb(100) single crystalline film epitaxially grown on Al<sub>2</sub>O<sub>3</sub> (11 $\bar{2}$ 0) was found to be most suitable for this purpose [7]. The growth sequence of the superlattice is shown schematically in figure 1. The in-plane orientation between hcp rare earth (0001) and bcc Nb (110) follows the Nishiyama-Wasserman relationship; namely, [11 $\bar{2}$ 0]Y // [002]Nb, and [1010]Y // [1 $\bar{1}$ 0]Nb [8]. After the initial deposition of a seed layer (Y) at 550 °C, the superlattices were grown at a relatively low temperature of 250 °C, to minimize the inter-diffusion between the rare earths such as Gd and Y. *In situ* reflection high energy electron diffraction (RHEED) patterns exhibit

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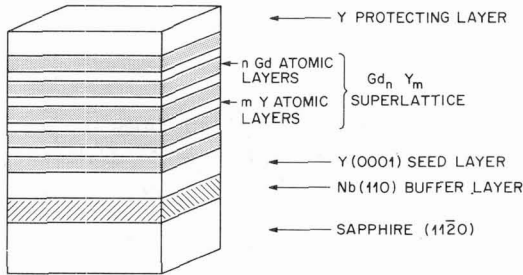


Fig. 1. - Schematics of a typical  $Gd_n - Y_m$  superlattice deposition sequence with a Nb(110) buffer layer grown on  $Al_2O_3$  (1120).

sharp streaks with distinct Kikuchi arcs, suggesting a layer-by-layer growth and a highly ordered, atomically smooth surface.

### 3. Structural analysis by X-ray diffraction

The overall structural properties were characterized by high-resolution X-ray diffraction from a Cu rotating anode X-ray source [9]. The  $[Gd_{N_{Gd}} - Y_{N_Y}]_m$  superlattice contains hcp  $N_{Gd}$  atomic planes of Gd and  $N_Y$  such planes of Y in each bilayer period that is repeated  $m$  times. The superlattices are three-dimensional hcp single crystals. The full widths at half maximum (FWHM) of the rocking curves are  $0.16^\circ$  and  $0.23^\circ$  for directions perpendicular and parallel to the plane, respectively. The coherence length along the growth direction is essentially the total film thickness, and the in-plane domains are ordered on a length scale of about 2000-3000 Å.

A quantitative analysis of the diffraction intensities indicates that the chemical interfacial widths are about 2-atomic layer thick where the compositional profiles vary from 90 % to 10 %. Due to the magnetostrictive effect in the ferromagnetic Gd, the amplitude and the interfacial width of the interplanar spacing (strain) modulation increase continuously at temperature below  $T_C$ . The strain modulation profile for a (21, 21) superlattice at  $T = 12$  K and 333 K are plotted in figure 2a.

### 4. Magnetic properties of Gd-Y superlattices

**4.1 INTERFACIAL MAGNETISM.** - Magnetic X-ray scattering experiment using a synchrotron radiation source was performed on the Gd-Y superlattices to determine the magnetic moment modulation in the superlattices [10]. Three possible models of the magnetic moment modulation profile for the (21, 21) superlattice are plotted in figure 2b. The solid curve, in that the full Gd moment is maintained at the center of the Gd array and the moment is smoothly reduced in approaching the interfaces, gives the best agreement with the measurements.

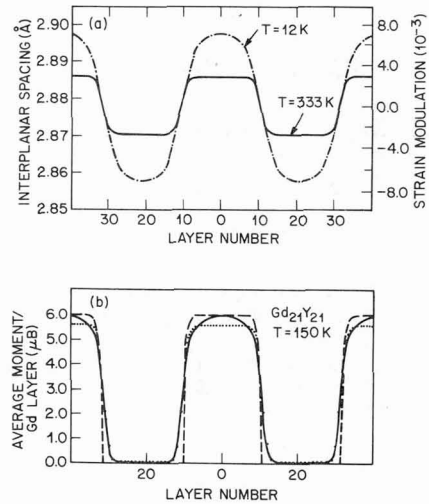


Fig. 2. - (a) The interplanar spacing modulation along the  $c$  axis at 15 K and 333 K for a (21, 21) superlattice. (b) Three models of the magnetic moment distribution at 150 K for the same sample.

The reduction of Gd magnetic moment near interface was also observed in the magnetization measurement using vibrating sample magnetometer (VSM), and the Faraday force method [9]. In the study of a series of superlattices with decreasing  $N_{Gd}$ , hence with increasing contributions of the interfaces, the magnetization property of the Gd array can be accounted for by a simple two-layer model: the central ferromagnetic Gd region of  $(N_{Gd} - 2)$  atomic layer thick retains the ideal magnetic thin film behavior even for  $N_{Gd}$  as small as 5. The two interfacial Gd atomic planes do not order ferromagnetically. The loss of the ferromagnetic order in the interfacial region has to do with the finite interfacial widths of the chemical and the strain modulations. The Gd moments might arrange in a disordered manner in the interfacial plane, which leads to a net cancellation of the overall moment of the interfacial layer.

**4.2 INTERLAYER EXCHANGE COUPLING EFFECTS.** - Superlattices provide a convenient means for studying such effect by choosing samples of the same  $N_{Gd}$ , but with varying  $N_Y$ . The dependence on  $N_Y$  of two characteristic magnetization parameters, the ratio of  $\sigma_r/\sigma(0)$ , and the saturation field  $H_S$  are shown in figure 3 for two series superlattices:  $N_{Gd} = 4$ , and 10 [11]. Here we define  $\sigma_r$  as the remanence,  $\sigma(0)$  as the spontaneous ferromagnetic moment, and the  $H_S$  as the saturation field at which  $\sigma$  no longer exhibits a rapid rise with field, and  $\Delta\sigma//\Delta H$  becomes constant. The data show a continuous oscillatory dependence on  $N_Y$  with a periodicity of approximately 7 atomic layers, extending over a range of 20 atomic planes.

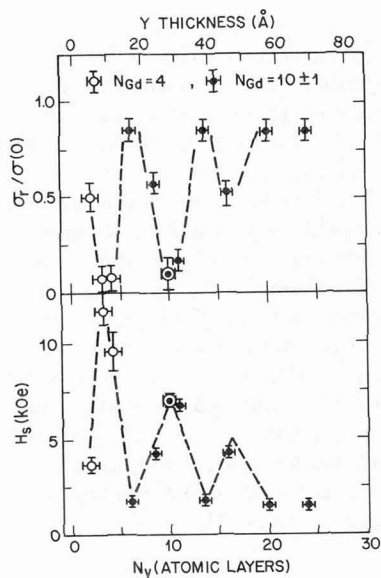


Fig. 3. - The oscillatory dependence of magnetization parameters of (a)  $\sigma_r / \sigma(0)$ , and (b)  $H_S$  on  $N_Y$  for two series of superlattices with  $N_{Nd} = 4$ , and 10.

Polarized neutron diffraction studies were performed at Brookhaven National Laboratory by Majkrzak *et al.*, and led to a clear picture of the magnetically ordered spin structures [12]. The overall magnetic order of the Gd-Y superlattices oscillates in two distinctly different states depending on the exact  $N_Y$ . One magnetic order is the simple ferromagnetic order where the moments in the plane of all Gd arrays are in parallel alignment, thus giving rise to a high  $\sigma_r$ , and a low  $H_S$ . The other magnetic order is the antiferromagnetic order, or so-called antiphase domain structure [12]. In zero field, the adjacent Gd arrays are  $180^\circ$  antiparallel to one another, hence a small  $\sigma_r$ . A field ( $H_S$ ) of about 6 kOe at 77 K is needed to fully align the spins with respect to the field. Figure 4a shows this antiferromagnetic spin structure at  $H = 150$  Oe, and  $T = 150$  K. The theoretical interpretation of the long-range coherent magnetic order in Gd-Y will be given in section 7.

##### 5. Magnetic properties of epitaxial Dy, Ho films, and Dy-Y, Ho-Y superlattices

Dy exhibits an incommensurate in-plane helical order at a  $T_N$  of 178 K. Due to a competition among the magnetic exchange coupling, magnetostriction, and the crystal field effect, the turn angle between successive spirals decreases continuously from  $43.2^\circ$  at  $T_N$  to  $26.5^\circ$  at  $T = 90$  K before collapsing to the ferromagnetic state through a first order transition. In addition to the similar helimagnetic order, bulk Ho also exhibits lock-in transitions at low temperature described recently by a spin-slip model [13].

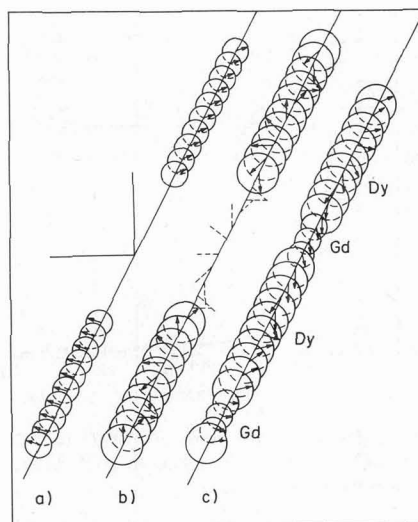


Fig. 4. - Schematic representation of the basal-plane component directions in three rare earth superlattices (a) Gd-Y canted antiphase domain structure (b) Dy-Y coherent incommensurate spiral, and (c) Gd - Dy asymmetric state.

In Dy-Y superlattices, possible modifications of the Dy helical order are discussed in terms of three aspects: phase, chirality, and frequency. Extensive studies by University of Illinois at Urbana and National Bureau of Standards showed that both the phase and chirality maintain their coherency in the Dy-Y superlattices [14-16]. Our studies on Dy-Y samples with varying  $N_Y$  agrees, in general, with their findings that the phase coherency and chirality are preserved so long as the  $N_Y$  is less than 20 [17]. This incommensurate coherent spiral structure is shown in figure 4b.

The Dy spiral wavevector in Dy-Y superlattices shows a much weaker temperature dependence than that in bulk Dy [18]. There is no evidence of the first-order ferromagnetic transition. In addition, the wavevector locks to a constant value over a temperature range  $\sim 50-90$  K. The wavevector of the Ho helix in Ho-Y superlattices also shows a modified temperature dependence with a trend similar to Dy-Y superlattices [19]. This behavior was explained by the lattice-clamping effect due to the epitaxial growth of the Dy or Ho with the non-magnetostrictive Y layer. The modified magnetostriction of Dy or Ho was not only observed in the Dy-Y or Ho-Y superlattices, but was also found in the epitaxial, single-layer Dy or Ho films grown on Y substrates. Magnetization measurements on the epitaxial Dy films show the systematic decrease of the ferromagnetic transition temperature with decreasing thickness. The data are plotted in figure 5 for  $H = 2.6$  kOe. These results indicate that the microscopic spin configuration of the magnetic rare earths can, in principle, be tailored by the coherency strain profile.

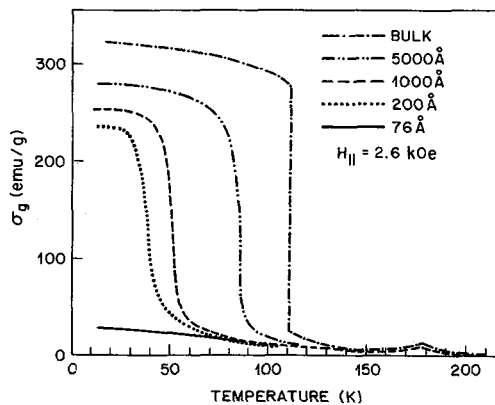


Fig. 5. - Moment vs. temperature at  $H = 2.6$  kOe for single layers of Dy films of 76 Å, 200 Å, 1000 Å, and 5000 Å thick, and a bulk Dy crystal.

## 6. Magnetic properties of Gd-Dy superlattices

The ferromagnetic/helimagnetic superlattices show rich magnetically-ordered spin states at low field and low temperature. Representative magnetic moment vs. temperature for a  $Gd_5-Dy_{10}$  superlattice is shown in figure 6 at a field of 100 Oe [20]. The Gd layers with  $N_{Gd} = 5$  are ferromagnetically ordered below a Curie temperature of 247 K. At temperatures below 200 K, the Dy layers develop a helical order with an interplanar turn angle similar to that seen in bulk Dy. The Gd spins couple coherently with the Dy spins at Gd-Dy interfaces through the interfacial exchange. The magnetic order of the bilayer follows an incommensurate super-spiral structure which yields a reduction of moment at  $130 \text{ K} < T < 200 \text{ K}$  [19].

As the Dy spiral turn angles decrease continuously with the lowering temperature, the wavevector of the super-spiral of the bilayer also decreases, eventually locks to a value to be commensurate with the bilayer

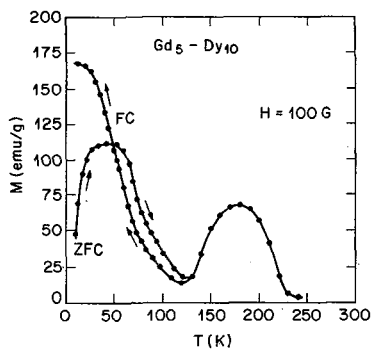


Fig. 6. - Moment vs. temperature at  $H = 100$  Oe for a  $Gd_5-Dy_{10}$  superlattice in zero-field cooling (ZFC), and field cooling (FC) cycles.

periodicity at  $T = 130 \text{ K}$ . The actual unit cell of the magnetic order is twice the unit cell of the chemical modulation. Polarized neutron diffraction studies by Majkrzak *et al.* showed that the magnetic order at 80 K and a field of 150 Oe is described by a so-called commensurate asymmetric state depicted in figure 4c [19]. The moments of adjacent ferromagnetically aligned Gd arrays are about at right angles to one another with one array aligned along with the applied field. The Dy moments fan out from nearly ferromagnetic alignment with the Gd moments at the interface to a maximum interplanar turn angle at the center of the Dy array which is close to the bulk Dy value. A somewhat similar spin-ordered state at low fields, the so-called unsymmetric state, was predicted in the recent calculation by Hinchev and Mills for superlattices containing alternating collinear ferromagnetic and antiferromagnetic layers [21].

## 7. Interlayer exchange coupling in Gd-Y and Dy-Y superlattices

The long-range, coherent magnetic spin structures observed in Gd-Y and Dy-Y superlattices were accounted for on the basis of the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction between magnetic rare earth layers intervened by the non-magnetic yttrium. The conduction electrons of the Y layer respond to the local exchange fields penetrating from the neighboring Gd (Dy) layers and propagate the exchange coupling indirectly in a manner described by the generalized susceptibility  $\chi_Y(q)$ . A semi-phenomenological calculation of the exchange function  $J_{Gd-Y}(r)$  was made by Yafet *et al.* [22] for a sandwich structure consisting of two monoatomic Gd planes separated by a Y medium. The exchange function  $J_{Gd-Y}(r)$  shows an oscillatory dependence on the  $N_Y$  with a periodicity corresponding to a wavevector of  $q_{max} = 0.28 \times 2\pi/c$ , where  $\chi_Y(q)$  exhibits a maximum at  $q_{max}$  [23]. Both the sign and the magnitude of  $J_{Gd-Y}(r)$  are in good agreement with the experimental observation. Note that the Gd arrays contain only collinear spin components. If the interactions beyond the nearest neighbor coupling are ignored, the coupling through the intervening Y can only lead to parallel or antiparallel ordering in Gd-Y superlattices.

Calculations were extended to Dy-Y sandwich structures where Dy layers are two-atomic plane thick [24]. It was shown that the turn angle between the successive Dy spirals separated by  $N_Y$  atomic layers of Y increases by  $360^\circ$  in approximately 7 atomic layers of Y, which corresponds to the value of  $q_{max}$  of  $0.28 \times 2\pi/c$ . Hence the mechanism based on the RKKY exchange coupling across Y produces the coherence in the phase and chirality of the Dy helical order as observed experimentally.

## 8. Conclusions

With the development of metal molecular beam epitaxy, highly perfect magnetic rare earth superlattices have been prepared with minimal interdiffusion. Modulation of the overall magnetic properties of the superlattices was demonstrated for the first time, along with the discovery of long range, coherent magnetic order that can be tailored with superlattice wavelength. The approach has now opened up a wide spectrum of exciting opportunities in the studies of thin film magnetism, which may eventually lead to novel device applications in the future.

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