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New configurations of rare-earth superlattices

R. Du, F. Tsui, and C. P. Flynn
Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801
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We have successfully grown high-quality Dy/Y rare-earth superlattices with their *a* or *b* axes perpendicular to the growth plane, at choice. Earlier efforts by molecular-beam-epitaxy methods produced only growth along the *c* axis. In other research, we have grown almost freestanding superlattices of Dy/Y and other hexagonal rare-earth superlattices. These new configurations make a variety of significant new experiments accessible.

Interesting magnetic phenomena are revealed by recent studies of magnetism in rare-earth superlattices and films. In earlier work with collaborators, we have examined the helical antiferromagnetism of Dy multilayers separated by nonmagnetic Y.[1] An unexpected result is that the Dy antiferromagnetism is transmitted *coherently* through the Y so that even the chirality of the helical order is preserved.[2] We observe analogous transmission for the *c*-axis-modulated Er structure in superlattices with Y.[3] These effects appear to be consequences of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction through the Y, linking spins in neighboring magnetic layers.[2,4] All the materials in this work have employed molecular-beam-epitaxy (MBE) growth of the superlattices, using Nb[110] grown on sapphire [11$\bar{2}$0] as a substrate for an initial Y buffer layer grown on its basal plane, preceding the actual hcp superlattice. A sketch of this arrangement is given in Fig. 1(a). This is the prescription worked out earlier in joint Bell Laboratories–University of Illinois research.[5] Since then, the Bell Labs workers have made a number of elegant studies mainly of superlattices with ferromagnetic Gd.[6] Our own research has focused largely on the interesting properties of superlattices containing the antiferromagnetic rare earths.

In both thin films and superlattices, the properties of Dy are modified. Specifically, the Néel temperature is lowered, the ferromagnetic transition in the thin epitaxial film at low temperatures is completely suppressed, and the helical period is changed to a different function of temperature. Our estimates of the magnetoelastic energy indicate that these are mainly the consequences of lattice "clamping" by the substrate, which modifies the change of available magnetoelastic energy in the transition.[1,2] In all superlattices grown to date, it is the basal plane that is clamped to the substrate. We recognize here that the anisotropy, both of the clamping and the RKKY interaction, makes the magnetic behavior of other film orientations particularly interesting. For this reason we have explored means to prepare superlattices that lack the clamping of a bulk substrate, and superlattices that are clamped on differently oriented planes. The new configurations that have been grown are discussed in what follows.

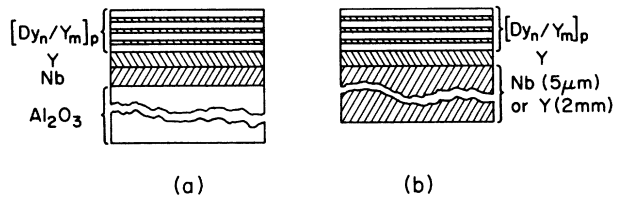

FIG. 1. In the conventional method, Nb[110] is grown on sapphire [11$\bar{2}$0], followed by Y[0001] and then the superlattice, as shown in (a). In the present work, Y is grown directly on a metal single crystal, as shown in (b). The crystals employed as substrates are either [110] Nb 5 μm thick or Y crystals 2 mm thick, with their *a* or *b* axes perpendicular to the growth plane.

To prepare rare-earth superlattices on thin substrates, we employ Nb foil grown as a [110]-oriented single crystal 5 μm thick by about 1 cm^2. The Nb crystal was first fabricated by an extension to thinner films of methods developed by Birnbaum and co-workers.[7] Our procedures involve cyclic heat treatment of an initially polycrystalline [111] textured foil at temperatures up to 2400 K, whereupon the desired structure emerges spontaneously. Earlier use of thin Nb single crystals in surface science applications has been reported by Strongin and co-workers.[8]

Superlattice preparation directly on the Nb foil [Fig. 1(b)] proceeded much as for conventional substrates except that care was necessary to avoid damage to the fragile foil. The [110] surface was first improved by added Nb followed by growth [0001] Y about 500 Å thick, and then the superlattice. Particular attention to thermometry was also required because the substrates could not be brought into thermal equilibrium by conduction from a base plate, owing to their geometry. Appropriate housing was therefore needed, and the foil temperature was measured directly at the growth temperature to ensure that correct growth condition were employed.

Figure 2 shows a Bragg scan on a Dy/Y superlattice with 64 periods each containing 12 Dy layers and 24 Y layers. The rocking curves of the Nb substrate and the Dy/Y superlattice were both about 1° wide. We believe that a significant part of this was due to a slight wrinkling that remained in the 5-μm foil after annealing, and that the microscopic crystal is more coherent than the width indicates. This belief is reinforced by the measured peak widths in the Bragg scan (Fig. 2), which are ~0.1° for Nb and 0.15° for the Dy/Y superlattice. The superlattices grown on foils are evidently single crystals, and not greatly inferior to those normally obtained on sapphire

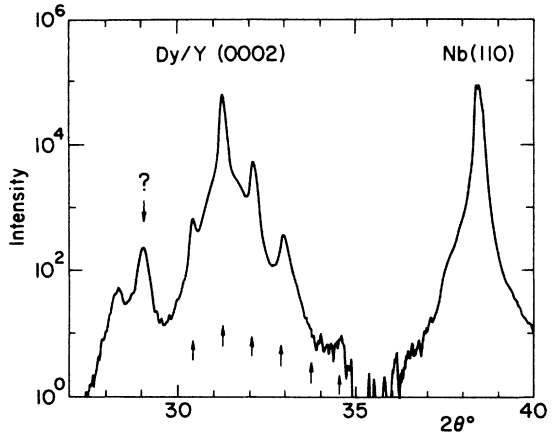

FIG. 2. Bragg scan of a $(Dy_{12}/Y_{24})_{64}$ superlattice grown along the c axis on a [110] Nb single crystal 5 μm thick. The Nb[110] peak near $2\theta = 38°$ is narrow enough that the Cu K α_1-α_2 splitting is resolved. Structure seen below $2\theta = 30°$ is of unidentified origin, possibly from differently oriented grains at an abundance of a few parts in 10^3.

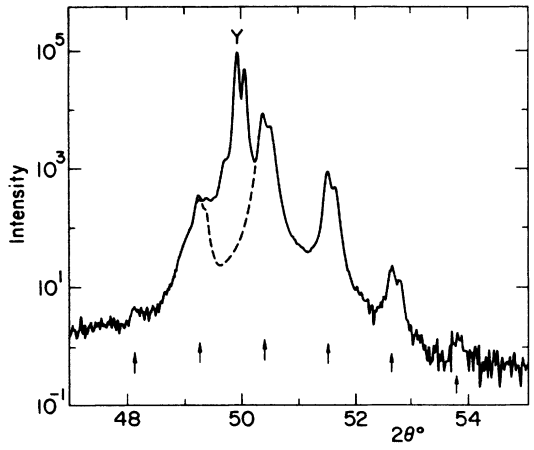

FIG. 3. Bragg scan of Dy/Y superlattice grown along its a axis on a Y single crystal 2-mm thick. The broken line indicates schematically the removal of the Y substrate peak. Note that the crystal quality is sufficient that several orders of superlattice sidebands exhibit α_1-α_2 splittings of the Cu K line.

substrates via Nb and Y. We have been equally successful with superlattices containing Er and Ho in place of Dy. One may reasonably expect that these new techniques will improve with practice and that materials of still higher quality will result.

We have, in addition, grown excellent Dy/Y superlattices along the hexagonal a and b axes using Y bulk single crystals as the growth template. The first metal superlattices grown with choice of orientation were of Nb/Ta, with [100], [110], [111], and [112] growth directions selected by choice of oriented Al_2O_3 or MgO substrates, and with comparably high quality achieved for all orientations.[9] The procedures we devised for the Nb/Ta system have later been confirmed by the Naval Research Laboratory (NRL) group,[10] but no similar flexibility has been reported for other metals. In the case of hexagonal rare-earth metals, suitable substrates for other selected orientations may possibly exist, but we have not yet found any, probably owing to the strong tendency for c-axis growth. To solve these problems, we have employed substrates of bulk single crystals cut with the desired orientation and subsequently polished and etched to a high level of surface perfection. Crystals about 2-mm thick and 1 cm^2, prepared in this way with a or c axes perpendicular to the surface, were purchased from the crystal preparation group working under B. Beaudry at the Ames Laboratory.

We chose to handle the crystals before growth using normal surface science methods. Surfaces were prepared for use by Ar-ion sputtering at room temperature followed by high-temperature anneals, all *in situ*. Y deposition at the growth temperature then preceded the growth of the actual superlattice. A variety of temperature-dependent surface reconstructions were observed by reflection high-energy electron diffraction (RHEED) before and during Y growth; these will be reported elsewhere. The most important discovery is that the a and b faces are badly reconstructed and fairly rough during growth at temperatures $\sim$400 °C suitable for the preparation of samples with minimal interdiffusion. Apparently, however, each layer must transform to the bulk crystal structure as it is buried, because the x-ray characteristics of the films are very satisfactory (as good, in fact, as the bulk crystal as purchased). Thus, both a- and b-oriented materials could in fact be grown to desired specifications.

A Bragg scan from an a oriented Dy/Y superlattice grown on a bulk Y single crystal is shown in Fig. 3. Films of generally similar quality could also be grown in the b orientation. The sample in Fig. 3 was grown with 40 layers each containing 43 Å of Dy and 44 Å of Y. The crystal quality is very good with the α_1-α_2 splitting of the Cu K line clearly resolved on the fundamental, and several orders of sidebands, with widths $< 0.1°$. The superlattice and bulk rocking curves are $\sim 0.5°$ wide. It was necessary to grow the a- and b-oriented films some 50 °C below the temperature for growth on the basal plane, in approximate consistency with the known anisotropy of the diffusion coefficient of Y.[11]

In summary, we have reported the successful growth of rare-earth superlattices both as almost freestanding films with the basal plane on 5-μm-thick [110] Nb single crystals, and as a- and b-oriented single crystals on bulk Y. In addition to their self-evident use in electron microscopy, specific heat, and various other measurements, we expect that these new structures can be used in future work to investigate the effects of substrate clamping and electronic anisotropy on the cooperative magnetic behavior exhibited by these interesting systems.

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