Enhanced Curie Temperatures and Magnetoelastic Domains in Dy/Lu Superlattices and Films

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We have grown high quality superlattices of Dy with nonmagnetic Lu and find that the 2.4% epitaxial compression nearly doubles the ferromagnetic T_C of Dy with little change in the Néel temperature. A helimagnetic phase exists over a narrow temperature range. Below T_C in superlattices, 300 Å orthorhombic domains form despite epitaxial constraints, each with a magnetostrictive distortion comparable to that of bulk Dy. For the thinnest intervening Lu layers, individual ferromagnetic Dy blocks have parallel alignment; the remaining samples show antiparallel alignment, coherent over many bilayer periods.

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A new regime in the study of rare-earth magnetism began with the discovery [1,2] that 3D, long-range order can occur in rare-earth/yttrium superlattice (SL) structures. Two important effects were found in the Dy/Y system: (i) Helimagnetic order propagates through nonmagnetic Y layers as thick as 100 Å and (ii) coherency strain and clamping to the substrate caused by the epitaxial constraint completely suppress the low-temperature ferromagnetic phase of dysprosium. In this Letter, we report significant new effects in the first Dy/Lu superlattices and films ever prepared. In the present case, the 2.4% epitaxial compression of the Dy lattice (opposed to the 1.6% expansion in the Dy/Y case) *enhances* the ferromagnetic transition temperature by nearly 100%. An enhancement [3] of 25% was found previously for Dy grown on Er (1% compression). Despite epitaxial constraints in the Dy/Lu samples, the onset of ferromagnetic order is accompanied by the appearance of microscopic, orthorhombically distorted domains. Such distortion necessarily induces long-range coherence of the easy axis along the growth direction. Our further results indicate that this mechanism, aided by local dipolar coupling, results in antiparallel stacking of the individual Dy ferromagnetic blocks for thick Lu interlayers.

Samples were grown by molecular beam epitaxy using methods described elsewhere [4]. An hcp Lu-(0001) buffer layer was grown on (110) Nb, followed by the growth of either Dy films ranging in thickness from 40 to 400 Å, or of Dy/Lu superlattices. In the superlattices, the Dy layer thickness was maintained at approximately 15 atomic planes (40 Å) per layer while the Lu layers were varied between 5 and 29 atomic planes (15–80 Å). Room-temperature x-ray diffraction confirmed good crystal quality, with coherence lengths of 700 Å along the c axis (growth direction) and mosaic widths $<0.3°$. Analysis of SL harmonics around the (00·2) reflection shows that interdiffusion involves at most 3–4 atomic layers at the interfaces.

In Fig. 1 we show both field-cooled (FC) and zero-field-cooled (ZFC) easy-axis magnetization data at 200 Oe and 1 kOe for a SL comprised of 60 bilayers, denoted $[Dy_{16}|Lu_{20}]_{60}$. Each period contains 16 atomic planes of Dy and 20 of Lu. Shown for comparison is the magnetization of bulk Dy, measured [5] at 1 kOe. Not discernible on this scale is a small cusp in the bulk data at 178 K that marks the onset of helimagnetic order. At this temperature the spins in each (00·1) plane order ferromagnetically in the plane, with successive planes rotated by an average turn angle ω to form a spiral. The first-order ferromagnetic transition at $T_C=85$ K is evident and is accompanied, in bulk Dy, by a 0.5% orthorhombic distortion of the basal plane [6]. A small feature near 178 K is also present in the superlattice data, but a gradual increase in magnetization sets in 60–70 K above the bulk T_C upon field cooling. Even at 10 K, a field in excess of 3 kOe is required to saturate the SL moment, in contrast to the $\simeq 1$ kOe anisotropy field [6] for bulk Dy. The ZFC magnetization is nearly zero at low temperatures, joining the FC curves at temperatures > 100 K.

In order to determine whether the magnetization data

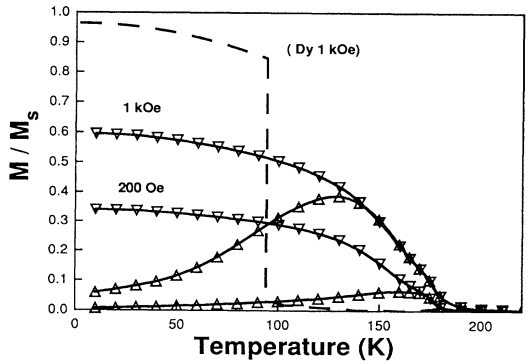

FIG. 1. Magnetization vs temperature for SL $[Dy_{16}|Lu_{20}]_{60}$ in fields of 200 and 1000 Oe alongside that of elemental Dy (1000 Oe). FC (grads) and ZFC (triangles) data are shown. The data are normalized to the saturation magnetization.

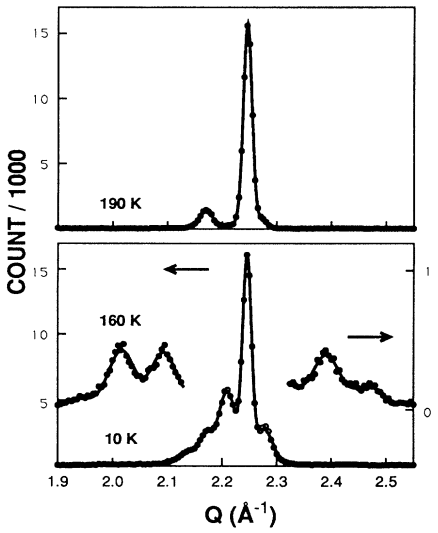

FIG. 2. Scattered neutron intensity about the (00·2) reflection in scans along c^* (growth direction) from SL $[Dy_{15}|Lu_{15}]_{40}$ at temperatures of 190 K (paramagnetic), 160 K (helimagnetic), and 10 K (antialigned ferromagnetic layers). Note the scale change for the 160 K data.

result from domain formation or antiparallel stacking of magnetized Dy blocks, we studied several SL's by neutron diffraction. Figure 2 shows the combined structural and magnetic scattered intensity for $[Dy_{15}|Lu_{15}]_{40}$ in scans along the c^* axis (growth direction). These data were obtained at the NIST reactor on a triple-axis spectrometer, with the analyzer tuned for zero energy transfer in order to reduce the inelastic background. At 190 K, only the (00·2) nuclear Bragg peak at $Q=2.25$ Å^{-1} is observed, along with its first-order SL satellite $2\pi/\Lambda$ lower at $Q=2.17$ Å^{-1}. The SL period is thus $\Lambda=80$ Å. The data at 160 K, shown on an expanded scale, display the characteristic multipeak structure of helical long-range order in a superlattice [1]. The centroids (first moments) of the magnetic peaks are located at 2.05 and 2.39 Å^{-1}. The widths of the individual peaks are all 0.052 Å^{-1}, after correction for instrumental resolution, corresponding to a magnetic coherence length of 1.5 bilayers. This is significantly shorter than the coherence observed in Dy/Y SL's of comparable composition [7]. Following earlier modeling [1], we find a turn angle of $28° \pm 1°$ per Dy plane at 160 K, as compared with 40° at the same temperature in elemental Dy [6]. The magnetic phase advance across each Lu layer is $45° \pm 2°$ (modulo $2\pi/N_{Lu}$) on average. This value agrees approximately with the calculated [8] maximum in the generalized susceptibility of Lu at 48°/plane.

The qualitatively different behavior of the 10 K data, shown at the bottom of Fig. 2, reveals greater coherence than that of the helimagnetic phase at 160 K, and is unlike anything observed in Dy/Y samples. The magnetic peaks at Q values almost exactly between the structural

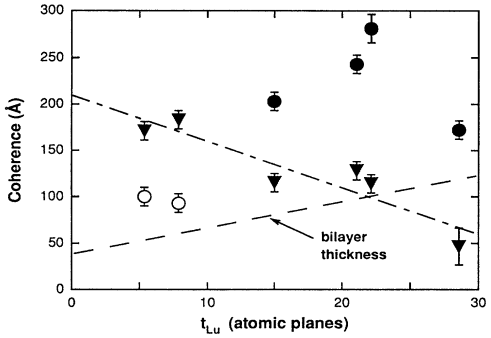

FIG. 3. Magnetic coherence length vs number of Lu interlayers for spiral (triangles), aligned ferromagnetic layers (open circles), and antialigned layers (filled circles). The actual spacing is 2.77 Å/Lu layer.

superlattice peaks correspond to a doubled SL unit cell. This structure appears at temperatures as high as 160 K in some samples, and coexists with the helimagnetic phase over a range $\simeq 30$ K. In the coexistence regime, the turn angle is approximately 25°/Dy layer, comparable to the 26.5°/layer at the T_C of bulk Dy [9]. Two-phase behavior of this type, induced by clamping of magnetoelastic strain, is currently being investigated explicitly [10].

A staggered ferromagnetic phase like that reported here for Dy/Lu also occurs in the Gd/Y system [11]. There, structures with aligned and antialigned ferromagnetic blocks alternate as the Y layer thickness is varied, with a seven-Y-plane period. In Dy/Lu, we have observed parallel alignment of Dy blocks for Lu thicknesses of 5 and 8 atomic planes per bilayer, but samples with Lu layer thickness of 15, 20, 22, and 29 planes all order in the antiparallel arrangement of Fig. 2, i.e., no return to aligned stacking is observed. As the Lu layer thickness is increased, the coherence of the high-temperature helical phase decreases until, at 80 Å of Lu, all interlayer coherence is lost, as shown in Fig. 3. Nonetheless, even for samples where the helical order is coherent only within a single Dy layer, the antiferromagnetic stacking structure observed at low temperature is coherent over $\simeq 175$–275 Å, or several bilayers. In contrast to this, the aligned-structure coherence is limited to only one or two bilayers (see Fig. 3).

The greatly enhanced Curie temperature, the changes in coherence lengths, and the hysteretic magnetic properties are linked by a novel magnetoelastic process reported for the first time in this Letter. Using synchrotron radiation at the X14a beam line at NSLS, we have discovered that the entire superlattice undergoes a cooperative magnetoelastic distortion, breaking into 300 Å, orthorhombic domains oriented along the three equivalent easy axes. The domain structure minimizes the area dependence of the strain energy much as magnetic domains avoid the volume dependence of the magnetic energy. This mecha-

nism has not been recognized in earlier research. The sold lines in Fig. 4(a) show a contour map of the (20·1) Bragg peak in the (h0·1) plane for $[Dy_{15}|Lu_{15}]_{40}$ at 10 K. This peak is sharp at room temperature but begins to broaden below 160 K. The 10 K data were fitted by the sum of three identical, asymmetric Gaussians, with the principal axes tilted with respect to the crystallographic axes. The centers are located at positions consistent with domains of orthorhombic distortion approximately 60% that of bulk Dy; the Gaussian widths correspond to domain sizes of $\simeq$300 Å. There is, in contrast, no splitting or significant increase in width in the c^* direction. On warming to room temperature, some residual strain remains [Fig. 4(b)]. This strain apparently relaxes, as data taken on the same sample in successive runs months apart show no signs of permanent damage. The process of domain formation is hysteretic, but does not seem to require the irreversible formation of dislocations or other plastic deformations. The distortion in each domain is only slightly smaller than the full magnetostrictive deformation exhibited by the bulk material at the ferromagnetic transition. The domain size is such that the magnetoelastic strain never exceeds about one-half of a lattice spacing; indeed, this may be the main factor determining the domain geometry. We conclude that the domains form as columns in which a common long a axis is shared among successive layers, thereby inducing long-range alignment of the easy axis and hence magnetic coherence. This is similar to the cooperative Jahn-Teller effect [12], in which strain coupling induces long-range order. The existence of structural domains leads to a significantly larger dipolar coupling between Dy blocks than would exist if the layer were homogeneous, and this competes with any ferromagnetic RKKY coupling between layers. This competition provides a possible explanation for the reduced coherence of the aligned state and the absence of alternation between aligned and antialigned phases with Lu layer thickness.

It is generally recognized [6] that the ferromagnetic transition in Dy is driven by temperature-dependent single-ion magnetoelastic terms in the spin Hamiltonian. At 85 K, the energy gain from a distortion of the hexagonal basal plane becomes sufficient to overcome the exchange energy barrier between helical and ferromagnetic phases. Because the magnetoelastic distortion within each domain is smaller than that in bulk Dy, the observed enhancement of T_C must also involve a reduction in the exchange barrier. Earlier work [3] showed that single Dy layers grown on Er have an enhanced T_C. We have grown single Dy layers of various thicknesses, sandwiched between 500 Å slabs of Lu. Measurements made at 200 Oe show a steady increase in T_C as the Dy film is made thinner: 100 K for 140 atomic planes (400 Å), 125 K for 52 planes (145 Å), and 175 K for 15 Dy planes (40 Å). FC and ZFC magnetization curves for the 40 Å sample are shown in Fig. 5. The shift in T_C follows the coherency strain in the Dy layer, as determined by x-ray diffraction [13] at 300 K.

In the standard three-plane model [6], the energy barrier between helical and ferromagnetic states is proportional to $(1-\cos\omega)^2/\cos\omega$, where ω is the turn angle. In Dy/Lu superlattices ω falls in the range $28° \leq \omega \leq 35°$ at T_N, compared with $\omega = 43°$ in bulk Dy. The consequent reduction in the energy barrier, caused by coherency strains, evidently permits the magnetoelastic energy to drive the system ferromagnetic at temperatures close to T_N. While we are unable to measure the turn angles of the thinnest films, the similar field and temperature dependences of the magnetizations for the films and the superlattices lead us to conclude that the high T_C is due to epitaxial strain. Further, the appearance of orthorhombic domains at the ferromagnetic transition results in three local easy axes within the basal plane. As a result, the ZFC magnetization will be uniformly distributed among the six easy directions, frozen in place at low tem-

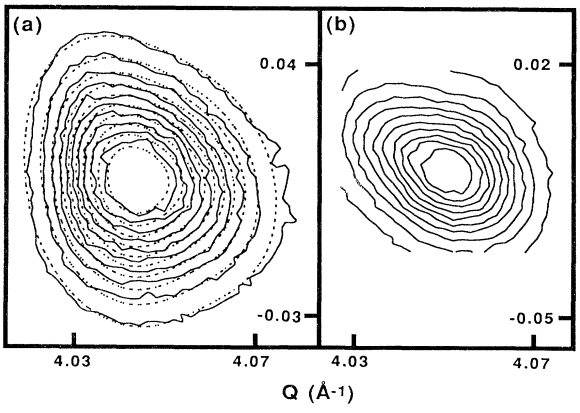

FIG. 4. X-ray intensity as a function of Q in basal plane scans through (20·1) at 10 K (a) and at room temperature (b) for SL $[Dy_{15}|Lu_{15}]_{40}$. Solid lines are data contours and the dashed lines are fits as described in the text.

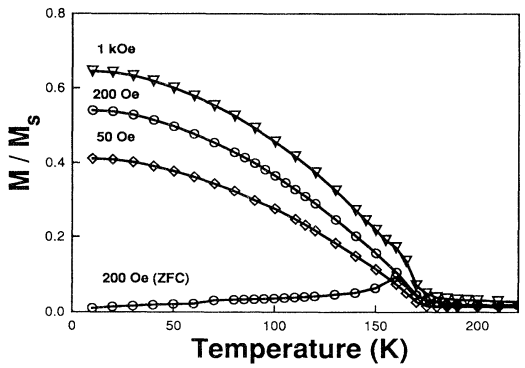

FIG. 5. FC magnetization vs temperature for a Lu/40 Å–Dy/Lu sandwich in fields of 50 Oe (diamonds), 200 Oe (circles, FC and ZFC), and 1000 Oe (grads).

peratures by anisotropy barriers even for films.

The situation is different for field cooling. In a film the magnetization must orient along those easy directions most nearly aligned with the field. Epitaxy, however, prevents the growth of these domains until the magnetic energy becomes large enough to detach the film or superlattice from its substrate. In superlattices, the addition of dipolar coupling between layers further reduces the ZFC magnetization.

In summary, we have found that the compressive epitaxial strain imposed on Dy by Lu substrates greatly increases the Curie temperature of the Dy. The magnetoelastic distortion that drives the transition in the bulk is observed also in superlattices, and by inference in films. In this new mechanism, the system forms columnar, orthorhombically distorted domains, for which the mean strain largely vanishes. From a broader perspective it appears likely that this pathway to magnetization may offer a general mechanism whereby barriers to magnetoelastic strain may be avoided. In the Dy-Lu system studied here, a competition between RKKY interactions that couple Dy blocks and dipolar interactions associated with domain formation evidently reduces any tendency for parallel alignment of Dy layers while strongly favoring antiparallel stacking.

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