Competing Anisotropies in Holmium-Erbium Superlattices

J. A. Simpson,[1] D. F. McMorrow,[2] R. A. Cowley,[1] D. A. Jehan,[1] R. C. C. Ward,[1] M. R. Wells,[1] and K. N. Clausen[2]
[1]*Oxford Physics, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, United Kingdom*
[2]*Risø National Laboratory, DK 4000 Roskilde, Denmark*
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The effect of competing crystal-field anisotropies on magnetic order has been investigated in a series of Ho/Er superlattices. For temperatures in the interval $T_N(\mathrm{Er}) \leq T \leq T_N(\mathrm{Ho})$ the Ho basal-plane order propagates coherently through the paramagnetic Er with a typical length scale of 1000 Å. Below $T_N(\mathrm{Er})$ the coherence length of the basal-plane order decreases, while the longitudinal component of the Er moments fails to order across the Ho block. It is argued that these results require an extension of current models of indirect exchange in superlattices to explicitly include the superlattice band structure.

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Magnetic superlattices have a number of unusual properties that both are challenging our understanding of exchange interactions in metals and are proving to be technologically useful. To date, studies of systems fabricated from alternating blocks of a magnetic element and a nonmagnetic spacer have dominated this field of research. In the $3d$ transition metal superlattices a giant magnetoresistance (GMR) effect has been discovered [1], and while a complete theory of this phenomenon is still lacking, it is known that scattering of the conduction electrons at the interface between the two elements plays an important role in determining the size of the GMR signal [2–4]. For rare-earth superlattices, helical magnetic order is found to propagate through nonmagnetic spacer layers (either Y or Lu) even when the thickness of the spacer exceeds 100 Å [5]. A detailed theory for this effect has also yet to be developed, although it has been argued that it can be understood within the framework of the RKKY model [6] of indirect exchange mediated by a spin polarization of the conduction electrons. In this Letter we report on studies of the magnetic structure of Ho/Er superlattices, where both elements are magnetic, but with crystal-field anisotropies that favor very different magnetic structures. Our results exhibit several unexpected features and provide further insight into the nature of the exchange in metallic superlattices.

Bulk Ho orders magnetically below $T_N \approx 132$ K into a helical structure. The localized $4f$ moments are confined to the hexagonal basal planes by the crystal field, forming a ferromagnetic sheet. The direction of the moments in these sheets rotates from one plane to the next along the c axis. At T_N the modulation wave vector describing the order is $\mathbf{q} \approx (2/7)\mathbf{c}^\star$, which reduces on cooling, until at 18 K it becomes equal to $(1/6)\mathbf{c}^\star$, and the moments tilt out of the basal plane to form a cone structure [7]. In the case of Er, the ordering at $T_N \approx 84$ K is to a longitudinal c-axis modulation with $\mathbf{q} \approx (2/7)\mathbf{c}^\star$. Between 52 and 18 K a component develops in the basal plane and projections of the moments form a cycloid in the a-c plane [8,9]. Below 18 K the moments form a cone with a ferromagnetic component along the c axis and a basal-plane helix with $\mathbf{q}=(5/21)\mathbf{c}^\star$. Thus within the temperature range 18 K $\leq T \leq$ 84 K the crystal fields in these two elements favor different orientations of the moments: Ho confined predominantly to the basal plane, and Er aligned mainly along the c axis.

There have been several studies of the magnetic structures of superlattices composed of Ho or Er grown in blocks separated by a nonmagnetic spacer such as Y or Lu [10–12]. In these systems the magnetic order is coherent through the nonmagnetic block, in the sense that the chirality of the order is preserved, and there is a well-defined phase change of the order across each nonmagnetic block. The coherence length describing the order is typically 1000 Å, and is largely independent of temperature. We have determined the magnetic structure of a series of three Ho/Er superlattices as a function of temperature using neutron diffraction. Results from this system have implications for the nature of the exchange interactions and the propagation of long-range order in magnetic superlattices. In particular, we argue that our results cannot be understood within conventional RKKY theory, unless it is modified to allow explicitly for the band structure of the superlattice.

The Ho/Er superlattices were grown by molecular beam epitaxy (MBE) following the method developed by Kwo [13]. Using this technique the superlattices grow with the c axis normal to the substrate and are epitaxially aligned to form good-quality single crystals (see, for example, Ref. [11]). Three superlattices were investigated with compositions as determined by x-ray diffraction of $\mathrm{Ho}_{43}/\mathrm{Er}_{10}$, $\mathrm{Ho}_{20}/\mathrm{Er}_{22}$, and $\mathrm{Ho}_{10}/\mathrm{Er}_{23}$, where the subscripts refer to the number of atomic planes of each element, and a typical sample contains fifty biblock units. The magnetic structures were determined using the TAS6 triple-axis spectrometer at Risø National Laboratory, Denmark. The superlattices were mounted in a variable temperature cryostat with the $(h\ 0\ \ell)$ crystallographic plane in the scattering plane, and measurements were made at 8, 30, 50, and 100 K. At each temperature the neutron wave-vector transfer $\mathbf{Q}$ was scanned along the $[00\ell]$ direction to give details of the ordering of moments

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in the basal plane, and also along [10ℓ], providing information on the alignment of moments along the c axis. The wave-vector resolution with 5 meV neutrons was typically 0.012 Å^{-1} (FWHM).

Initially, we shall discuss the results obtained at 100 K, a temperature selected to be below T_N of Ho, but above that of Er. As an example, the scattering observed from Ho_{40}/Er_{13} is presented in Fig. 1. The well-resolved magnetic peaks show that basal-plane order is established, and that this order is coherent through the Er blocks with a coherence length of ≈ 600 Å. (After allowing for the contribution from the instrumental resolution, the real-space coherence length ξ was calculated from the FWHM in reciprocal space, ΔQ, using $\xi = 2\pi/\Delta Q$.) As shown by the solid line in Fig. 1, an excellent fit to both the [00ℓ] and [10ℓ] data was achieved using the same model developed for Ho/Y and Ho/Lu. (For a complete description of this model see [11].) The modeling also confirmed that the interfaces between the Ho and Er blocks were resticted to four or five atomic planes, the same width found for other rare-earth superlattices [5,11]. Therefore, in the paramagnetic state, Er behaves in the same manner as nonmagnetic Y or Lu; the long-range order propagates through the Er despite the localized Er $4f$ moments being disordered. The turn angle between successive Ho planes was determined from the fits as $46° \pm 1°$, compared to the bulk value at 100 K of 44°, and for paramagnetic Er the corresponding angle is $51° \pm 1°$.

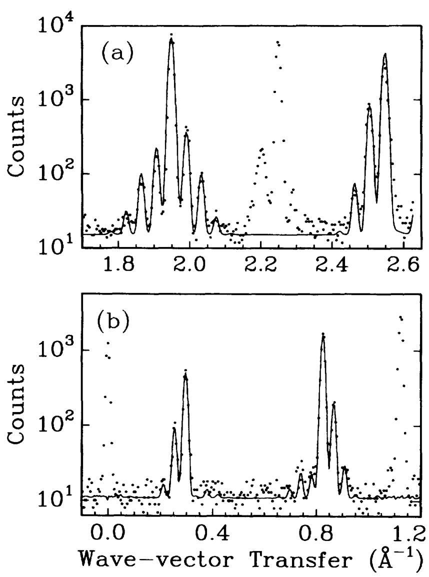

FIG. 1. The neutron scattering observed from the $Ho_{40}Er_{13}$ sample at 100 K: (a) shows the scattering for wave-vector transfer along [00ℓ] and (b) that along [10ℓ]. The solid line is a fit to the data of a model with basal-plane helical ordering of Ho and no ordering of the Er moments. The peaks near ℓ=2.2 Å^{-1} along [00ℓ] and ℓ = 0 and 1.1 Å^{-1} along [10ℓ] are the (002), (100), and (101) nuclear Bragg peaks and are not included in the model.

Fits to the data from the other two superlattices at the same temperature yielded, within error, identical results.

On cooling below 100 K, the scattering changed in two ways. First, along the [00ℓ] direction, the widths of the peaks increased steadily with decreasing temperature. Although the coherence length of the basal-plane order decreases as the temperature is reduced, as shown in Fig. 2 for all three superlattices, it remains much greater than the superlattice period. Second, the magnetic scattering along [10ℓ] consisted of well-resolved peaks from the coherent basal-plane order and a broad peak, as shown in Fig. 3 for Ho_{20}/Er_{22} at 8 K. Least-squares fits of a sum of Gaussians to the [10ℓ] data revealed several key features of the broad peak: its position corresponds to the turn angle found in bulk Er at the same temperature and its width to the value expected if the c-axis Er moments in each block order independently. [For example, the width of the broad peak in Fig. 3 from Ho_{20}/Er_{22}, which has an Er block length of 63 Å, is 0.097 Å^{-1}(FWHM). This corresponds to a real-space coherence length of 65 Å.] Thus, although the basal-plane order is coherent across several biblocks, the longitudinal Er order is confined to a single Er block. At present we have been unable to unambiguously determine the detailed structure adopted by the Er moments, but from the [00ℓ] data we have been able to estimate the turn angle through the Ho blocks. The temperature dependence of this turn angle and that of the broad peak along [10ℓ] are given in Fig. 4. We consider that the changes observed in the magnetic coherence length are unrelated to the lattice mismatch between Ho and Er. Such effects have not been reported for other superlattices, such as Ho/Y, where the fractional mismatch is approximately 4 times greater.

The lack of coherence of the longitudinal Er moments in these superlattices is surprising, especially in view of the result that long-range order has been observed

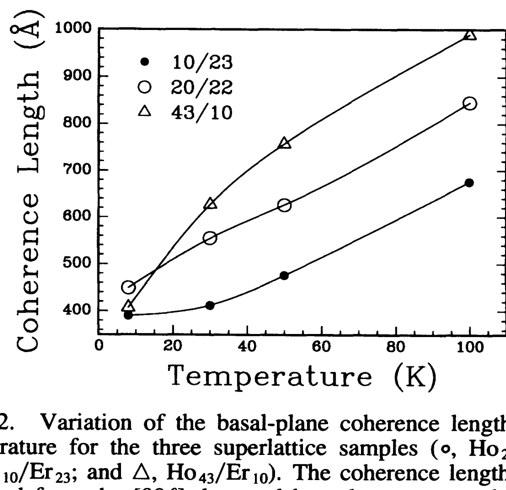

FIG. 2. Variation of the basal-plane coherence lengths with temperature for the three superlattice samples (∘, Ho_{20}/Er_{22}; •, Ho_{10}/Er_{23}; and △, Ho_{43}/Er_{10}). The coherence lengths were obtained from the [00ℓ] data and have been corrected for the instrumental resolution.

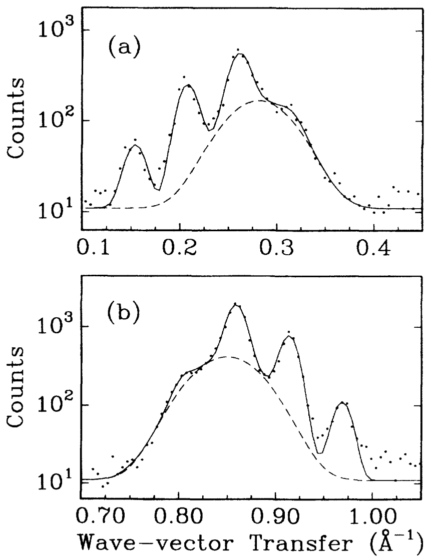

FIG. 3. Fits to the [10ℓ] intensity for the Ho_{20}/Er_{22} sample at 8 K with a series of Gaussian peaks, showing how the profile is formed by a combination of sharp (solid lines) and broad (dashed line) peaks. The broad peak has a real-space correlation length determined by the thickness of the Er block (≈ 60 Å) as described in the text.

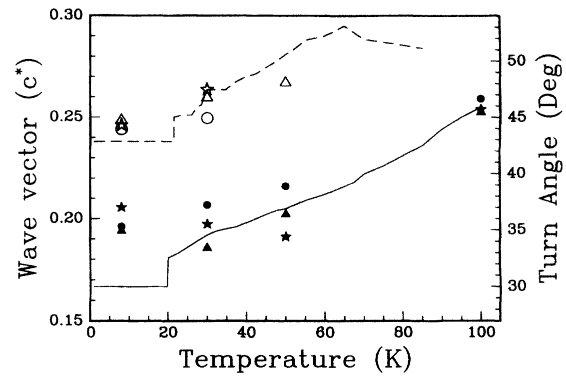

FIG. 4. Wave vector (and equivelant turn angle) for the Ho and Er moments (★, Ho_{10}/Er_{23}; •, Ho_{43}/Er_{10}; and ▲, Ho_{20}/Er_{22}). This is calculated for the Er from the position of the broad [10ℓ] peak (open symbols), and a model comprising helical basal-plane order in Ho (filled symbols). The dashed and filled lines are the bulk variation for Ho and Er, respectively.

in the Er/Y system [10]. We shall now consider the implications that our results have on models of exchange in metallic superlattices.

Previous attempts to explain the coherent propagation of magnetic order in rare-earth superlattices composed of alternating blocks of magnetic and nonmagnetic ions have argued that the observed features can be understood within the spirit of conventional RKKY theory [14,15]. That is, for a given magnetic ion, the strength of the coupling through the nonmagnetic spacer should depend on the the size of the ordered moment in the magnetic block and the conduction electron susceptibility of the spacer layer: the driving force and the response. We believe that it is difficult to reconcile this simple model with either the observed lack of coherence of the longitudinal Er moments in Ho/Er superlattices, or other known characteristics of rare-earth superlattices. For the Ho/Er superlattices, this model predicts that as the conduction electron susceptibilities of Ho and Er are similar, the coherence of the basal-plane and longitudinal order should also be similar. There are other apparent inconsistencies which we choose to highlight. First, the magnetic coherence length in systems such as Ho/Y is independent of temperature. On the basis of the conventional picture it should decrease close to T_N as the localized Ho moment decreases and hence the degree to which the conduction electrons in the spacer are spin polarized is reduced, but this has not been observed. Second, the magnon dispersion relationships for the rare earth elements [16] and the critical concentrations of their alloys with Y [17] indicate that the exchange extends to approximately sixth-nearest neighbors only. In contrast, magnetic order in the Ho/Y superlattices extends over at least forty atomic planes [11]. Third, in some cases Ho blocks in Ho/Lu [12] or Dy in Dy/Lu [18] multilayers order ferromagnetically, with either a ferromagnetic or antiferromagnetic coupling across the nonmagnetic spacer, so that the turn angle across the nonmagnetic spacer is $n\pi$. In contrast, when there is helical order in the same samples, the turn angle across the nonmagnetic spacer is not an integer multiple of π. This shows that the magnetic interactions through the nonmagnetic layers are dependent on the nature of the magnetic order, and cannot be described by a Heisenberg $\mathbf{S}_1 \cdot \mathbf{S}_2$ interaction.

A possible explanation of these results may be obtained from a model which takes account of the band structure of the magnetically ordered superlattice, and so differs from the conventional RKKY interaction which treats the magnetic ordering as a perturbation. Electron states in the ordered phase are then determined by the band structure, allowing for the superlattice periodicity. The effects of magnetic order on the band structure of the rare-earth elements are largest for electrons close to the Fermi surface, as discussed by Elliott and Wedgwood [19]. Using a simple free-electron model of the conduction electrons, they show that the electrons most affected by the development of magnetic order, described by wave vector $\mathbf{q}$, are those with wave vectors $\mathbf{k}\pm = (k_x, k_y, \pm\frac{q}{2})$. For two elements with similar conduction bands forming a magnetic/nonmagnetic superlattice, conduction electrons in each block will have similar wave functions. Consequently, electrons at the Fermi surface will propagate between two layers and, if there is negligible additional spin scattering at the interfaces, this will give rise to a continuous spin-density wave in the nonmagnetic blocks similar to that in magnetic blocks. The driving force for coherent long-range order is the decrease in the energy of the conduction electrons if they are no longer confined to a single block, and the correlation length is then determined by the electron mean-free path, rather than the height of the peak in the conduction electron susceptibility.

These considerations of the electron states offer an explanation for the observed lack of coherence in the c-axis Er moments in the Ho/Er superlattices. Helical magnetic order leads to electrons at the Fermi surface with a wave function $|\mathbf{k}_\uparrow^+\rangle + |\mathbf{k}_\downarrow^-\rangle$, where the arrows represent the spin directions with respect to the c axis. There is then a helically polarized conduction spin-density wave in both the Ho and Er blocks. In contrast, a c-axis modulation leads to states at the Fermi surface of the form $|\mathbf{k}_\uparrow^+\rangle + |\mathbf{k}_\uparrow^-\rangle$ and $|\mathbf{k}_\downarrow^+\rangle - |\mathbf{k}_\downarrow^-\rangle$. As it is not possible to match these states onto those associated with helical order in the Ho blocks, there will be no coherence of the longitudinal Er moments between Er blocks, because the conduction electrons in the helically ordered state cannot transmit the c-axis phase information. In a similar manner ferromagnetic basal plane order in the Ho or Dy for the Ho/Lu and Dy/Lu systems [12,18] leads to a ferromagnetic spin-density wave polarized in the basal plane. This then produces a similar spin-density wave in the nonmagnetic spacer, giving rise to an interaction through the spacer which is either ferromagnetic or antiferromagnetic rather than helical in character.

It is also worth noting that other mechanisms may lead to a reduction in the magnetic coherence length. As is well known, for the rare-earth elements [16] the gaps opened up in the Fermi surface by the formation of a localized moment reduce the conduction electron susceptibility, and may in exceptional circumstances produce a phase transition, as in metallic Tb. Alternatively, random strains may lead to a reduction in the coherence length, as has been recently suggested for thin films of Ho [20]. Similarly, magnetoelastic and dipolar effects can influence the ordering of ferromagnetic systems. However, we consider that none of these mechanisms provide a satisfactory explanation of the experimental results presented in this Letter.

In conclusion, we have studied the evolution of the magnetic structure of Ho/Er superlattices with temperature using neutron diffraction. Above the ordering temperature of bulk Er, the Ho blocks align in a basal-plane spiral with coherence through the Er layers. At low temperatures Ho order is coherent through the Er, but the c-axis component of the Er moment fails to order through the Ho. We have suggested that these effects arise from conflicting boundary conditions on the conduction electron wave functions at the Ho/Er interface, and indicated the direction for future calculations of interactions in rare-earth superlattices.

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