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1996 J. Phys.: Condens. Matter 8 L187

(http://iopscience.iop.org/0953-8984/8/11/005)

J. Phys.: Condens. Matter **8** (1996) L187–L194. Printed in the UK

LETTER TO THE EDITOR

Persistence of helical magnetic order in dysprosium–holmium superlattices

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Received 14 December 1995

Abstract. The magnetic structures of two Dy/Ho superlattices have been studied by neutron scattering using a time-of-flight spectrometer. For temperatures below the Néel temperature of bulk Dy, T_N(Dy), but above T_N(Ho), the Dy blocks order in a helical magnetic structure which has long-range coherence across several superlattice repeats, despite the Ho atoms being paramagnetic. Below ∼140 K helical order also develops in the Ho blocks, and the ferromagnetic transition of Dy at lower temperatures is suppressed, so the helical phase is found at all temperatures down to 2 K.

The application of molecular beam epitaxy (MBE) to the growth of metallic superlattices has opened up new areas of research in magnetism. Magnetic systems may now be produced that exhibit properties not observed in the bulk. In the case of the rare earths, many systems have now been investigated. To date most of these studies have focused on superlattices formed from one magnetic rare-earth element and one of the non-magnetic elements Lu, Y and Sc. For the Dy/Y [1], Dy/Lu [2], Ho/Y [3] and Ho/Lu [4] systems, the two key results are that the helical magnetic order characteristic of the bulk propagates through the non-magnetic element up to distances in excess of 100 Å, and that phase transitions, such as the helical-to-ferromagnetic transition in Dy, are modified in the superlattice.

More recently there has been a trend to explore the effects produced if two magnetic elements are used [5]. These may be two heavy rare earths that exhibit competing crystal-field anisotropies, as in the cases of Ho/Er [6] or Er/Dy [7], or one heavy and one light rare earth, for example Ho/Pr [8], or even two light rare earths such as Nd/Pr [9]. This second type of superlattice displays a range of unusual properties, as in the case of Ho/Er where the basal-plane components of the moments have long-range coherence along the *c*-direction, but the ordering of the *c*-axis component of the Er moments is coherent across one Er block length only [6]. Such observations indicate that further theoretical work is required to describe the magnetic interactions in superlattices, and that the study of new systems may provide additional experimental evidence to complement this.

In this letter we present the results of a neutron scattering study of two Dy/Ho superlattices. Here, both elements are magnetic and have crystal-field anisotropies that confine the magnetic moments to the basal plane. The magnetic structure of Dy was first investigated using neutron scattering by Wilkinson *et al* [10]. Below a Néel temperature of 179 K the moments order to lie in the basal plane. The moments are ferromagnetically coupled within each basal plane, but the orientation rotates through an

0953-8984/96/110187+08$19.50

angle in moving between successive planes along the *c*-direction. The angle is characterized by a wave vector, $\boldsymbol{q}$, which has a value of $0.240\boldsymbol{c}^\star$ at the onset of order. The wave vector reduces on cooling until at $T_C = 87$ K, $\boldsymbol{q} = 0.147\boldsymbol{c}^\star$. Below T_C the magneto-elastic energy causes a transition to a ferromagnetic structure, and this is associated with an orthorhombic distortion of the basal-plane lattice.

The magnetic structure of Ho was first determined by Koehler *et al* [11], and is also a basal-plane spiral. The Néel temperature is ≈132.2 K and $\boldsymbol{q} = 0.275\boldsymbol{c}^\star$ at T_N. In a similar manner to that of Dy the wave vector reduces on cooling; however, the magnetic structure is a spiral with $\boldsymbol{q} \neq 0$ for all $T \leqslant T_N$(Ho). As the crystal-field anisotropy increases rapidly below ∼30 K, the wave vector locks into values commensurable with the chemical lattice, and the moments form configurations known as spin-slip structures. These were first observed by Gibbs *et al* [12] and the model was further refined by Cowley and Bates [13]. Below ≈20 K the wave vector locks into a value of $(1/6)\boldsymbol{c}^\star$ and the moments tilt out of the basal plane due to the temperature dependence of the axial anisotropy [14].

The Dy/Ho system therefore presents several temperature regions of possible interest: T_N(Ho) $\leqslant T \leqslant T_N$(Dy), where bulk Dy is helical, and Ho is paramagnetic; T_C(Dy) $\leqslant T \leqslant T_N$(Ho), where both Dy and Ho may order as helices; and $T \leqslant T_C$(Dy), when the Dy may order ferromagnetically, and alter the magnetic structure in the Ho blocks.

Two Dy/Ho superlattices were grown by MBE following the same method as was used for the Ho/Y system [3, 15]. A buffer layer of Nb was first deposited on a sapphire substrate, followed by a seed layer of Y to allow the lattice parameter to relax to its bulk value. Alternate layers of Dy and Ho were then deposited at a substrate temperature of 400 °C with the growth direction along the *c*-axis, and this was repeated 50 times. The samples had compositions Dy_{32}/Ho_{11} and Dy_{16}/Ho_{22}, where the subscripts refer to the number of planes in each bi-block unit. These were characterized by x-ray diffraction using a triple-axis spectrometer mounted on a Stöe rotating-anode generator operating at 6 kW. Measurements were made with the wave-vector transfer, $\boldsymbol{Q}$, along the $[00\ell]$ reciprocal-lattice direction, and also transversely through the (002) Bragg reflection. The mosaic spread is determined from this latter measurement, and was approximately ∼0.3° for both superlattices. This value is consistent with those obtained for previous rare-earth superlattices (for example the value obtained for the Ho/Y system is ∼0.2° (FWHM) [3]).

The magnetic ordering was investigated by neutron scattering using a time-of-flight neutron scattering technique. This method is a convenient way of observing elastic scattering provided that $\boldsymbol{Q}$ is along a direction that passes through the origin of reciprocal space. A scan with the wave-vector transfer along $[00\ell]$ provides information solely about the basal-plane components of the moments, and the crystal-field anisotropies of both Dy and Ho favour alignment of moments in this plane. The Dy/Ho system is therefore an ideal system for a time-of-flight experiment, and this technique may then offer advantages of high resolution and rate of data collection compared to a reactor-based triple-axis spectrometer method. The experiments were performed using the IRIS high-resolution time-of-flight spectrometer [16] at the ISIS Facility of the Rutherford Appleton Laboratory. This instrument provides good resolution ($\delta Q/Q \approx 2.5 \times 10^{-3}$) at low Q by the use of long wavelengths and back-scattering geometry. The samples were mounted in a helium-flow cryostat, which allowed temperature control to ±0.1 K, with the $(h0\ell)$ crystallographic plane in the scattering plane. The elastic scattering with wave-vector transfer along the $[00\ell]$ reciprocal-lattice direction was obtained from the signal measured by a single high-angle detector. Measurements were made in temperature steps of either 5 K or 10 K on warming between 5 K and 200 K.

The scattering from the Dy_{32}/Ho_{11} sample at 160 K, and from the Dy_{16}/Ho_{22} sample at 140 K is shown in figure 1. These are both temperatures above the bulk value of T_N(Ho)

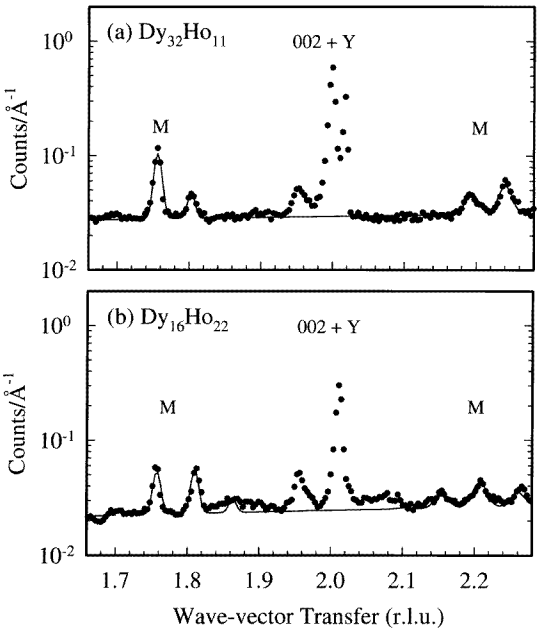

Figure 1. The neutron scattering with the wave-vector transfer along $[00\ell]$ for (a) Dy_{32}/Ho_{11}, $T = 160$ K, and (b) the Dy_{16}/Ho_{22} sample at $T = 140$ K. The solid line is a fit to the data of a model with basal-plane helical ordering of Dy and no ordering of the Ho moments. The peaks near $\boldsymbol{Q} = 2\boldsymbol{c}^*$ are the (002) nuclear Bragg peaks and are not included in the model.

but below T_N(Dy). In addition to the (002) nuclear Bragg scattering, figure 1 shows a series of sharp peaks with $\boldsymbol{Q}$ above and below the (002) peak. This indicates that the Dy moments have ordered in a spiral structure which is coherent across several superlattice repeats. We have adopted a procedure for modelling the magnetic ordering similar to that used to describe the Ho/Y system [3]. We assume a helical ordering in the Dy blocks with a moment A_{Dy}. The possibility of helical order developing in the Ho regions is also allowed for, and this has a moment length A_{Ho}. The model attempts to account for the fact that the interfaces between successive blocks are not ideal, but instead extend over a small number of atomic planes, λ. Properties such as the lattice parameters and turn angles are assumed to vary in a smooth manner across an interface region. The lattice parameter in the lth atomic plane may be calculated from the lattice parameters in the centre of a Dy or Ho block, d_{Dy} and d_{Ho} respectively, using the expression

$$d_l = \frac{(d_{Dy} + d_{Ho})}{2} + \frac{(d_{Dy} - d_{Ho})}{2} \tanh[(l - I)/\lambda] \tag{1}$$

where I is the position of the interface. The turn angle between successive atomic planes, β_l, may also be calculated from an expression similar to equation (1) from the turn angles in the centre of Dy and Ho blocks, ψ_{Dy} and ψ_{Ho} respectively. For moment lengths A_{Dy} and A_{Ho} for the Dy and Ho spirals, the moment length in the lth atomic plane is given by

$$J_l = \frac{(A_{Dy} + A_{Ho})}{2} + \frac{(A_{Dy} - A_{Ho})}{2} \tanh[(l - I)/\lambda]. \tag{2}$$

For a coherent magnetic structure extending over the whole superlattice, the magnetic

scattering along $[00\ell]$ is then proportional to the function

$$I(\boldsymbol{Q}) \propto |A^{+}(\boldsymbol{Q})|^2 + |A^{-}(\boldsymbol{Q})|^2 \tag{3}$$

where

$$A^{\pm}(\boldsymbol{Q}) = \sum_{m=1}^{N} \mathrm{e}^{\mathrm{i}m(LQ\pm\Phi)} \sum_{l}^{one\ bilayer} J_l\, \mathrm{e}^{\mathrm{i}(QR_l\pm\beta_l)}. \tag{4}$$

In equation (4), Φ is the total phase advance across one bi-block unit of length L along c, and

$$R_l = \sum_{j=1}^{l} d_j. \tag{5}$$

The observed neutron scattering may then be simulated by convoluting with the resolution function of the instrument, which we have assumed to be a Gaussian, and adding a sloping background. The model was then used in a least-squares fitting routine to obtain the best-fit values for the parameters ψ_{Dy}, ψ_{Ho}, d_{Dy}, d_{Ho} and $r = A_{Ho}/A_{Dy}$. The values of the other salient parameters (number of atomic planes in the Dy and Ho blocks, n_{Dy} and n_{Ho} respectively) were held fixed at values that produced consistent fits for all temperatures, and in particular the interface width (λ) was approximately four lattice planes. Agreement between theory and experiment is generally good, and is shown by the solid line in figure 1. Within error, the samples ordered at the Néel temperature of bulk Dy. In the temperature region $\sim$140 K $\leqslant T \leqslant T_N$(Dy), the fits all gave values of r of zero within error. This indicates that the Ho moments remain paramagnetic above their bulk Néel temperature, and despite this the Dy spiral is coherent across regions of disordered Ho moments. A similar effect has also been observed in the Ho/Er [6] and Dy/Er [7] systems.

The integrated intensity of the (002) Bragg peak was determined at each temperature and was found to be constant down to the lowest temperatures investigated. This shows that the Dy blocks do not undergo the transition to a ferromagnet as in the bulk, since this would cause additional scattering at this position. The data taken at 40 K are shown for both samples in figure 2. The magnetic scattering consists of a series of sharp peaks with $\boldsymbol{Q}$ above and below the (002) reflection as in the high-temperature data, although their position has changed as the wave vector of the spiral has varied. The low-temperature data were fitted using the above model, and good agreement is still obtained, as shown by the solid line in figure 2. Magnetic order develops on the Ho moments in a continuous fashion, and the variation of r is shown in figure 3, indicating that T_N(Ho) $\sim$ 140(3) K, which is slightly higher than in the bulk. The magnetic structure below T_N(Ho) is therefore a spiral for both elements, and the low-temperature ferromagnetic transition for Dy is suppressed.

Our results for the turn angles obtained from this modelling are summarized in figure 4, where they are compared to the bulk variation. We estimate that the error in the derived values of the turn angles is $\pm 1^{\circ}$. For the Ho blocks, the turn angle is constant above T_N(Ho) with a value of 51°. This supports the idea that the variation in the turn angle is driven by the ordered moment. ψ_{Ho} gradually reduces on cooling below T_N(Ho) with very similar behaviours obtained for the two samples. In contrast, the Dy turn angle reduces on cooling until $T \sim 60$ K, and below this there is a small increase in ψ_{Dy} to a low-temperature value of $\approx 30^{\circ}$. The values shown in figure 4 are the results from fits where all of the parameters could vary freely; however, a negligible difference in χ^2 is obtained if ψ_{Dy} is held constant at 30° at low temperatures. Below $\sim$30 K, the results are consistent with the Dy moments locking to the $(1/6)\boldsymbol{c}^{\star}$ structure, and a $(1/5)\boldsymbol{c}^{\star}$ lock-in occurring in the Ho blocks.

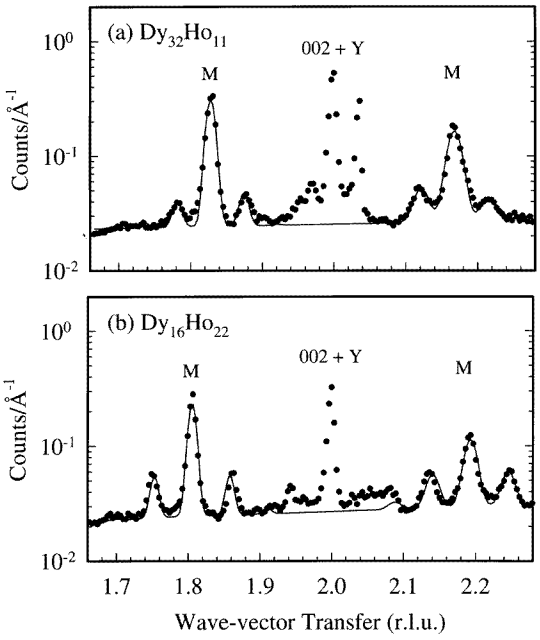

Figure 2. The neutron scattering with the wave-vector transfer along $[00\ell]$ observed at $T = 40$ K from (a) the Dy_{32}/Ho_{11}, and (b) the Dy_{16}/Ho_{22} sample. The solid line is a fit to the data of a model with basal-plane helical ordering of both the Dy and Ho moments. The peaks near $Q = 2c^*$ are the (002) nuclear Bragg peaks and are not included in the model.

Previous studies of Dy-based superlattices include those of Dy/Y [1] and Dy/Lu [2], where very different behaviours were found for ψ_{Dy} in each case. In the former there is an expansive basal-plane strain of the Dy layers, which results in the ferromagnetic phase being suppressed at all temperatures. In contrast, there is a compressive strain for the Dy basal planes in Dy/Lu, and the transition temperature to a ferromagnet is enhanced. The strain for the Dy layers in the Dy/Ho system is the same as that for Dy/Lu, although the lattice mismatch is much smaller: 0.4% as compared to 2.5%. On this basis, the ferromagnetic phase of Dy would be expected, possibly with a slight increase in the transition temperature from the bulk value. The absence of this phase is therefore somewhat surprising, and may indicate that the strain is not the sole factor determining the wave vector of magnetic order in a superlattice.

A possible explanation for this effect may be found by considering the magnetic structure of the superlattice as a whole, rather than as isolated Dy or Ho blocks. The conduction electrons can form a coherent helical spin-density wave in both materials, but if there is a transition to a ferromagnetic state, this spin-density wave would be disrupted. This leads to a higher energy state and the possible loss of coherence between the ordering in different blocks. Alternatively, if a helical/ferromagnetic structure did have long-range coherence, there might be an increase in energy if the moments at the interface had to make a smooth transition between the two magnetic structures. Thus it may be the case that, although the magneto-elastic energy for an individual Dy block favours a ferromagnetic alignment of the moments, the overall energy of the superlattice is minimized when both elements have helical order which is coherent across several superlattice repeats.

Further insight into the behaviour of ψ_{Dy} may be found from the theoretical work of

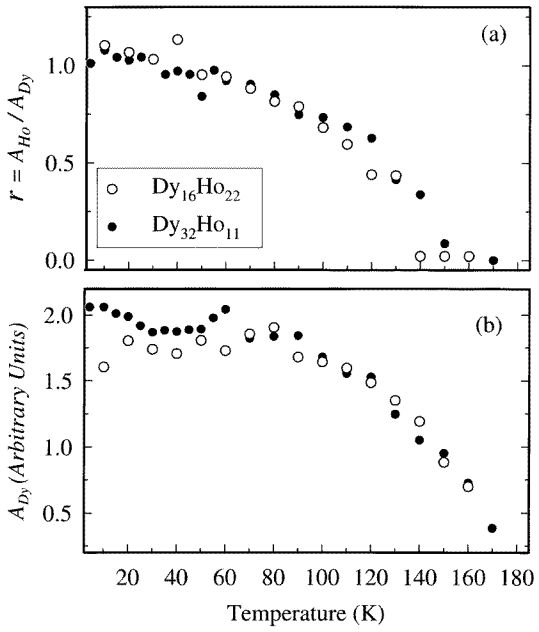

Figure 3. (a) The variation of the ratio of the Ho moment length to the Dy moment length ($r = A_{Ho}/A_{Dy}$). This indicates T_N(Ho) $\sim$ 140 K for the Ho moments in both samples. (b) The overall scale factor of the scattering, which is proportional to A_{Dy}, the Dy moment length. This behaves differently from r, and indicates that T_N(Dy) is close to the bulk value of 179 K for both samples.

Seno *et al* [17], who considered the possible values of $\boldsymbol{q}$ for a spiral structure using a model with a crystal-field anisotropy of sixfold symmetry and a two-neighbour exchange interaction. They demonstrated that when the crystal-field energy becomes large compared to the exchange interaction, values of $\boldsymbol{q}$ commensurable with the crystal lattice are formed with a wave vector determined by the ratio of the two exchange constants. As these constants change, many commensurable states are formed with $\boldsymbol{q} = n\boldsymbol{c}^\star$ if $n \geqslant \frac{1}{6}$. However, the only state possible with $n < \frac{1}{6}$ is $n = 0$, i.e. the ferromagnet. For the Dy/Ho system, the crystal-field energy is small at high temperatures and the wave vector is not commensurable with the chemical lattice. On cooling, the exchange energy causes $\boldsymbol{q}$ to decrease, with a value of approximately $(1/6)\boldsymbol{c}^\star$ obtained at $\sim$60 K. As the temperature is further reduced, the exchange energy might be expected to favour a further reduction in $\boldsymbol{q}$, but the crystal-field energy has increased sufficiently to cause lock-ins of the spiral wave vector. If the only stable state with $\boldsymbol{q} < \frac{1}{6}\boldsymbol{c}^\star$ is the ferromagnet, which is energetically unfavourable for the superlattice, this could account for the behaviour of ψ(Dy) below 60 K. The wave vector at these temperatures locks in to $(1/6)\boldsymbol{c}^\star$ as this is the smallest value which minimizes the crystal field and exchange energies in the superlattice as a whole.

In conclusion, we have investigated the magnetic structure of two Dy/Ho superlattices. The Dy moments order in a spiral at all temperatures, and below a Néel temperature of $\sim$135 K helical order also develops in the Ho blocks. The ferromagnetic transition of Dy is not observed, despite the strain imposed on the Dy layers favouring this. It is argued that a ferromagnetic phase for Dy moments would result in an energy state higher than would be obtained for a coherent helical ordering throughout the superlattice. The low-temperature

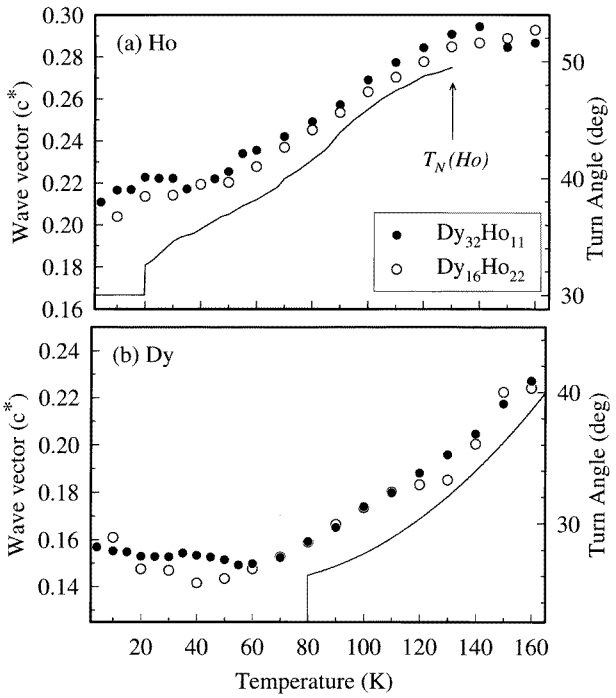

Figure 4. The wave vector and equivalent turn angle for (a) Ho and (b) Dy moments deduced from the model in the text (○, Dy_{16}/Ho_{22}; ●, Dy_{32}/Ho_{11}). The bulk variation for each element is shown by the solid lines. Ψ_{Dy} is observed to decrease until ∼60 K is reached and a small increase occurs on cooling, reaching a low-temperature value of ≈30°. However, as discussed in the text, the data at low temperatures are also consistent with $\Psi_{Dy} = 30°$.

turn angle in the Dy blocks is then restricted to being ≈30°, since this structure has the smallest possible wave vector other than that of the ferromagnet.

The work in Oxford was funded by the EPSRC. We are grateful for the technical staff at ISIS for the help throughout the experiment.

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