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The magnetic phase transition of a lattice-matched holmium thin film

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Abstract. Neutron and x-ray scattering measurements have been made of the magnetic phase transition in a thin single-crystal film of holmium. The 5000 Å film was grown on a seed of a lutetium and yttrium alloy with a concentration chosen such that the basal plane was lattice matched to the holmium film. It was capped by 10 000 Å of a similar film. The measurements below T_N gave the critical exponent $\beta = 0.37 \pm 0.03$. Above T_N the scattering suggests that the anomalous long-length-scale scattering is at least 20 times weaker than has been observed in similar experiments on bulk and thin-film holmium. These results show that the long-length-scale scattering is associated with the crystallographic surface rather than the end of the magnetic layer. The implications of this result for the origin of this type of scattering are discussed.

1. Introduction

The development of high-resolution x-ray scattering techniques and their application to the study of the critical scattering at structural and magnetic phase transitions has led to the discovery of critical scattering close to phase transitions with two different length scales. One of these length scales is associated with the normal critical scattering while the other is much longer and arises from scattering that occurs close to the surface of the crystal. This scattering has now been observed at a variety of structural phase transitions and at magnetic phase transitions in the rare-earth metals and actinides [1–6] and these results have been summarized in a review [7]. Despite this experimental activity there has been far less theoretical progress in understanding the origin of the second and longer-length-scale component of the critical scattering. Altarelli and co-workers [8] have suggested that there are defects at the surfaces of the crystals which are associated with long-range strain fields and that these induce a crossover to a new disordered fixed point. An alternative viewpoint [7] is that the effect is intrinsic to ideal materials and occurs because of the effect of the surface on the critical fluctuations. The order parameter couples non-linearly to the strain but above the transition temperature the strain is effectively clamped in the bulk of the crystal and can follow the fluctuations only close to the surface. Neither of these explanations is as yet fully convincing. In the former case, the defects have not been identified for any of the materials, and it is unexplained why the phenomena are so similar in metals and insulators

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at both magnetic and structural phase transitions, while in the latter case the theory has not been developed quantitatively.

The neutron and x-ray scattering experiments described in this paper were performed to provide more information about the nature of this long-length-scale scattering. Experiments to study the magnetic scattering from bulk holmium [3, 4] were the first to show the existence of two length scales at a magnetic phase transition and are still some of the most detailed studies of the phenomena. We have chosen to study the magnetic transition of a thin film of holmium grown by molecular beam epitaxy. The film was grown on a seed of an yttrium–lutetium alloy with a concentration chosen to provide lattice matching with the holmium layer. The film was then capped by another similar alloy film with a thickness larger than that of the holmium film. This sample was designed so as to reduce the strain occurring at the interfaces of the holmium film and also to clamp the holmium film so that the magnetic material did not have a free surface and so the magnetic fluctuations could not couple to the elastic surface waves. The results are compared with similar measurements made on a holmium film grown on a 2400 Å yttrium seed and capped with a 30 Å yttrium layer [9].

The next section of the paper describes the properties of the holmium film and the experimental measurements. The results are described in the third section and discussed in a final section.

2. The experiments

The sample was grown by molecular beam epitaxy (MBE) using the LaMBE Facility, Oxford [10]. A 1000 Å Nb (110) film was deposited on a sapphire substrate at a temperature of 1100 K. The substrate temperature was then reduced to 600 K and an alloy of yttrium and lutetium was deposited with a thickness of 6660 Å. The concentration of the alloy was chosen such that the basal-plane lattice parameter of the alloy would be the same as that of bulk Ho at a temperature just above the magnetic ordering temperature. The alloy grew with the (001) planes perpendicular to the growth direction. A layer of holmium (001) was then grown with a thickness of 5000 Å and followed by a capping layer with the same concentration of yttrium and lutetium as that of the seed and a thickness of 10 000 Å. *In situ* RHEED measurements showed that the basal-plane lattice parameters of the alloy and holmium differed while they were growing. Nevertheless detailed x-ray measurements of the sample *ex situ* showed that the seed layer, holmium layer and capping layer all had the same basal-plane lattice constant to an accuracy of $\pm 0.01\%$. This is different from the behaviour of most of the rare-earth films and superlattices for which the seed and capping layers have basal-plane lattice constants that differ from those of the magnetic material. At room temperature the basal-plane lattice parameter was measured as 3.5730 Å, while the lattice parameter c of the alloy was 5.6370 Å, and that of the holmium was 5.6205 Å. The corresponding lattice parameters for bulk holmium are 3.578 Å for the basal plane and 5.618 for the c-axis. The mosaic spreads were measured as 0.073° for the alloy and 0.061° for the Ho film, both of which are less than those observed for other rare-earth materials grown by the same techniques.

The neutron scattering measurements were carried out at the NBSR BT7 reflectometer located at the National Institute of Standards and Technology Center for Neutron Research. The incident neutron energy was fixed at 14.8 meV using the (002) reflection of a vertically focused pyrolytic graphite monochromator. Higher-order neutrons were suppressed with a pyrolytic graphite filter. The BT7 reflectometer is similar to a two-axis diffractometer equipped with three adjustable slits, two in front of the sample and one behind. The sample

was mounted in a closed-cycle cryostat with the [001] direction aligned along the translation axis of a motorized goniometer stage and measurements were made of the scattering near the $(0, 0, q)$ reflection arising from the helical magnetic order [11]. The measurements exploit the excellent resolution available for small scattering wave vectors [9, 12].

The magnetic x-ray scattering measurements were performed with the X22C beamline of the National Synchrotron Light Source at Brookhaven National Laboratory. This beamline has a doubly focusing nickel-coated mirror, and a fixed-exit Ge(111) double-crystal monochromator. The scattered x-rays were measured using the (111) reflection of Ge with an energy resolution of about 10 eV. The sample was mounted in a closed-cycle cryostat with the (001) face in a vertical scattering geometry. The incident x-ray energy, 8.07 keV, was chosen so as to maximize the magnetic x-ray scattering. The measurements were made by scanning the wave-vector transfer near the $(0, 0, 2 - q)$ and $(0, 0, 2 + q)$ reflections.

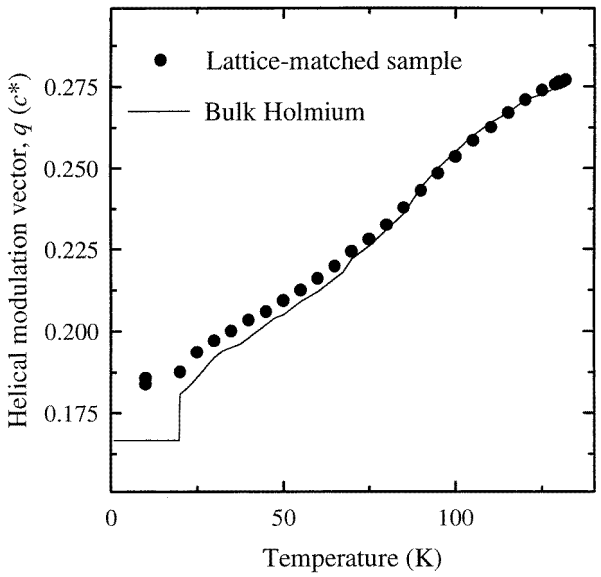

Figure 1. The temperature dependence of the modulation wave vector of the helical magnetic structure of a lattice-matched Ho film. The solid line shows the corresponding wave vector for bulk holmium [4].

3. Experimental results

The temperature dependence of the wave vector describing the helical magnetic ordering is shown in figure 1 where it is compared with similar measurements for bulk holmium [4]. Above about 60 K there is excellent agreement between the results for the film and the bulk samples but at lower temperatures the results for the film are slightly larger. After cooling rapidly from above the ordering temperature to below 20 K, two peaks were observed corresponding to wave vectors of $0.1849 = 22/119\ \boldsymbol{c}^*$ and $0.1833 = 11/60\ \boldsymbol{c}^*$, while after cooling more slowly only one peak was observed with a wave vector of $0.1833\ \boldsymbol{c}^*$. These wave vectors are larger than were observed at low temperatures for the cone phase of bulk holmium [11], but smaller than those observed for holmium films grown on yttrium seed

layers [9, 13]. Figure 2 shows the temperature dependence of lattice parameters a and c for the lattice-matched film and of the alloy substrate and cap. The lattice parameters a are within error identical for the holmium film and for the alloy films and independent of temperature. The lattice parameter c of the holmium film increases with decreasing temperature and increasing magnetic order while that of the alloy decreases with decreasing temperature in a similar manner to that for bulk lutetium or yttrium. Both lattice parameters c increased with increasing temperature between 135 K and room temperature.

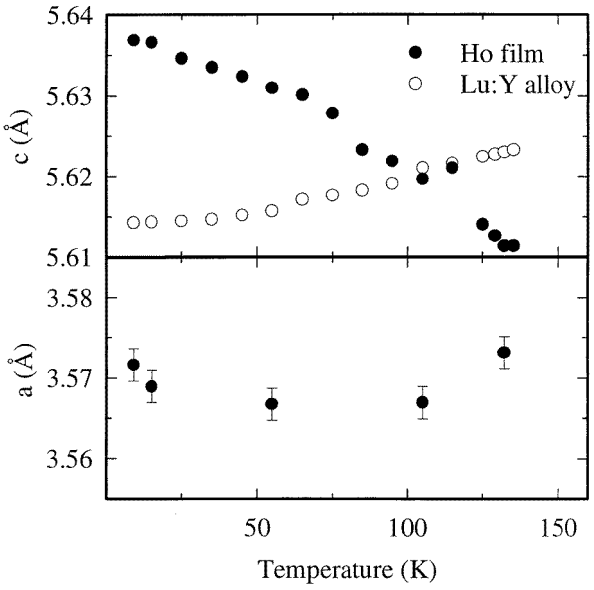

Figure 2. Lattice parameters as a function of temperature for a holmium film grown between films of a lattice-matched lutetium–yttrium alloy. The lattice parameters a of the alloy and holmium films are the same.

Close to T_N the wave vector of the helical ordering was 0.2770 $\boldsymbol{c}^*$, as shown in figure 1. The temperature dependence of the intensity of the magnetic scattering was measured using both x-ray and neutron scattering techniques. In the former case, scans were performed varying the wave-vector transfer radially through the $(0, 0, 2-q)$ magnetic reflection, while for the neutron scattering the wave-vector transfer was varied both radially and transversely through the $(0, 0, q)$ magnetic reflection. The observed intensity was then fitted to obtain the integrated intensity of the peak and the results for the temperature dependence of the intensity are shown in figure 3. These results were then fitted to a power law:

$$I(T) = A(T_N - T)^{2\beta}$$

and the results of the x-ray measurements gave the values $T_N = 131.04 \pm 0.05$ K and $\beta = 0.345 \pm 0.045$. The results from the neutron scattering results depended on the range of the temperatures used in the fits. Agreement between the data from the radial and transverse scans was obtained when the data between 129 K and 131.5 K were fitted and the results were $T_N = 131.61 \pm 0.02$ K and $\beta = 0.390 \pm 0.040$. The difference between the values of T_N probably arises from the calibrations of the thermometers in the two different cryostats. The x-ray data were refitted holding the value of $\beta = 0.39$, and the sum of the squares of the deviations doubled over that obtained when β was allowed to vary. We

conclude that the best estimate of β is 0.370 ± 0.03. This result is consistent with the value of β (0.37–0.39) found from experiments on bulk Ho [4, 14]. It is inconsistent with the value of 0.5 found in experiments on other Ho films [9, 13] which have a similar film thickness but are not lattice matched to the substrate or cap and have a cap thickness of only 300 Å [9] or 30 Å [13]. These films have a negligible clamping of the long-wavelength fluctuations while the larger value of β is possibly associated with the more intense long-length-scale scattering that is observed for these films.

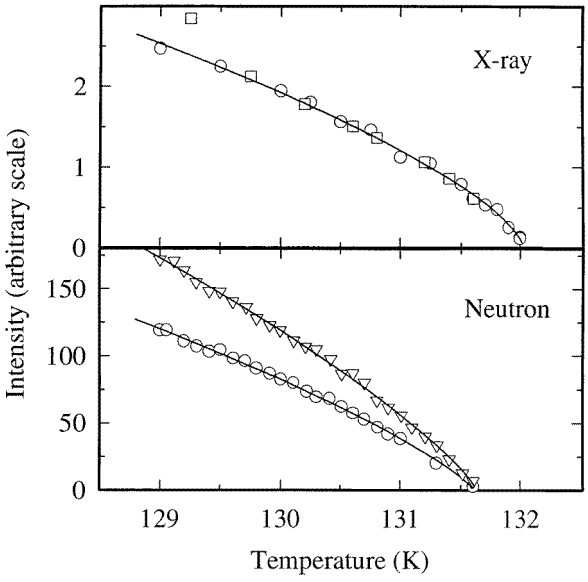

Figure 3. The temperature dependence of the magnetic Bragg scattering from a lattice-matched Ho film as measured with x-ray and neutron scattering techniques. The circles and squares for the x-ray measurements are the results of two different experiments, and for the neutron scattering measurements the circles are from longitudinal measurements and the triangles from transverse measurements. The solid lines are the results of power-law fits as described in the text and the two measurements give different values for T_N probably because of the calibration of the two different thermometers.

The neutron and x-ray scattering was measured for temperatures just above T_N. Figure 4 shows the neutron scattering observed at 0.22 K above T_N when the wave-vector transfer is scanned transversely through the (0, 0, q) magnetic Bragg reflection. The solid line in figure 4 shows a fit to the scattering with an anisotropic Lorentzian profile convoluted with a Gaussian approximation to the resolution function [15]. The ratio of the inverse correlation lengths in the basal plane and along the c-axis was assumed to be the same as for bulk holmium, 1.38 [4], while the parameters of the resolution function were obtained from the shape of the Bragg peak below T_N. The fit gives a chi-squared of 1.48 and the Lorentzian has a FWHM in the basal plane of 0.013 ± 0.001 Å^{-1}. Fits were also performed to the sum of a Lorentzian and an anisotropic Lorentzian-squared form. The ratio of the basal-plane and c-axis parameters for the Lorentzian squared was fixed at 0.78 [4]. The chi-squared for this model reduces to 1.22 and the FWHM of the Lorentzian increases to 0.019 ± 0.002 Å^{-1} while that of the Lorentzian squared is 0.0015 ± 0.0006 Å^{-1}. The width of the Lorentzian component deduced from measurements on bulk Ho [4] was 0.004 Å^{-1}, and the extrapolation of the power law obtained at high temperatures to 0.22 K above

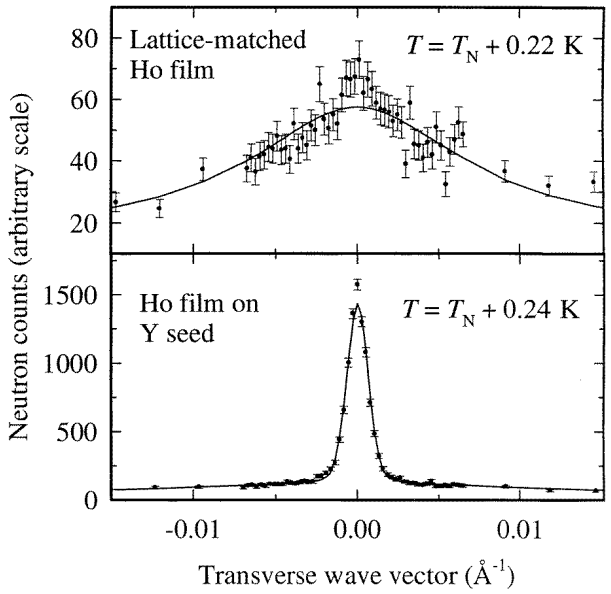

Figure 4. The neutron scattering observed in a transverse scan through the $(0, 0, q)$ magnetic Bragg reflection. The upper part is from the lattice-matched film 0.22 K above T_N, and the lower from a film on an yttrium seed 0.24 K above T_N. The solid line in the upper part shows a fit to a Lorentzian form convoluted with the experimental resolution and with a constant background.

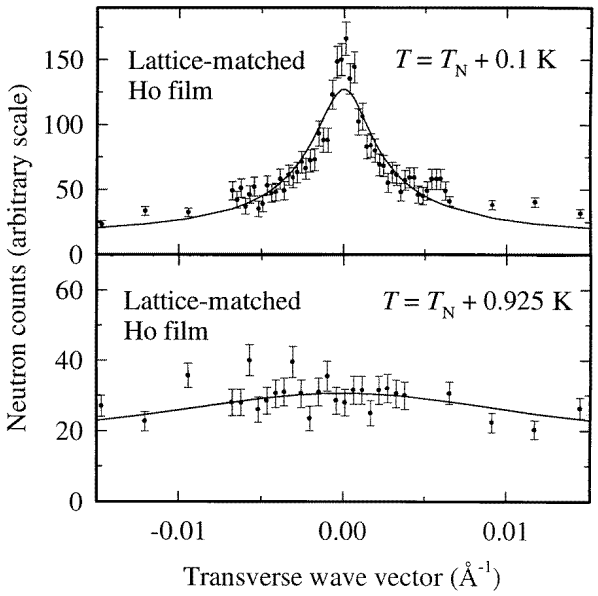

Figure 5. The neutron scattering observed in transverse scans through the $(0, 0, q)$ magnetic Bragg reflection at 0.10 K and 0.925 K above T_N. The solid lines show fits to a Lorentzian form convoluted with the experimental resolution and with a constant background. The large-wave-vector parts of the lower diagram have been omitted from the figure.

T_N was 0.016 Å^{-1}. Assuming that the broad Lorentzian arises from conventional critical scattering, the width of the Lorentzian component is expected to be the same in the samples suggesting that the fits to the Lorentzian and Lorentzian-squared profiles overestimate the contribution of the Lorentzian-squared component in the lattice-matched film. In figure 4 we also compare the results for the lattice-matched film with our earlier results for a holmium film on an yttrium substrate [9]. The results for the scattering are very different and in the previous experiment the two-component nature of the scattering is clearly visible. The ratio of the peak heights of the narrow and broad components of the scattering is 15 ± 2. In contrast there are not clearly two different components in the data from the lattice-matched film and if fits are used to estimate the contribution of the two components to the profile shown in the upper part of figure 4, the ratio of the peak heights is 0.75 ± 0.2. We conclude that the data shown in figure 4 have a relatively much smaller Lorentzian-squared component than was observed in our earlier experiment with a film on an yttrium substrate [9]. The new data suggest that there is a reduction in the intensity of the Lorentzian-squared component by a factor of at least 20 and are probably consistent with the absence of any Lorentzian-squared component. A similar conclusion can be drawn from a comparison of the scattering observed 0.6 K below T_N and 0.22 K above T_N. In the present experiment the peak count rate decreases by a factor of about 240, whereas in the $(0, 0, q)$ neutron scattering experiment from bulk Ho the ratio was about 28. In our experiment at 0.22 K above T_N the amplitude of the Lorentzian-squared component is less than 0.75 of the observed scattering. If the difference in the two ratios is a result of the decrease in the amplitude of the Lorentzian-squared component, then this decrease is by a factor of about 22.

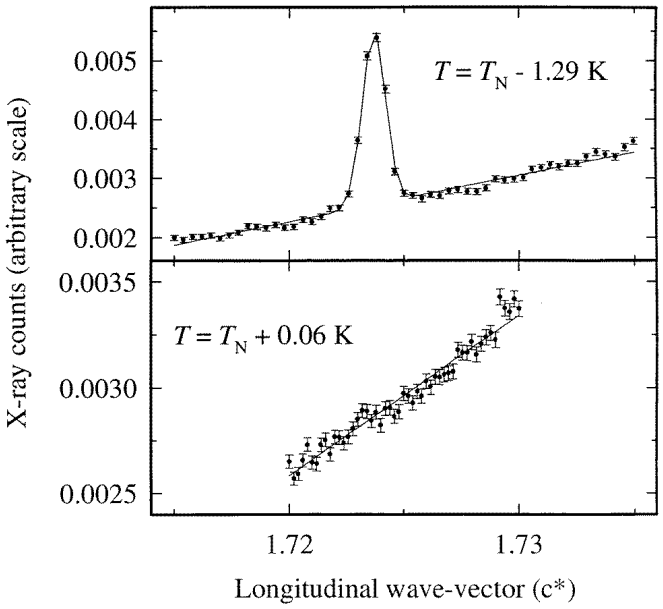

Figure 6. The x-ray scattering observed in a longitudinal scan through the $(0, 0, 2-q)$ magnetic Bragg reflection at 1.29 K below and 0.06 K above T_N. The solid lines show fits to a sloping background and in the case of the upper diagram a Gaussian peak.

Similar experiments were performed at temperatures of 0.1 K and 0.91 K above T_N. At the higher temperature the data, figure 5, are consistent with a single-Lorentzian peak

having a width consistent with the Lorentzian profile observed at the same temperature for bulk Ho [4]. The lower-temperature results are shown in figure 5 where again the solid line is a fit to a Lorentzian profile. This fit gives a chi-squared equal to 2.33 and the Lorentzian FWHM is 0.0022 ± 0.0003 Å^{-1}. This is larger than the width of the Lorentzian observed for bulk Ho [4] but smaller than the extrapolation of the high-temperature power law. The fit shown in figure 5 is improved if a Lorentzian-squared form is added in the fitting procedure. Chi-squared is reduced to 1.53 while the width of the Lorentzian peak increases to 0.013 Å^{-1}. This is substantially larger than the Lorentzian width for the bulk when it is extrapolated from high temperatures [4], and so this fit probably overestimates the size of the Lorentzian-squared component. Given the uncertainties of the fitting procedure and the difficulty of ensuring that the temperature of the sample is uniform and constant close to T_N these fits do not provide clear evidence for a two-component line shape. We conclude that the neutron scattering data are consistent with the narrow component of the scattering being either absent or at least 20 times smaller than was observed in other similar experiments using a film [9] or bulk Ho [3, 4].

X-ray scattering measurements above T_N with films are made difficult by the charge scattering from the truncation rods associated with the film, niobium and sapphire interfaces. This gives the large background streak parallel to $\boldsymbol{q}$, shown in figure 6, so when the wave-vector transfer is scanned perpendicular to $\boldsymbol{q}$ an intense peak is observed and the weak magnetic scattering is not measured reliably. We have therefore performed longitudinal scans of the wave-vector transfer but there is inevitably a large background preventing very accurate measurements from being made. Figure 6 shows the scattered x-ray intensity at temperatures 1.29 K below and 0.06 K above T_N. The former shows a magnetic Bragg peak whereas the latter has no significant peak. A series of detailed fits produced an estimate of the maximum peak intensity that is consistent with the data 0.06 K above T_N. This maximum is 0.009 of the intensity observed 1.29 K below T_N. Figure 1 of reference [4] suggests that the ratio of the scattered x-ray intensity from bulk holmium 0.23 K above T_N to that observed 1.1 K below T_N is 0.06. Furthermore, this ratio is increased to about 0.12 if the temperature is 0.06 K above T_N. Comparing this ratio with that obtained in the present experiments suggests that the x-ray scattering from our film is at least 13 times weaker than that observed from bulk holmium. Since the x-ray scattering is dominated by the narrow component of the critical scattering this means that this component is at least 13 times weaker in our film than in the bulk material.

4. Summary and conclusions

High-resolution neutron and x-ray scattering measurements have been made of the magnetic phase transition of a thin film of holmium. The single-crystal film is 5000 Å thick and grown on a lattice-matched lutetium–yttrium alloy and capped by 10 000 Å of a similar alloy. The results show that the temperature dependence of the order parameter is given by the exponent $\beta = 0.37 \pm 0.03$ in agreement with recent measurements on bulk holmium [4, 14], but in disagreement with other measurements on thin films [9, 13]. Above T_N the measurements show that the critical scattering with a long length scale is very much reduced in comparison with its intensity in similar experiments [3, 4, 9]. Our measurements are consistent with a reduction in its intensity by a factor of at least 20 and are not inconsistent with the complete absence of the long-length-scale component. The difficulty of separating this scattering from the Bragg scattering close to T_N may be why the earlier measurements on thin films [9, 13] gave a value of the exponent β of about 0.5.

Previous measurements on a number of different systems have shown that the sharp

component of the critical scattering is associated with the surface of the sample. In the experiments reported here, the magnetic surface of the thin film is a buried interface between the magnetic holmium layer and the non-magnetic lutetium–yttrium alloy. The absence or near absence of the narrow component in these experiments suggests that the long-length-scale component is associated with the structural surface and not the magnetic surface. Surface relaxation and reconstruction is usually restricted to the first few atomic layers [16] and so cannot produce a new very long length scale for the critical fluctuations [7]. The result is consistent with the observations that show that the narrow part of the scattering arises from a strained part of the crystal that must be at the structural surface [2].

Unfortunately our results do not directly distinguish between the two possible explanations for the observation of the sharp component of the critical scattering. If this scattering is associated with defects at the surface of the crystal then the growth of the film between thick lattice-matched alloy films might result in a reduction in the number of relevant defects at the surface of the magnetic layer. Alternatively, if the scattering is associated with the coupling between the order parameter and the free strains at the surface, the thick cap clamps the upper surface of the magnetic layer inhibiting its response to the order parameter fluctuations. The theory of surface waves suggests that this clamping would occur for fluctuations of a length scale shorter than the thickness of the cap, 10 000 Å. The temperature dependence of the width of the narrow component suggests that the length scale of the narrow component in the bulk is shorter than this when the temperature is more than 0.05 K above T_N. Although our experiments do not distinguish between the two explanations, the mosaic spread of our Ho film is about 0.06° whereas the bulk samples [4] had a smaller mosaic spread. If the sharp critical scattering arises from defects it is then surprising that this scattering is absent or much smaller in the film with a mosaic spread which is larger than that in the bulk samples in which the scattering is observed.

Acknowledgments

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