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A study of the magnetic critical scattering from the longitudinally modulated antiferromagnets thulium and erbium

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Abstract. The temperature dependence of the magnetic critical scattering above T_N and the staggered magnetization below T_N has been measured in the longitudinally modulated antiferromagnets thulium and erbium using neutron diffraction. The transitions, which occur at $T_N = 57.65 \pm 0.10$ K in thulium and at $T_N = 86.04 \pm 0.20$ K in erbium appear to be continuous second-order transitions. As longitudinally modulated antiferromagnets thulium and erbium would be expected to belong to the $d = 3$, $n = 2$ universality class of the $d = 3$ XY-model. From an analysis of the critical scattering the exponents $\nu_c = 0.43 \pm 0.02$, $\nu_{ab} = 0.40 \pm 0.02$ and $\gamma = 0.90 \pm 0.04$ have been deduced for thulium and $\nu_c = 0.41 \pm 0.04$, $\nu_{ab} = 0.35 \pm 0.05$ and $\gamma = 0.73 \pm 0.06$ for erbium. These values are different to the theoretical values for the $d = 3$ XY-model of $\nu = 0.65$ and $\gamma = 1.3$. However, from the temperature dependence of the staggered magnetization the exponents $\beta = 0.37 \pm 0.03$ for thulium and $\beta = 0.35 \pm 0.03$ for erbium have been determined which are consistent with the theoretical value of $\beta = 0.35$ for the $d = 3$ XY-model. It is speculated that the difference above T_N between theory and experiment could arise if the exchange interaction were temperature dependent.

1. Introduction

It is well known that continuous magnetic phase transitions can be classified according to universality classes [1]. The parameters which define a universality class include d, the spatial dimensionality and n, the number of components of the order parameter. For values of n less than 4 it is believed [2] that the critical exponents describing the static properties of the phase transition only depend upon the values of d and n. This means that transitions with what appear to be quite dissimilar natures should be governed by the same set of critical exponents. This is the case for the paramagnetic to longitudinally modulated antiferromagnetic transitions of thulium and erbium, which should belong to the universality class of the $d = 3$ XY-model.

The transition in thulium is known to occur at about $T_N \approx 56$–58 K [3, 4] and that in erbium at $T_N \approx 85$–86 K [5]. In the ordered state just below T_N the magnetic moments in these elements are along the direction of the hexagonal c-axis (both thulium and erbium have the HCP structure) but the size of the magnetic moment

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varies sinusoidally with a wavevector τ which is oriented along the c^*-axis. Although the staggered magnetization (order parameter) in thulium and erbium is uniaxial the phase transition belongs to an $n = 2$ universality class because the number of components of the star of the ordering wavevector (i.e. τ) is 2. The value of n is defined as the number of spin degrees of freedom multiplied by the number of components of the star of the ordering wavevector [6]. The best known model system with $n = 2$ is the XY-model, in which the magnetic moments are constrained to lie in the x–y plane but are free to rotate isotropically in that plane.

In this paper we report the results of neutron diffraction measurements of the magnetic critical scattering associated with the paramagnetic to longitudinally modulated antiferromagnetic transitions in thulium and erbium. We also report results for the temperature dependence of the staggered magnetization in the critical region.

2. Experimental details

Neutron scattering measurements on both thulium and erbium are difficult because they have large absorption cross sections for thermal neutrons. The crystal of erbium used was a special isotopic sample of ^{170}Er which has the lowest neutron absorption cross section. This crystal was a semi-circular-shaped button, of radius 14 mm and thickness 8 mm. While the a^* reciprocal lattice vector was in the plane of the button the c^* reciprocal lattice vector was at approximately 45° to the plane. The crystal of thulium used was in the shape of a thin plate of dimensions 7.2 mm by 4.1 mm by 0.9 mm and was one of the crystals used by Fernandez-Baca *et al* [4] in their spin-wave measurements. The c-axis of the crystal was perpendicular to the plate. In both sets of measurements the crystals were oriented with their $(H, H, 0)$ and $(0, 0, L)$ reciprocal lattice directions in the scattering plane.

The crystals were contained in aluminium sample cans which were filled with helium exchange gas. These cans were then attached to the copper block of a modified closed cycle refrigerator. The modification to the refrigerator was to add an extra mass of 100 g of lead to the the copper block in order to increase the thermal mass. This produced stable temperatures when in equilibrium but also made it slow to change temperature.

The neutron diffraction measurements were performed at the High Flux Isotope Reactor, Oak Ridge National Laboratory using the HB1A and HB2 triple-axis spectrometers. For all of the critical scattering measurements the spectrometers were operated in a two-axis mode with the analyser crystal removed. The monochromator on HB1A is a double-bounce system consisting of two pyrolitic graphite crystals utilizing the $(0, 0, 2)$ planes for scattering with a fixed incident neutron energy of 14.75 meV. The HB2 spectrometer has a conventional single-bounce pyrolitic graphite monochromator and was operated with an incident neutron energy of 14.8 meV. For the measurements on both spectrometers a pyrolitic graphite filter was placed in the incident beam to remove contaminant neutrons scattered from higher-order planes in the monochromators. The beam collimation used on HB1A was 20′ monochromator to sample and 20′ sample to detector and that used on HB2 was 10′ and 10′ respectively.

3. Results

In the first subsection we report the results of the staggered magnetization measurements and in the second subsection the results of the critical scattering measurements.

3.1. Staggered magnetization

In both thulium and erbium we have measured the temperature dependence of the intensity of the magnetic $(1, 1, \overline{\tau})$ Bragg peak. In figures 1(*a*) and 1(*b*) we show the measured integrated intensity through the Bragg peak position by solid circles for temperatures below T_N and by crosses for temperatures above T_N. The solid lines in these figures are the results of fitting the data below T_N to the power law equation

$$I = I_0 t^{2\beta} \tag{1}$$

where $t = |T - T_N| / T_N$ is the reduced temperature and β is the critical exponent describing the staggered magnetization [1]. Initially both T_N and β were allowed to vary in the fits. However, the values of T_N deduced were very similar to the values deduced from power law fits of the parameters describing the critical scattering above T_N. As a consequence average values of T_N were obtained and the data re-fitted with T_N fixed at these average values. These latter fits lead to the values $\beta = 0.37 \pm 0.03$ and $T_N = 57.65 \pm 0.10$ K for thulium and $\beta = 0.35 \pm 0.03$ and $T_N = 86.04 \pm 0.20$ K for erbium. We note that the initial free-fit values of β and T_N lie within the ranges quoted in the error bars.

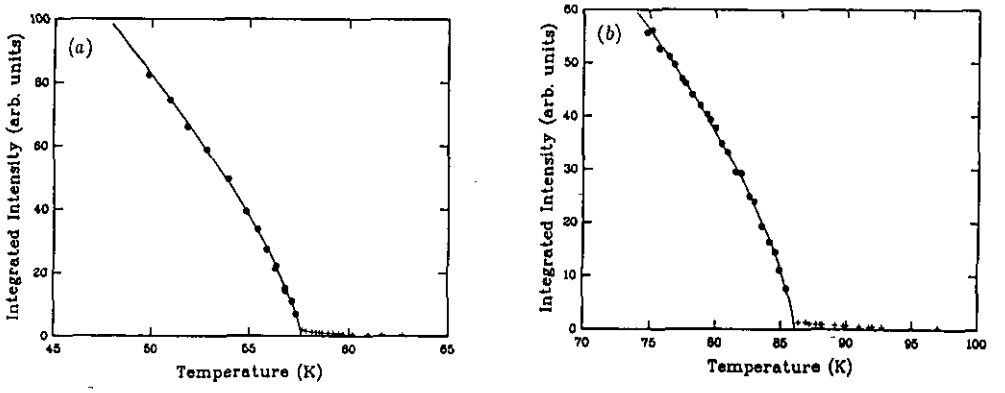

Figure 1. The temperature dependence of the integrated intensity measured in a scan through the position of the $(1, 1, \overline{\tau})$ magnetic Bragg peak are shown (*a*) for thulium and (*b*) for erbium. The solid circles indicate data taken below T_N, the crosses data taken above T_N and the solid lines are the power law fits to the data below T_N as described in the text.

These values for β appear to agree well with the theoretical value of $\beta = 0.35$ for the $d = 3$ XY-model [1]. However there are two effects which could mean that the precise values are less reliable than the error bars would indicate. The first is the effect of extinction on the Bragg peak intensity. This is an especially complicated

effect in a critical phenomena measurement because the extinction parameters are dependent upon the value of the staggered magnetization which undergoes a large variation in the temperature range not far below the critical point. As a consequence any effect due to extinction can be assumed to be highly temperature dependent and virtually impossible to quantify accurately. In erbium and thulium this may be a less serious effect than in other materials because of the relatively large absorption cross sections which, in principle, should reduce the effect of extinction. The second effect which may perturb the value of β is the effect of the critical scattering below T_N. The experimentally measured intensity is the sum of the scattering due to the Bragg peak and the structure factor corresponding to the critical scattering below T_N, both convoluted with the experimental resolution function. The separation of these two terms in an experiment is very difficult in practice. Well below T_N the critical scattering is very weak compared to the Bragg peak, while close to T_N where it may make a more significant contribution to the intensity it is very narrow and difficult to separate from the Bragg peak lineshape. In order to give some idea of the relative contribution made by this effect we have shown by the crosses in figures 1(*a*) and 1(*b*) the integrated intensity measured above T_N. In mean-field theory and in model systems where theoretical calculations have been performed, such as the $d = 2$ and $d = 3$ Ising models, the intensity of the critical scattering below T_N is less than that above T_N. Consequently the experimentally measured intensity above T_N sets an upper limit to the possible contribution made by the critical scattering at the equivalent temperatures below T_N. Unfortunately without a precise knowledge of the ratio of the critical amplitudes above and below T_N it is not possible to subtract a fraction of this scattering from the overall intensity measured.

In light of the discussion above it is therefore necessary to be cautious in concluding that the β values agree with the theoretical value for the $d = 3$ XY-model. It would however be very surprising if either of the effects discussed above were to make substantial changes to the measured β values, for example changing them from 0.37 to 0.50. In conclusion therefore it seems that the measured values of β are consistent with erbium and thulium belonging to the universality class of the $d = 3$ XY-model.

3.2. *Magnetic critical scattering*

In order to characterize the critical scattering around T_N we have performed scans in a two axis mode through the $(1, 1, \bar{\tau})$ magnetic satellite position along the $(1, 1, L)$ and $(H, H, \bar{\tau})$ directions. These scans were carried out at various temperatures between 57 K and 63 K for thulium and 86 K and 98 K for erbium. Further scans were carried out at 149.7 K for both thulium and erbium to assess the background level in the critical scattering measurements.

The magnetic structure of thulium and erbium is such that only the longitudinal staggered susceptibility χ^{zz} should diverge at T_N and the transverse staggered susceptibilities (χ^{xx} and χ^{yy}) should not. In the scans described previously around $(1, 1, \bar{\tau})$ the scattering geometry is such that all three susceptibilities will in principle be observed. Therefore in order to assess the contributions of χ^{xx} and χ^{yy} in these scans we have also carried out scans through the position $(0, 0, 2 + \tau)$ along $(0, 0, L)$ and $(H, H, 2 + \tau)$ at a number of temperatures for both thulium and erbium. At the position $(0, 0, 2 + \tau)$ scattering from χ^{zz} is forbidden because the wavevector transfer of the neutron is parallel to the direction of the staggered magnetization. It should be noted that the neutron scattering cross section is such that scattering can only occur

from components of the magnetization perpendicular to the wavevector transfer [1]. Hence the magnetic scattering around $(0,0,2+\tau)$ should only be due to χ^{xx} and χ^{yy}. These scans were, within the counting statistics, independent of temperature and the intensity of the scattering measured was consistent with the background level measured around $(1,1,\bar{\tau})$ in both thulium and erbium. It can therefore be concluded that χ^{xx} and χ^{yy} do not make a significant contribution to the observed scattering at $(1,1,\bar{\tau})$ compared to that from χ^{zz}.

In figures 2(*a*) and 2(*b*) we show the results of scans along $(1, 1, L)$ at 62.7, 59.5 and 57.9 K for thulium and at 97.0, 91.0 and 87.2 K in erbium respectively. The background which, as described earlier, was measured at high temperature is shown as a dashed line in these figures. The critical scattering has been fitted to a Lorentzian lineshape $S(Q)$ given by

$$S(Q) = \chi \Big/ \left(1 + \left(\frac{Q_c - \tau}{\kappa_c}\right)^2 + \left(\frac{Q_{ab}}{\kappa_{ab}}\right)^2\right) \tag{2}$$

where Q_c and Q_{ab} are wavevector components along the c^* axis and in the plane perpendicular to c^* respectively, κ_c and κ_{ab} are the inverse correlation lengths in those directions and χ is the staggered susceptibility. The Lorentzian was convoluted with the resolution function, which was measured from the $(1, 1, \bar{\tau})$ satellite Bragg peak below T_N. In the thulium measurements the major axis of the resolution function within the scattering plane was found to be oriented at 17.68° to $(H, H, 0)$ direction with a full width at half maximum (FWHM) of 0.0404 Å^{-1}. The FWHM of the minor and vertical axes of the resolution function were 0.0322 Å^{-1} and 0.1025 Å^{-1} respectively. In the erbium measurements the major axis of the resolution function was along the $(H, H, 0)$ direction with a FWHM of 0.0425 Å^{-1}. Along the minor and vertical axes the FWHM was 0.0123 Å^{-1} and 0.1297 Å^{-1} respectively. In figures 2(*a*) and 2(*b*) the solid lines are the best fit results to equation (2) at these temperatures. At all of the temperatures studied equation (2) gave a good fit to the data with a normalized weighted least squares value close to 1.

In figures 3(*a*) and 3(*b*) we show the squares of the best fit parameters κ_c and κ_{ab} and in figures 4(*a*) and 4(*b*) the parameter χ^{-1} as a function of temperature. Figures 3(*a*) and 4(*a*) are the results for thulium and 3(*b*) and 4(*b*) for erbium. The reason for plotting κ^2 against temperature rather than κ will become apparent in the discussion section. The solid lines in figures 3(*a*), 3(*b*), 4(*a*) and 4(*b*) are the result of power law fits to the equations

$$\kappa_c = \kappa_c^+ t^{\nu_c} \tag{3}$$

$$\kappa_{ab} = \kappa_{ab}^+ t^{\nu_{ab}} \tag{4}$$

$$\chi = \chi^+ t^{-\gamma} \tag{5}$$

As for the power law fits of the staggered magnetization we initially allowed the value of T_N to vary in these fits. However, because the resulting values were similar to each other and to those deduced from the staggered magnetization, the data were re-fitted with the average value of T_N kept fixed. In table 1 the resulting critical exponents and transition temperatures are listed. The initial free-fit results lie within the range of the error bars quoted in table 1.

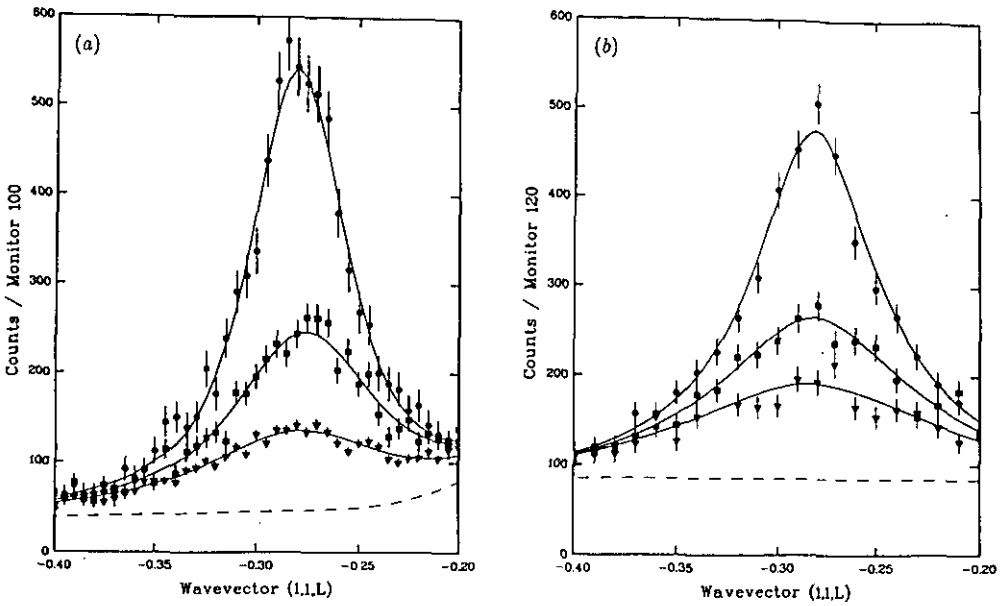

Figure 2. The results of scans along $(1, 1, L)$ through the satellite positions are shown (a) for thulium at 57.9 K (circles), 59.5 K (squares) and 62.7 K (triangles), and (b) for erbium at 87.2 K (circles), 91.0 K (squares) and 97.0 K (triangles). The solid lines are the fits to equation (2) convoluted with the resolution function as described in the text.

4. Discussion

As described in the introduction we anticipated at the outset of this study that thulium and erbium would belong to the same universality class as the $d = 3$ XY-model (the $n = 2$ class). The critical exponents ν, γ and β for the $d = 3$ XY-model are also listed in table 1 for comparison with the experimental results. There is substantial disagreement between the values of ν and γ we have found for thulium and erbium, and those of the $d = 3, n = 2$ class, while the experimental value of β is consistent with the theoretical value.

The values of ν and γ obtained experimentally are very surprising. This is not just because they are different to the $d = 3$ XY-values but because they are smaller than the mean-field values of $\nu = 0.5$ and $\gamma = 1.0$. It might be suspected that the observed values of ν and γ have arisen because of a cross-over to mean-field results away from the $d = 3$ XY-values. There are three arguments against this being an explanation for the values of the exponents ν and γ. Figures 3(a) and 3(b) were deliberately plotted in the unusual form of κ^2 against T because the mean-field value of ν is 0.5 and consequently in a temperature range where mean-field behaviour was relevant a plot of κ^2 against T should be a straight line. It should be noted that mean-field theory not only predicts a different value for the critical exponent but also a different value for T_N which could lead to difficulty in recognizing a mean-field region. However, this problem is overcome by plotting κ^2 against T where mean-field behaviour should identify itself by a straight line dependence which extrapolates to the mean-field value for T_N. The same argument applies to figures 4(a) and 4(b) where the data in a plot of the inverse susceptibility χ^{-1} against T should in any

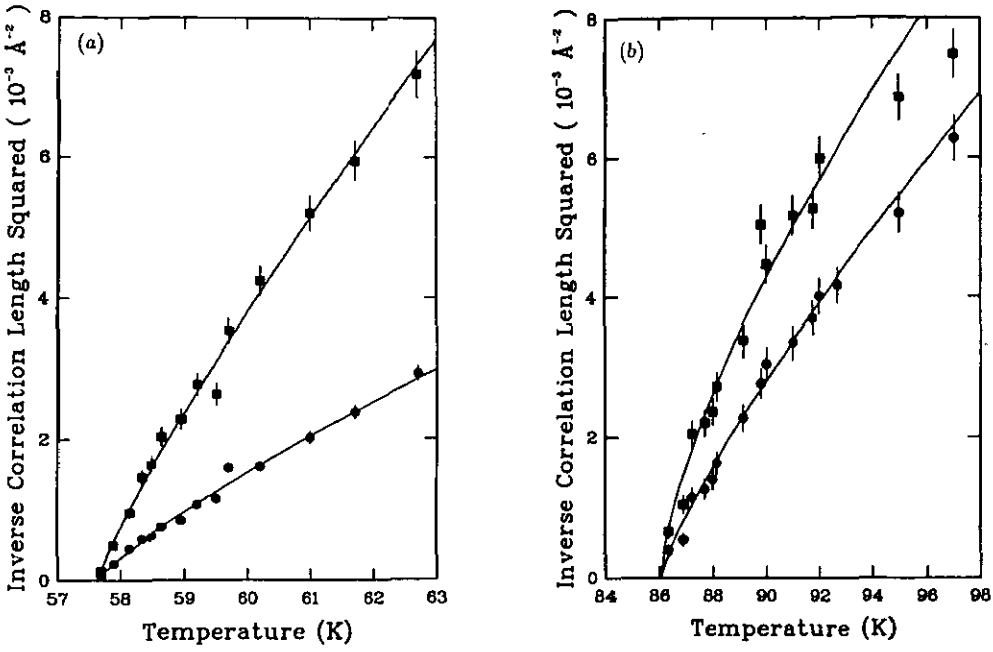

Figure 3. The temperature dependence of the squares of the inverse correlation lengths obtained from the fits to the critical scattering above T_N in (*a*) thulium and (*b*) erbium is shown. The solid circles refer to the values of κ_c and the solid squares to κ_{ab}. The solid lines are the results of power law fits as described in the text.

mean-field regime of temperature follow a straight line, extrapolating to the effective mean field value of T_N, because the mean-field value of the exponent γ is 1. Secondly, even accounting for a possible change in the effective value of T_N, it is difficult to see how a crossover from an exponent of $\nu = 0.65$ to $\nu = 0.5$ could lead to the observed values of $\nu \approx 0.4$. We note that such a crossover would appear in figures 3(*a*) and 3(*b*) as a convex function, whereas the data has a concave appearance. A similar argument applies for the value of the susceptibility exponent γ. Thirdly the observed values of the exponent β for thulium and erbium are both consistent with the $d = 3$ XY-model value. This agreement appears to hold over a range of reduced temperature extending out to $t \approx 0.14$ below T_N in both thulium and erbium. In our critical scattering measurements above T_N the range of reduced temperatures we have studied is $t \leqslant 0.14$, a similar range to that below T_N. It would seem unlikely that the critcal region below T_N would be many times larger (if larger at all) than that above T_N. As a consequence it seems unlikely that the values of ν and γ result from a mean-field crossover effect.

At this time we do not have any explanation for these results which can be fully justified on experimental or theoretical grounds. However, there is a possibility which could, at least qualitatively, explain these results. If the exchange interaction in thulium and erbium were to decrease with increasing temperature above T_N then this could effect the measured values of κ and χ in a way that leads to the behaviour we have observed. We have estimated that the exchange would have to have fallen by up to 15% at a reduced temperature of $t = 0.1$ in order to account for the

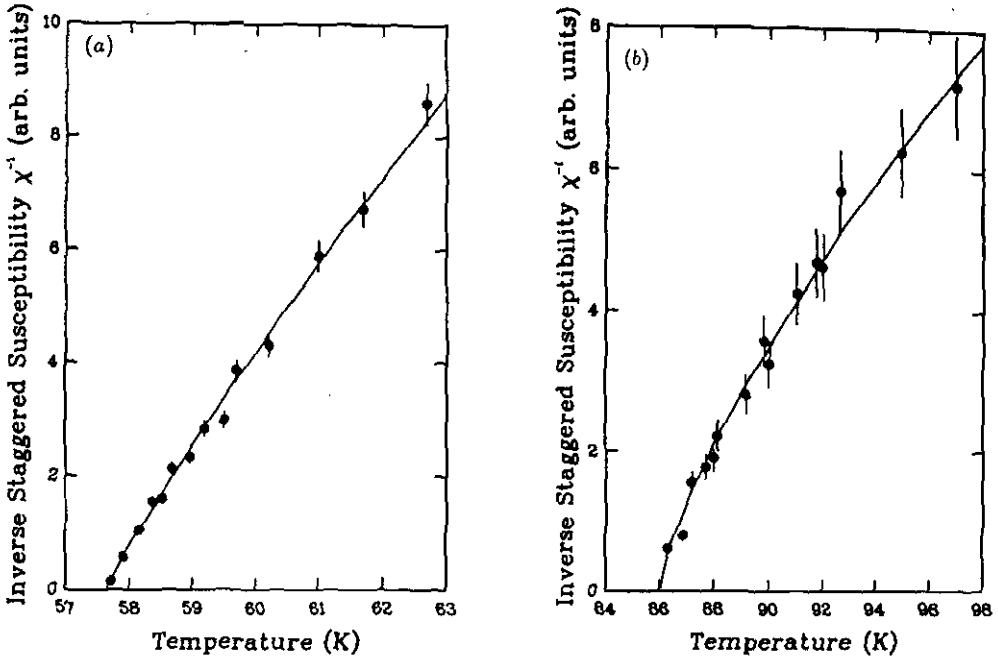

Figure 4. The temperature dependence of the inverse of the staggered susceptibility χ^{-1} obtained from fits to the critical scattering above T_N is shown for (*a*) thulium and (*b*) erbium. The solid lines are the results of power law fits as described in the text.

Table 1. The values of the critical exponents β, ν_c, ν_{ab}, and γ and the corresponding values of T_N found from the power-law fits described in the text are given for thulium and erbium. Also given for comparison are the theoretical values for the $d = 3$ XY-model which were taken from [1].

	Thulium	Erbium	$d = 3$ XY-model
T_N	57.65 ± 0.10	86.04 ± 0.20	—
	$T < T_N$		
β	0.37 ± 0.03	0.35 ± 0.03	0.35
	$T > T_N$		
ν_c	0.43 ± 0.02	0.41 ± 0.04	0.65
ν_{ab}	0.40 ± 0.02	0.35 ± 0.05	0.65
γ	0.90 ± 0.04	0.73 ± 0.06	1.30

observed behaviour. In appendix A we give a derivation of this estimate and the assumptions involved. The possibility that the exchange interaction in thulium might be temperature dependent has been speculated upon by McEwen *et al* [7] when examining the low-temperature magnetic excitations. In another rare earth, holmium, it is known from an analysis of the spin-wave spectra that the exchange interaction does have an explicit temperature dependence [8] below T_N. There is, however, a need for caution before accepting this explanation. The effect in holmium occurs below T_N and it is not clear that it is of a sufficient size to account for the effects we have observed in thulium and erbium. Another aspect which also suggests caution is the fact that the measured β exponents are consistent with the $d = 3$ XY-model.

If the exchange interaction varied with temperature below T_N then this would be expected to have a significant effect on the magnetization. Since this does not appear to happen it must be assumed that the exchange does not vary as strongly below T_N as above. Further work is required to resolve this question and it would be very interesting to know whether the specific heat capacity exponents above and below T_N agreed with the $d = 3$ XY-model or not.

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Appendix A

If it is assumed that the exchange interaction above T_N in thulium and erbium is dependent upon temperature then it is possible to estimate the amount of change from the experimental results subject to an assumed dependence of the critical amplitude on the exchange. This can be done by equating the observed temperature dependence of the inverse correlation length to an inverse correlation length given by the 'true' critical exponent with an effective Néel temperature which is temperature dependent. This is given by

$$\overline{\kappa^+}\left(\frac{T - T_N}{T_N}\right)^{\overline{\nu}} = \kappa^+\left(\frac{T - T_N^*}{T_N^*}\right)^{\nu} \tag{A1}$$

where $\overline{\kappa^+}$, $\overline{\nu}$ and T_N are the experimentally observed critical amplitude, exponent and Néel temperature, and κ^+, T_N^* are the temperature-dependent critical amplitude and Néel temperature and ν is the theoretical value for the $d = 3$ XY-model. It should be noted that κ^+ and T_N^* are temperature dependent because the exchange is (assumed to be) temperature dependent. The dependence of T_N^* on the exchange is linear; however, the dependence of κ^+ is in general unknown. In mean-field theory the critical amplitude κ^+ is proportional to the square root of the exchange and therefore we have assumed this to be the case for our estimate. Furthermore we have assumed that the constants of proportionality in the relations $\overline{\kappa^+} \propto \sqrt{T_N}$ and $\kappa^+ \propto \sqrt{T_N^*}$ are the same. Under these conditions equation (A1) can be re-written as

$$\varepsilon + \varepsilon^{(2\nu-1)/2\nu} t^{\overline{\nu}/\nu} = 1 + t \tag{A2}$$

where $t = |T - T_N|/T_N$ is the reduced temperature and $\varepsilon = T_N^*/T_N$. Since the Néel temperature is linearly proportional to the exchange, ε is also the ratio of the exchange at temperature T to the value at T_N. In figure A1 we show the numerical solution of equation (A2) for ε as a function of t from 0.0 to 0.1 using the experimental value of $\overline{\nu} = 0.4$ and the values of $\nu = 0.65$ (solid line) and 0.5 (dashed line) which would be appropriate for the $d = 3$ XY-model and mean-field theory respectively. From figure A1 it can be seen that at $t = 0.1$ the exchange would have to be 15% smaller than at T_N in order to account for the observed value of ν rather than the $d = 3$ XY-value.

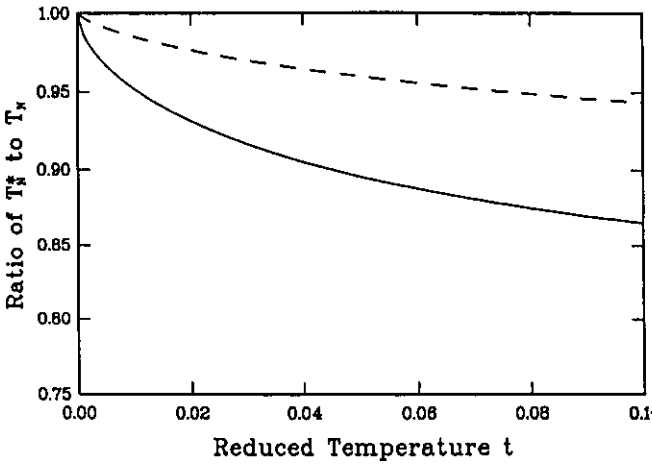

Figure A1. The value of ε, the solution of equation (A2), is shown as a function of reduced temperature t, for the cases where $\nu = 0.65$ by the solid line and $\nu = 0.50$ by the dashed line.

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