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ANOMALOUS MAGNETIC CRITICAL BEHAVIOR OBSERVED IN NEUTRON SCATTERING FROM HoSb AND DySb*

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Neutron diffraction measurements on HoSb and DySb show an anomalously large intensity from magnetic critical scattering above a well-defined Néel point T_N. The origin of this unusual critical behavior above T_N is not understood at present, although it appears related to strong magnetoelastic coupling which for both systems results in a cubic-to-tetragonal lattice distortion at T_N.

THE RARE EARTH pnictides are an interesting class of metallic magnets since most of the compounds exhibit a structural transformation at low temperatures in addition to magnetic ordering. Recent measurements on HoSb and DySb have shown evidence of anomalous critical behavior near the antiferromagnetic ordering temperature T_N. Specific heat measurements on HoSb[1] give unusually large values of the critical exponents α and α' suggestive of tricritical behavior.[2] Moreover, the temperature derivative of the electrical resistance R obeys the same power law divergence above T_N as the specific heat C_p.[1,3] In DySb, specific heat and neutron scattering measurements[4] have shown a first order transition with the magnetic and structural phase transitions occurring at the same temperature. These experiments were supported by resistivity measurements showing a step-discontinuity with hysteresis at T_N.[5,6] However, despite the first order nature of the transition, the resistance also showed downward curvature above T_N characteristic of critical scattering of electrons as observed in GdSb and HoSb.[3]

It has been suggested[1,3] that the anomalous critical behavior displayed by these rare earth pnictides is associated with strong magnetoelastic coupling which results in a cubic-to-tetragonal lattice distortion at T_N.[4,7] Alternatively, it is conceivable that such anomalous results are not critical phenomena at all, and the large exponents are actually effective exponents obtained by forcing a power law divergence to a first order transition which has been broadened by chemical and structural inhomogeneities. In this note, we establish by neutron diffraction measurements on HoSb and DySb the existence of an anomalously large intensity from magnetic critical scattering above a *well-defined* T_N. We will show that in several respects our results for HoSb are inconsistent with the explanation of a first order transition which has been broadened by sample inhomogeneities.

HoSb and DySb are semimetals of the cubic rock-salt structure above T_N. Below T_N they are type 2 antiferromagnets[8,9] with moments nearly parallel to a [100] direction and with ferromagnetic sheets perpendicular to a [111] direction. The samples used in the present investigation were single crystals ~ 3 × 1 × 1 mm (mosaic spread $\lesssim 0.5°$ in the cubic phase) cut from the same ingots as the resistivity and specific heat samples.[1,3] Measurements were performed on both double and triple axis spectrometers

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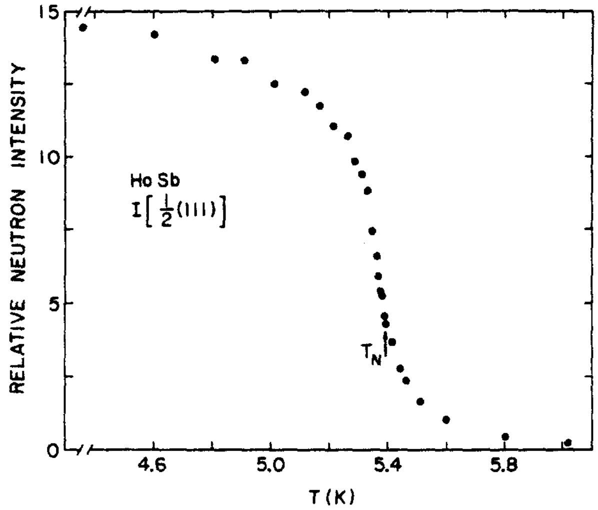

FIG. 1. Intensity of ½ (111) magnetic peak vs temperature for HoSb. Arrow indicates T_N as determined by the abrupt onset of the structural transformation (Fig. 2).

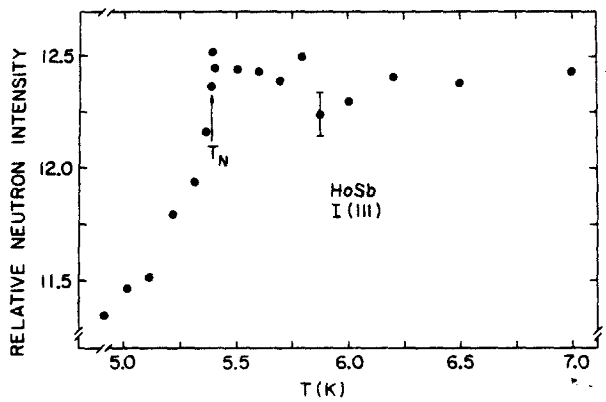

FIG. 2. Intensity of the (111) nuclear peak of HoSb vs temperature.

at the Brookhaven High Flux Beam Reactor with a filtered incident neutron beam of energy 13.5 meV. In the experiments the horizontal collimation was 20 min throughout and the vertical collimation was determined by the sample size and the 2 in. vertical dimension of the slit system. The monochromator and analyzer were bent and flat pyrolytic graphite, respectively, having a mosaic spread of 0.4° full width at half maximum.

In Fig. 1 we have plotted the scattered intensity at the 1/2 (111) magnetic superlattice point vs temperature for HoSb as measured on the 2-axis spectrometer. A similar curve was obtained from an $E = 0$, q-scan on the 3-axis spectrometer. Unlike a typical magnetization curve, there is a very large tail to the intensity which is observed up to ~ 5 K above T_N. An inflection point occurs near $T_N \simeq 5.4$ K where the intensity is ~ 25 per cent of the extrapolated $T = 0$ value. For comparison, in $RbMnF_3$, which is regarded as an ideal Heisenberg antiferromagnet, the intensity at the 1/2 (111) magnetic superlattice point at T_N is only a few per cent of the $T = 0$ value.[10] From the ratio of the nuclear (200) and magnetic 1/2 (111) peaks of HoSb at 4.2 K, we estimate a saturation moment of about $10\mu_B$ per Ho^{3+} ion in reasonable agreement with the value of $9.3\mu_B$ reported earlier.[8] Therefore, the relatively large magnetic intensity at T_N cannot be attributed to a failure of attaining the full Ho^{3+} moment at low temperatures. Despite this large scattering, the intensity of the magnetic 1/2 (111) peak reproduced well and showed no hysteresis in slow temperature sweeps in opposite directions.

We also monitored the structural transformation in HoSb by measuring the intensity of the nuclear (111) reflection near T_N on the 3-axis spectrometer as shown in Fig. 2. There is a sudden, though continuous, change in intensity at $T = 5.39 \pm 0.01$ K which agrees with the inflection point in the magnetic intensity (Fig. 1) and also T_N as determined from the peak in C_p and the maximum in dR/dT.[1,3] The integrated intensity of the (111) nuclear reflection remained constant in the 2-axis scans in this temperature range and we conclude that the abrupt change in peak intensity is due to structural domains and twinning of the crystal at the onset of the tetragonal distortion. Thus the structural transition temperature T_S is determined quite well and is equal to T_N as determined from the macroscopic measurements. In addition, the sharpness of the structural transition suggests that chemical and strain inhomogeneities do not smear the transition. If the large magnetic intensity above $T_N = T_S$ were due to transformed regions of the sample, we would expect the structural transition to be similarly broadened.

To establish that the anomalously large magnetic intensity observed above T_N is critical scattering, we measured the temperature dependence of the q-width of the 1/2 (111) peak in 2-axis and 3-axis $E = 0$ scans.

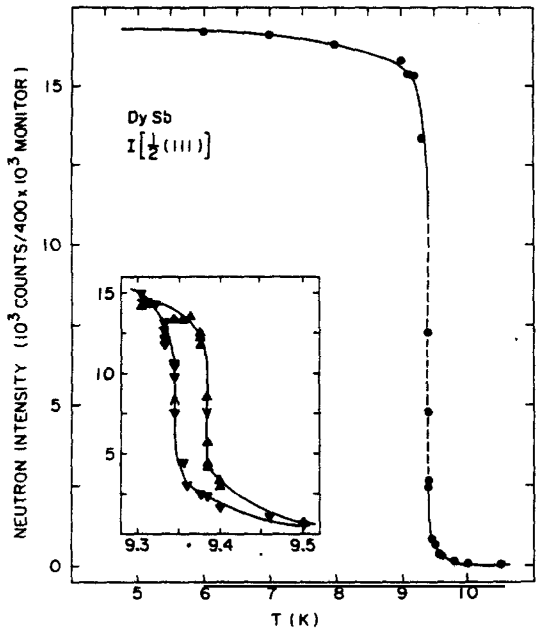

FIG. 3. Intensity of ½(111) magnetic reflection of DySb vs T. Insert shows hysteresis near T_N: (▲) cooling; (▲) heating.

In both cases, we found that the full q-width at half maximum (FWHM) is determined by the spectrometer resolution at $T = 5.4$ K, but it increases by ~ 25 per cent at 7.0 K. This is consistent with a correlation length for magnetic critical fluctuations which increases as T_N is approached from above. If a broadened first order transition were occurring, the magnetic intensity at high temperatures would be due to a Bragg peak from the transformed region of the sample and the line width would be temperature independent. For $(T - T_N) \lesssim 3$ K, the energy width of the 1/2(111) peak was accounted for by the 0.08 meV resolution of the 3-axis spectrometer.

Although the q-width of the 1/2(111) peak increases upon heating above T_N, the peak remains strikingly narrow even at $T = 7.0$ K $[\epsilon = (T - T_N)/T_N = 0.3]$. It is of interest to compare the intrinsic q-width of the ~~critical scattering~~ at $\epsilon = 0.3$ with that of the classical antiferromagnet $RbMnF_3$. In order to obtain the intrinsic q-width, we have fitted the line shape observed in a 3-axis scan through 1/2(111) ($\vec{q} \parallel [110]$ to a Lorentzian folded with the instrumental resolution function calculated by Cooper and Nathans.[12] At $\epsilon = 0.3$ we estimate the width of the Lorentzian $\kappa \lesssim 2 \times 10^{-3}$ Å^{-1}.[13] The corresponding lower bound on the correlation length ξ is $\xi = 2\pi/\kappa \approx 6000$ Å or about 1000 lattice spacings. This value of ξ is about a factor of 100 greater than that obtained for $RbMnF_3$ at the same ϵ.[10] We emphasize that we have only made an order of magnitude estimate of κ for HoSb but that this is sufficient to demonstrate the dramatically narrower q-width of the critical scattering as compared with $RbMnF_3$.

As a final check that the large magnetic intensity above 5.4 K results from magnetic critical scattering, we measured the neutron intensity as a function of temperature at the reciprocal lattice point 1/2(0.94, 0.94, 1). This point is sufficiently far from the superlattice reflection 1/2(111) to avoid detecting Bragg scattering below T_N, but is is close enough to 1/2(111) to detect critical scattering of small momentum transfer. As the temperature was varied through T_N we observed a sharp peak in intensity which is characteristic of critical scattering in magnetic systems.

DySb, in contrast to HoSb, has a step discontinuity with hysteresis in the intensity of the 1/2(111) magnetic reflection at $T_N \simeq 9.36$ K as shown in Fig. 3.[14] Nevertheless, just as in HoSb there is a relatively large magnetic intensity above T_N which can be observed for $(T - T_N) \lesssim 4$ K. We have also verified in DySb that the q-width of the 1/2(111) peak increases upon heating above T_N. In 3-axis scans along the [110] direction the FWHM is determined by the spectrometer resolution at 9.5 K and increases by ~ 25 per cent at 10 K.

The neutron scattering experiments reported here have provided additional evidence of anomalous critical behavior near the magnetic-structural phase transition in HoSb and DySb. In the case of HoSb the possibility of a broadened first order transition in the absence of critical effects has been ruled out. Since we now have available the results of neutron, ultrasonic,[15,16] specific heat[1,4] and resistivity[3,5,6] measurements, it seems useful to conclude by emphasizing the basic features of the phase transition in HoSb and DySb. The unusual magnetic critical behavior appears to be associated with the *simultaneous* structural transformation of the solid. In the related compound GdSb which has no observable tetragonal distortion,[6] the specific heat and resistivity exhibit critical behavior typical of metallic magnets.[1,3] In addition to the magnetic critical effects above T_N which we have discussed here, there is a pronounced softening of the elastic constant $1/2\,(c_{11} - c_{12})$ preceding the tetragonal distortion in DySb[15] and HoSb.[16] Thus there may be local fluctuations in the interatomic distance as well as the magnetization above T_N. The fact that the magnetic and structural transitions occur together suggests the presence of a strong magnetoelastic interaction coupling magnetic and displacive fluctuations of the ions. It would be desirable to perform inelastic neutron experiments on larger samples than presently available in order to elucidate the dynamical aspects of this phase transition. Finally, we note that while our measurements have been confined to HoSb and DySb, it is likely that the an anomalous critical behavior would be common to other rare earth pnictides which have simultaneous structural and magnetic ordering.

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