

goes an order-disorder phase transition from CsCl to bcc structure. As a result the two Λ modes must become degenerate at T_c , and since the gradients $\nabla\omega_\Lambda$ of these bands are of opposite signs at 296°K, they must go through zero at T_c , or, in other words, they become flat there. Calculations of $g(\omega, T)$ as well as $C_\nu(T)$ for β -brass are already in progress.¹²

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SOFT SPIN-WAVE MODES AND THE CONE-TO-SPIRAL TRANSITION IN HOLMIUM METAL

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It is suggested that the cone-to-spiral magnetic transition in holmium metal near 20°K is caused by a soft spin-wave mode of nonzero wave vector. Neutron inelastic scattering measurements are presented which support this proposal.

The existence of a soft spin-wave mode, analogous to the soft phonon mode in a ferroelectric,¹ may be inferred from the form of the spin-wave dispersion law for holmium. For the conical phase this may be written²

$$\hbar\omega(\vec{q})/S = [J(\vec{k}_0 + \vec{q}) - J(\vec{k}_0 - \vec{q})] \cos\theta + (F_1 F_2)^{1/2}, \quad (1)$$

where

$$F_1 = 2J(\vec{k}_0) - J(\vec{k}_0 + \vec{q}) - J(\vec{k}_0 - \vec{q}),$$

$$F_2 = [2J(\vec{k}_0) - J(\vec{k}_0 + \vec{q}) - J(\vec{k}_0 - \vec{q})] \cos^2\theta + 2[J(\vec{k}_0) - J(\vec{q}) - K(0) + L] \sin^2\theta,$$

and

$$L = 6K_4 S^2 \cos^2\theta + 15K_6 S^4 \cos^4\theta.$$

S is the total angular momentum, $J(\vec{q})$ is the Fourier transform of the exchange interaction $J(\vec{r}_{ij})$, \vec{k}_0 is the wave vector of the periodic magnetic structure, and $K(0)$, K_4 , and K_6 are two-, four-, and sixfold anisotropy parameters, respectively. The dispersion relation for the spiral phase may be obtained by putting the cone angle θ equal to $\frac{1}{2}\pi$. For the conical phase the exchange and an-

isotropy constants are related by the cone equilibrium condition²

$$3K_6 S^4 \cos^4\theta + 2K_4 S^2 \cos^2\theta + J(\vec{k}_0) - J(0) - K(0) = 0. \quad (2)$$

Since $J(\vec{q})$ has its maximum value when $\vec{q} = \vec{k}_0$, the term F_1 in Eq. (1) is necessarily positive for nonzero values of \vec{q} . For the spiral phase the term F_2 is a minimum when $\vec{q} = \vec{k}_0$, when it reduces simply to $-K(0)$. Hence $K(0)$ must be negative if this spin-wave mode and the spiral structure itself are to be stable. For the conical phase the constant K_4 may be eliminated from L by using the cone equilibrium condition (2), and the corresponding condition on $K(0)$ for the stability of this mode may be written

$$K(0) > \frac{3}{2}[J(\vec{k}_0) - J(0)] - 3K_6 S^4 \cos^4\theta - \frac{1}{4} \cot^2\theta [2J(\vec{k}_0) - J(0) - J(2\vec{k}_0)].$$

K_6 may be calculated from crystal field theory.^{3,4} Although its exact value depends on the details of the model used, the term $3K_6 S^4 \cos^4\theta$ is probably very small and negative in sign. The last term is therefore the only one which gives a negative

contribution to the complete expression. However it is of order $\frac{1}{3}\cot^2\theta$ smaller than the first term, and since the cone angle for holmium is about 79° ,⁵ it too can be neglected. In the conical phase, we therefore have the condition

$$K(0) > \frac{3}{2}[J(\vec{k}_0) - J(0)]$$

for the mode $\vec{q} = \vec{k}_0$ and the conical structure itself to be stable. This condition on the stability of the mode $\vec{q} = \vec{k}_0$ is in fact more stringent than that on the stability of the conical structure and of spin waves of small wave vector obtained from the cone equilibrium condition.²

The energy of a rare-earth ion in a hexagonal crystal field may be expanded in terms of the spherical harmonics Y_2^0 , Y_4^0 , Y_6^0 , and Y_6^6 ,^{6,7} and the first three of these contribute to the two-fold axial anisotropy $K(0)S_\zeta^2$. According to a calculation by Kasuya⁴ the dominant contribution at 0°K arises from Y_6^0 and is positive in sign, in agreement with the discussion given above, and there is also a significant negative contribution from Y_2^0 . Callen and Callen⁸ have shown that the contribution due to a term Y_{2l}^0 varies with temperature as the $l(2l+1)$ th power of the ordered atomic moment. $K(0)$ should therefore decrease rapidly with increasing temperature as the positive contribution from Y_6^0 decreases in magnitude with respect to the negative contribution from Y_2^0 . At some temperature the value of $K(0)$ will no longer satisfy the inequality (3) and the conical structure will become unstable. The transformation to a stable spiral structure requires that $K(0)$ should now change discontinuously to zero, and then become negative on further increasing the temperature. A discontinuous change in $K(0)$ can arise from discontinuous changes, at the phase transition, in quantities such as the ordered moment and lattice parameters on which the crystal field parameters themselves depend. Féron and Pauthenet⁹ have measured the field dependence of the c -axis magnetization over a wide range of temperatures, and there is evidence from their results that the anisotropy parameters change sharply in the neighborhood of the phase transition.

In this model, therefore, the spin wave with wave vector \vec{k}_0 may be expected to become unstable at the transition temperature. We have carried out neutron inelastic scattering measurements on a single crystal of holmium in order to search for this instability, using a triple axis spectrometer at the Chalk River NRU reactor in

the constant \vec{Q} mode of operation.¹⁰

It was not possible to make measurements on the mode $\vec{q} = \vec{k}_0$ because the corresponding neutron groups could not be resolved from the strong elastic background, but by using focusing techniques we were able to observe a mode of slightly higher energy at $\vec{q} = (0.2\pi/a, 0.2\pi/a, k_0)$. Measurements were made at a series of temperatures in the range from 4.6 to 48°K, and the temperature variation of the energy of this mode is given in Fig. 1. The transition temperature for our specimen, as determined from the temperature dependence of the magnetic contribution to the (110) Bragg reflection,⁵ was $22.5 \pm 0.5^\circ\text{K}$.

Figure 1 shows a striking decrease of about 35% in the spin-wave energy between 4.6°K and the transition temperature, followed by a slight rise in energy, with a weak maximum at about 40°K. It is possible that this variation may contain a contribution from the temperature dependence of the exchange constants $J(\vec{r}_{ij})$. However, analysis of our earlier measurements of the complete c -axis dispersion curves,¹¹ and of similar measurements by Nicklow et al.,¹² suggests that the exchange constants decrease by not more than 10% on increasing the temperature to 48°K. Almost the whole of the spin-wave energy variation in this temperature range may therefore be attributed to variation of the anisotropy parameters.

Because $J(\vec{k}_0) - J(\vec{q})$ is not zero for the mode $(0.2\pi/a, 0.2\pi/a, k_0)$ the term F_2 in Eq. (1) contains

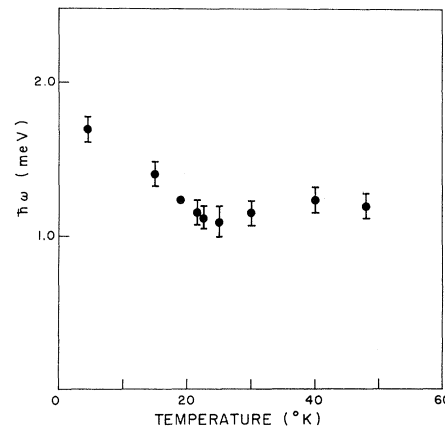


FIG. 1. The points give the measured energy $\hbar\omega$ of the spin-wave mode $(0.2\pi/a, 0.2\pi/a, k_0)$ in holmium metal at a series of temperatures in the range from 4.6 to 48°K. The errors shown correspond to the uncertainties in the energies relative to the energy at 19°K.

a nearly constant exchange contribution, and so this term, and thus the spin-wave energy, will not go to zero with the anisotropy contribution on approaching the transition temperature.

In summary, we have proposed a model in which the cone to spiral transition in holmium is caused by an instability of the spin-wave mode of wave vector \vec{k}_0 arising from the strong temperature dependence of the anisotropy parameters. Calculations of these parameters lend support to our arguments, and we have carried out neutron inelastic scattering measurements which demonstrate the influence of this instability on a spin-wave mode of wave vector near \vec{k}_0 .

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EFFECT OF THE MOLECULAR FIELD ON THE ELECTRICAL RESISTIVITY NEAR A MAGNETIC TRANSITION: GdNi_2 †

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We have measured the temperature dependence of the electrical resistivity of the cubic, Laves-phase, ferromagnetic, intermetallic compounds GdNi_2 , GdPt_2 , and GdRh_2 , and for GdNi_2 , we have analyzed the temperature derivative of the electrical resistivity in the neighborhood of the magnetic transition. Above the Curie temperature our data are very well described by the molecular-field treatment of the long-range spin fluctuations of the short-range order, giving for the first time an experimental result in agreement with this treatment.

For some time there have existed theoretical treatments by de Gennes and Friedel¹ and Kim² of the effect of short-range spin ordering on the electrical resistivity near a magnetic transition. To date, there have not been any experimental data which can be reasonably described in terms of these theories. These studies have used a molecular-field or Ornstein-Zernicke³ approximation for the long-range behavior of the spin-spin correlation function and the Born approximation to relate this correlation function to the electrical resistivity. The basic model used is that of an "s-d" exchange interaction between the conduction (s) electrons and the assumed lo-

calized d or f electrons. Recently, the measurements of Craig et al.⁴ on the electrical resistivity of Ni near its Curie point showed a behavior very different from that predicted by these theories. Fisher and Langer⁵ have argued that these theories are inadequate because the anomalous behavior of the electrical resistivity near a magnetic critical point is determined largely by the short-range rather than the long-range spin fluctuations, at least for temperatures above the transition temperature T_c . In this paper we report the observation of the effect of the long-range nature of the critical fluctuations of the short-range spin ordering on the tempera-