

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-

ture dependence of the electrical resistivity of GdNi₂, a cubic, Laves-phase, ferromagnetic, intermetallic compound, and demonstrate that the molecular-field theories of de Gennes and Friedel¹ and Kim² give a good semiquantitative fit to our data for temperatures above T_c .

We have measured the temperature dependence of the electrical resistivity of a series of cubic, Laves-phase, gadolinium intermetallic compounds and have analyzed, in some detail, the case of GdNi₂ which shows the most striking anomalous behavior near the ferromagnetic-ordering temperature. The saturation magnetization and Curie temperature of GdNi₂ were measured independently by Skrabek and Wallace⁶ and Crangle and Ross.⁷ Our measurements of the electrical resistivity were made by the standard four-point probe technique on a sample that had been annealed at 1000°C for one week. Data were taken at 1°K intervals throughout the critical region. The temperature derivative of the resistivity $d\rho/dT$ was obtained from the $\rho(T)$ data by point-by-point differentiation using a computer.

In Fig. 1 we plot our $\rho(T)$ data and the computed $d\rho/dT$ for GdNi₂. Similar results were obtained in GdPt₂ and GdRh₂. At high temperatures $\rho(T)$ is essentially linear in T but as the temperature is lowered there is a plateau in the neighborhood of 80°K and then a further decrease to the lowest temperatures measured. The $d\rho/dT$ curve shows a very broad maximum at high temperatures and a sharp decrease until about 80°K followed by a very sharp rise over a temperature interval of a few degrees as the temperature decreases. At about 75°K there is a sharp

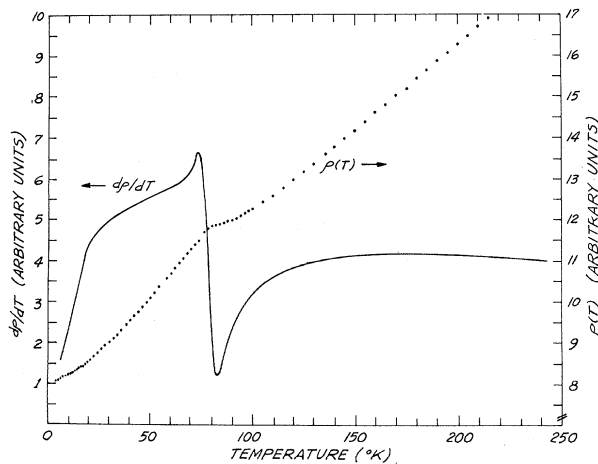


FIG. 1. $\rho(T)$ and $d\rho/dT$ for GdNi₂. $d\rho/dT$ was obtained from $\rho(T)$ data by computer differentiation.

maximum followed by a decrease in $d\rho/dT$ at lower temperatures.

The general shape of the $d\rho/dT$ curve in the neighborhood of 75°K is consistent with the $\rho(T)$ behavior predicted by the molecular-field theories of de Gennes and Friedel and Kim for the critical region, and we analyze our results using expressions derived by Kim.³ In particular, the electrical resistivity of a system due to a local s - d exchange interaction in the Born approximation is given by⁵

$$\rho = \rho_0 \frac{1}{2} \int_0^\pi \sum_I \Gamma(\vec{R}_I) \exp(i\vec{K} \cdot \vec{R}_I) \times (1 - \cos\theta) \sin\theta d\theta, \quad (1)$$

where ρ_0 is the electrical resistivity in the high-temperature limit where spin-spin correlations are negligible, θ is the scattering angle, $\hbar\vec{K}$ is the momentum transfer, and $\Gamma(\vec{R}_I)$ is the static spin-spin correlation function for two spins separated by \vec{R}_I :

$$\Gamma(\vec{R}_I) = [\langle \vec{S}_I \cdot \vec{S}_0 \rangle - \langle \vec{S}_I \rangle \cdot \langle \vec{S}_0 \rangle] / S(S+1). \quad (2)$$

Using the molecular-field approximation [essentially taking $\Gamma(R) \propto R^{-1} \exp(-\kappa R)$, where κ is the inverse range of correlation] for the spin-spin correlation function, Kim² shows that the electrical resistivity can be written as

$$\begin{aligned} \rho T > T_c \\ &= 4\rho_0 \frac{T}{T_0} \left[1 - 2 \frac{T - T_c}{T_0} \ln \left(1 + \frac{T_0}{2(T - T_c)} \right) \right], \end{aligned} \quad (3)$$

and

$$\begin{aligned} \rho T < T_c \\ &= 4\rho_0 \frac{T}{T_0} \left[1 - \frac{4}{3} \frac{T_c - T}{T_0} \ln \left(1 + \frac{T_0}{4(T_c - T)} \right) \right]. \end{aligned} \quad (4)$$

In Eqs. (3) and (4), T_0 is a temperature-independent parameter which depends on the details of the model for $\Gamma(\vec{R}_I)$. In the molecular-field approximation where $\kappa^2 = a|T - T_c|$, $T_0 \approx 8k_F^2 a^{-1}$ with k_F the Fermi momentum of the relevant conduction electrons. In any more realistic model, T_0 would depend on the band structure as well as the details of the form of $\Gamma(\vec{R}_I)$.

This theory predicts a cusp in $\rho(T)$ and an infinite discontinuity in $d\rho/dT$ at T_c with $d\rho/dT$ positive for $T = T_c^-$ and negative for $T = T_c^+$. In Fig. 2 we show several curves of $d\rho/dT$ as a function of T obtained from differentiating Eqs. (3) and (4). The parameter γ is equal to $4T_c/T_0$ and we have, for the purposes of plotting, taken

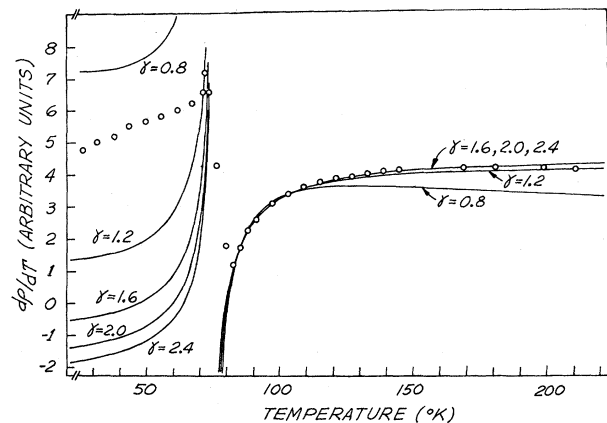


FIG. 2. Solid curves: $d\rho/dT$ due to magnetic short-range-order scattering following Kim. Circles: $d\rho/dT$ data for GdNi_2 . The theoretical curves were plotted to agree with the data at two temperatures (85 and 105°K), using $T_c = 75^\circ\text{K}$. The parameter γ is defined in the text.

$T_c = 75^\circ\text{K}$ (corresponding to what we believe is the Curie temperature of our sample of GdNi_2 , which is in good agreement with the value obtained by Crangle and Ross⁷). If one recalls that the measured resistivity includes a temperature-dependent term due to electron-phonon scattering and if one smooths the divergence in $d\rho/dT$, there is a remarkable similarity between the $d\rho/dT$ data of Fig. 1 and the theoretical curves for $d\rho/dT$ in Fig. 2, at least for temperatures above the theoretically predicted discontinuity. The fact that the measured $d\rho/dT$ never goes negative is explained by the finite electron-phonon scattering, which for the case of $\rho_{\text{phonon}} \sim T$ would shift the theoretical $d\rho/dT$ curves by a constant positive amount.

Because of the width of the transition region in $d\rho/dT$ there is some ambiguity in choosing T_c in the absence of a model for describing the width. We have therefore chosen T_c to be 75°K in GdNi_2 , about the temperature where $d\rho/dT$ shows a sharp change. This value could be in error by about 2 or 3°K corresponding to half the width of the transition region (the range over which $d\rho/dT$ drops dramatically with increasing temperature). This value of T_c is in excellent agreement with the value obtained by Crangle and Ross⁷ and is about 15°K lower than the value obtained by Skrabek and Wallace.⁶

We have also shown in Fig. 2 points for the computed $d\rho/dT$ obtained from our data on GdNi_2 using $T_c = 75^\circ\text{K}$. In order to make a comparison between our data and theory, we have added to Eq. (3) a term BT to represent the phonon con-

tribution to $\rho(T)$ and have used two temperatures (85 and 105°K) to fix the constants ρ_0 and B . The values of ρ_0 , B , and γ which best fit the data are not particularly significant. There are obvious differences between the calculated and experimental results especially below T_c , since the theory includes only the effects of short-range order. What is significant is that the temperature dependence of $d\rho/dT$ for $T > T_c$, where the short-range order of the spins should strongly perturb the electrical resistivity, is very well described by a molecular-field treatment of the spin-spin correlations as expressed by Eq. (3).

We can make the following comments about the fit of our data to the theoretical curves as shown in Fig. 2. First of all, the theory does predict a broad maximum in $d\rho/dT$ above T_c when $\gamma < 1.0$. Such a broad maximum is evident in our data (see Fig. 1). However, a fit of our data with a γ value less than unity does not appear to be possible. In fact, a choice of $\gamma \approx 1.6$ gives good agreement with the data over an interval of nearly 150°K above T_c . The explanation of this small disagreement between experiment and theory is probably due either to an insufficiency in the theory (γ is effectively temperature dependent) or to a small residual temperature dependence of the phonon contribution to $d\rho/dT$ (B is temperature dependent). Secondly, below T_c , where there is long-range order present in the spins, Eq. (4) is clearly insufficient to describe the temperature dependence of the electrical resistivity since it takes into account only the effects of short-range order in the spins. Finally, at the lower temperatures, the phonon contribution to the electrical resistivity must certainly have a temperature dependence different from BT , with B a constant. Since we have no way of separating out the electron-phonon scattering contribution from the spin-disorder scattering contribution at the lower temperatures, and indeed have no theoretical prediction of the spin-disorder scattering contribution valid over the entire temperature interval from 0°K to T_c , it would be premature to speculate about the situation in this temperature range. We can say, however, that the good agreement between our data and the molecular-field theory of Kim² for temperatures above T_c , where there can only be short-range spin order and where the electron-phonon scattering contribution to $d\rho/dT$ is probably small if not negligible, is strong confirmation of the essential correctness of the molecular-field treatment of the spin fluctua-

tions, at least in the system GdNi₂.

Our results on the cubic Laves-phase compounds GdPt₂ and GdRh₂ are similar to those of GdNi₂ although the anomalies in $\rho(T)$ and $d\rho/dT$ near T_c are not as striking as in the case of GdNi₂. We obtain values for T_c of about 23°K in GdPt₂ and 70°K in GdRh₂ compared with 36 and 74°K, respectively, obtained by Crangle and Ross.⁷

In summary, our experiment gives the first clearly quantitative agreement with the molecular-field description of the spin-disorder scattering near the critical point, and we conclude that this agreement illustrates the validity of such a theory in a system such as GdNi₂ where there is clearly a well-localized Gd moment. We believe that the difference of the critical behavior in these compounds from that in Ni⁴ or PdFe alloys⁸ probably is due to the fact that, in these gadolinium compounds, the magnetic f electrons are well localized. Therefore, a local "s-d" exchange interaction, a feature common to the theories discussed earlier,^{1,2,5} is a better representation of the true situation than in the case of Ni or PdFe alloys where the d electrons are not well localized. One is led to make a distinction⁹ between the statistical treatment of the problem and the model interaction used. It is quite possible that the major source of the qualitative difference between theory and experiment lies not so much with the details of the sta-

tistical treatment of the critical spin fluctuations as with the model interaction used to describe the scattering of the conduction electrons.

There is clearly a need for further theoretical study of this point.

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TWO-MAGNON SCATTERING OF NEUTRONS

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We report the experimental observation of the inelastic scattering of slow neutrons by two-magnon processes and present theoretical calculations which account for the shape and wave-vector dependence of the spectrum.

Two-magnon processes in antiferromagnets have recently attracted considerable interest not only in infrared absorption¹ and Raman scattering^{2,3} experiments, but also in theoretical studies. Elliott et al.^{4,5} have shown that the shape of the two-magnon spectrum observed in these optical experiments is influenced by magnon-magnon interactions, and Fleury⁶ has strikingly confirmed their predictions in RbMnF₃. In this Letter we report the observation of neutron inelastic scattering by pairs of magnons in the antiferromagnet cobalt fluoride. Neutron scattering differs from optical experiments because the wave-vector dependence of the scattering may be observed, and because two-magnon scattering arises from the correlation between pairs of spins rather than from the four-spin correlations sug-