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Electrical Resistivity near the Magnetic Transition of Some Rare-Earth Laves Phase Compounds*

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From the measured temperature dependence of the electrical resistivity $\rho(T)$, we have computed the temperature derivative $d\rho/dT$ with particular emphasis on its behavior in the critical region for six ferromagnetic rare-earth cubic Laves phase compounds: $GdAl_2$, $GdCo_2$, $GdNi_2$, $GdPt_2$, $GdRh_2$, and $DyAl_2$. All samples are characterized by pronounced changes in $d\rho/dT$ in the vicinity of the magnetic ordering temperature T_c. In $GdNi_2$, $GdPt_2$, and $GdRh_2$, as the temperature is decreased toward T_c, a minimum in $d\rho/dT$, followed by a maximum, is observed. This behavior is consistent with that predicted from the long-range nature of the short-range spin fluctuations in the framework of the molecular field theory. The behavior of $d\rho/dT$ for $GdAl_2$ and $DyAl_2$ shows only a broad maximum, whereas for $GdCo_2$, $d\rho/dT$ has a sharp peak at about T_c – very similar to that found in transition metals.

I. INTRODUCTION

Studies of the magnetic properties of rare-earth metals and their intermetallic compounds have recently been a subject of great interest. Rare-earth atoms possess a well-localized $4f$ magnetic moment. Although, in general, the overlap between the partly filled $4f$ shells of the neighboring atoms is small, thus resulting in a weak direct-exchange coupling, there exists a strong indirect-exchange mechanism provided by the conduction electrons. It is this indirect exchange which is responsible for the magnetic ordering of these materials. One interesting class of systems is that of the binary rare-earth cubic Laves phase compounds RX_2. The crystallographic and the magnetic properties of these compounds which allow for a large variation of magnetic or nonmagnetic X elements have been studied by various authors.[1-5] These investigations have been almost exclusively directed toward the determination of magnetization, magnetic-ordering temperature, and to the mechanisms of conduction-electron polarization. Little attention, however, has been paid to the critical behavior in these magnetic compounds. In this paper, we present a comprehensive experimental study of the temperature derivatives of the electrical resistivity $d\rho/dT$ over a wide range of temperatures around and in the close vicinity of the ordering temperature for six ferromagnetic rare-earth intermetallic Laves phase compounds, wherein the moment on the rare-earth sites is believed to be very well localized.

The magnetic ordering in these compounds may be viewed as determined by the relative strengths of the three principal types of couplings present: R-R, R-X, and X-X, where each coupling includes contributions from the conduction-electron polarization. For the compounds in which Fe, Co, or Ni are absent the s-f exchange provides the basic mechanism of the dominant magnetic interaction. On the other hand, when any of these three are present, the ordering of the magnetic sublattice (Fe, Co, or Ni) is important. In GdX_2 various values of the moments are observed at the X sites: about 1.7 μ_B on Fe atoms, about 1.1 μ_B on Co atoms, and with practically no moment on Ni sites. For $GdFe_2$ and $GdCo_2$, the high ordering temperatures ($\approx$ 800 and 400 °K, respectively) are strong evidence for the existence of other than direct-exchange interactions. Thus, these compounds offer an opportunity to investigate systems which from one point of view might appear to be similar to transition metals, where itinerant electrons play an important role, and from another viewpoint are similar to those with well-localized spins. Transport measurements near the magnetic-ordering transition in magnetic conductors are extremely sensitive to the details of the magnetic interactions.[6,7] Both in pure rare-earth metals[8-10] and binary intermetallic compounds,[11-13] the transport measurements display dramatic anomalies directly related to the onset of magnetic order.

While the temperature dependence of the electrical resistivity ρ for $T \leq T_c$ is determined by the long-range nature of the magnetic order, it is of interest to study the effect of short-range order on $\rho(T)$ for $T > T_c$. Since the total resistivity is generally a monotonic function of temperature, the determination of the temperature derivative of the resistivity $d\rho/dT$ enables us more carefully to study the effects of both long- and short-range order. Recently, there have been a number of experimental investigations of $d\rho/dT$ in both pure metals[14] and alloy systems,[15,16] and it is found that the critical behavior is, in general, complex, revealing different regions of temperature dependences in the vicinity of the ordering temperature, and the divergences in $d\rho/dT$ are usually much stronger than simple logarithmic. According to Fisher and Langer[17] and Suezaki and Mori,[18] the nature of the singularity of the magnetic part of $d\rho/dT$ near the ordering temperature can be associated directly with the critical behavior; therefore the study of $d\rho/dT$ is of more fundamental importance, not only in its own right but also in that it provides a reasonably good criterion for determining the ordering temperature.

Since no comprehensive theory of the critical behavior of transport properties in metallic systems exists, these systems provide us with an opportunity to investigate in a systematic and detailed manner those questions which may lead to a better description of the nature of the magnetic transition. In our previous work[19] on the resistivity of $GdNi_2$, we have shown that the behavior of $\rho(T)$ and $d\rho/dT$ may be understood in terms of the effect of the long-range nature of the critical fluctuations of the short-range spin-spin correlations in the framework of the molecular field theory. The applicability of this model to these compounds is an important consideration in pursuing this study in similar systems. In this light, we have investigated the critical behavior of other rare-earth compounds. In Sec. II, we give the experimental description of this study, and in Sec. III, we present the results of our measurements and attempt to provide a qualitative understanding.

II. EXPERIMENTAL DESCRIPTION

We have measured the temperature dependence of the electrical resistivity of six rare-earth intermetallic compounds: $GdAl_2$, $GdCo_2$, $GdNi_2$, $GdPt_2$, $GdRh_2$ and $DyAl_2$,[20] all exhibiting ferromagnetic ordering. The temperature range studied in these measurements was 4.2–300°K except for $GdCo_2$, in which case the range was extended to 500 °K. All samples were cut from large ingots into small bars of approximately 15 mm lengths and 1 mm × 1 mm in cross section. The sample shapes were optimized for our measurements, and current and voltage contacts were made with pure indium solder for all but $GdCo_2$, where a higher melting-temperature (≈ 300 °C) solder was employed. A standard four-point probe technique was used to determine the voltage across the sample while a known and well-regulated current (to 1 part in 10^5) was passed through the sample. The current magnitude was so adjusted (about 100 mA) as to provide the maximum number of significant figures measurable on a Honeywell Model 2779 microvolt potentiometer. At each temperature the current was reversed to avoid any extraneous emf's. The sample was in good thermal contact with a large Cu block into which were mounted Ge and Pt resistance thermometers, and a Cu-constantan thermocouple. In different temperature regions, various cryogenic fluids (liquid He, liquid N_2, and ice water) were used as baths which were weakly coupled to the Cu block. A manganin wire heater was imbedded in the block to vary its temperature and an automatic temperature controller regulated the temperature of sample and block to several millidegrees. The absolute accuracy of the temperature determination was about 0.1 °K, but the changes in temperature were always determined to better than 0.01 °K. The data, which were taken at 1 °K intervals in the temperature region about 50 °K on either side of the ordering temperature, were point-by-point computer fitted over a five-point span to a best-fit second-order polynomial, and then point-by-point differentiated to give $d\rho/dT$. To facilitate the critical analysis, $d^2\rho/dT^2$ was also obtained from the $d\rho/dT$ data in the same manner.

III. RESULTS AND DISCUSSION

In Fig. 1, we present data for both the electrical resistivity ρ and the computed temperature derivative $d\rho/dT$ for $GdNi_2$, $GdRh_2$, and $GdPt_2$. As is clear from these curves, the resistivity is essentially linear in T for the higher temperatures and shows a marked change in slope as the ordering temperature is approached. The details of these changes are more distinctly demonstrated in the $d\rho/dT$ curves which are superimposed over the $\rho(T)$ plots. All three $d\rho/dT$ curves show a broad maximum at high temperatures well above T_c followed by a minimum as T is decreased, which is further followed by a sharp rise toward a well-defined maximum. The temperature corresponding to the maximum for each system is associated with the ordering temperature. For temperatures below this maximum, there is, in general, a drop in $d\rho/dT$ for all samples. In $GdPt_2$, the maximum is rather sharp and roughly symmetric with a width of about 24 °K. In the other two samples the width is much larger though the maximum is well defined, and in addition there is a noticeable asymmetry. As is evident from these curves the depth of the minimum of $d\rho/dT$ for $GdPt_2$ and $GdRh_2$ is much smaller than that of $GdNi_2$. The temperature difference between maximum and minimum is larger in $GdPt_2$ and $GdRh_2$ (10 and 14 °K, respectively) as compared to only a few degrees in $GdNi_2$.

The nature of the transition in all three of these compounds is similar, and can be described in terms of the $\rho(T)$ behavior predicted by the molecular field theories of de Gennes and Friedel,[21] and Kim[22] for the critical region; and in our previous

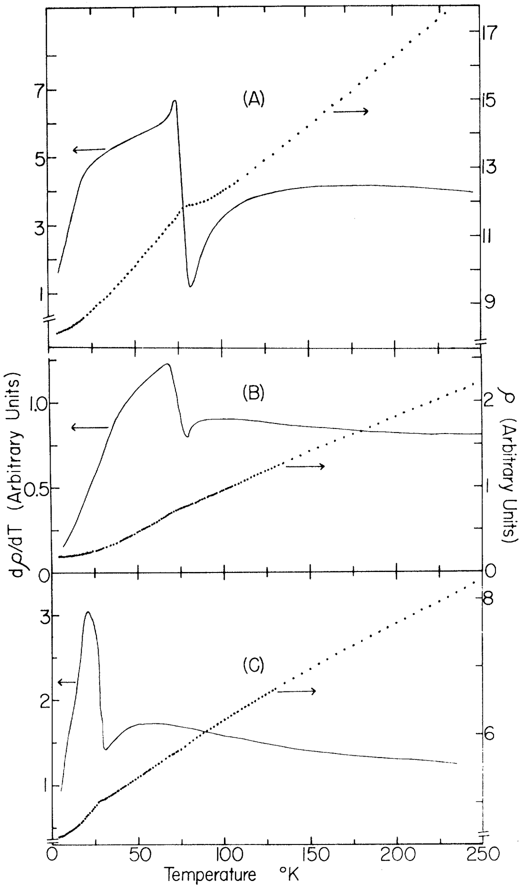

FIG. 1. The electrical resistivity $\rho(T)$ and its temperature coefficient $d\rho/dT$ of (A) $GdNi_2$, (B) $GdRh_2$, and (C) $GdPt_2$. The arrows indicate the respective scales. Note the breaks in the various ordinates.

publication[19] we have quantitatively analyzed this behavior for $GdNi_2$. In these systems we believe that the basic scattering mechanism of the magnetic contribution to the electrical resistivity is provided by the local s-f exchange interaction. Furthermore, close to T_c the long-range part of the critical fluctuations in the short-range order contributes significantly to the temperature dependence of the scattering. The resistivity[21,22] can be written in terms of a static spin-spin correlation function $\Gamma_{ij}(\vec{R})$ of two localized spin moments. $\vec{R}$ is the separation between two spins at ith and jth sites. In the molecular field approximation $\Gamma(R) \propto R^{-1}\exp(-\kappa R)$, where κ is the inverse correlation range. The use of this correlation function leads to a cusp in $\rho(T)$ and an infinite discontinuity in $d\rho/dT$ with positive value of $d\rho/dT$ for $T \leq T_c$ and negative ones for $T \geq T_c$. In Fig. 1, the calculated contribution, assuming this form for $\Gamma(R)$, is in good agreement with the experimental data for $GdNi_2$ as reported previously.[19] When other scattering mechanisms to the resistivity, in particular the electron-phonon contribution, are taken into account, the divergence in the magnetic part is smoothed out, thus giving the experimentally observed maximum followed by a minimum at higher temperatures. The exact behavior of the nonmagnetic parts of ρ is not known at low temperatures for these systems, and there is no unique method to separate out the electron-phonon scattering contributions from the spin-disorder resistivity. For systems with low ordering temperatures, this is especially a problem since the nonmagnetic part of ρ depends more strongly than linearly on temperature. This fact, coupled with crystal-field effects and the precise quality of the samples, is important in determining the relative differences of the maximum and minimum in the critical behavior of $d\rho/dT$. Therefore, we cannot carry out any detailed quantitative comparison between the theory of the magnetic contribution and the experimentally determined quantities for $GdPt_2$ and $GdRh_2$.

Figure 2 shows the $\rho(T)$ and the computed $d\rho/dT$ behavior for $GdCo_2$, $GdAl_2$, and $DyAl_2$. In the case of $DyAl_2$, at temperatures not far above the transition, there is a small slope in $d\rho/dT$, thereby possibly indicating a nonlinear dependence of the phonon contribution. As we approach the critical temperature from above, $d\rho/dT$ rises markedly, and then gently bends over to give a weak maximum at $T \approx 48\,^\circ\mathrm{K}$ (seen more clearly by looking at the second derivative of ρ). At the lowest temperature $d\rho/dT$ decreases, at first slowly, but then strongly. Immediately below T_c, if one subtracts out the extrapolated (from high temperatures) phonon contribution to $\rho(T)$, then one obtains a constant $d\rho/dT$ region extending to about 28 °K.

Fisher and Langer[17] have pointed out that the decrease in the magnetic part of the resistivity due to the incoherent part of the scattering is proportional to $M(T)^2/M_0^2$, where M is the spontaneous magnetization. This results in $d\rho/dT \propto (T-T_c)^{2\beta-1}$ (where β is the well-known magnetization critical exponent). When $d\rho/dT$ is constant, as we find for $DyAl_2$ between approximately 28 and 48 °K, $\beta = \frac{1}{2}$, which is just that predicted by the simple molecular field theory. In $DyAl_2$ the weakness of the maximum sets a limit on how well we can determine T_c from the resistivity data. Conversely, $GdAl_2$ shows a sharp and well-defined maximum in $d\rho/dT$ at about 157 °K which we associate with the Curie temperature. There is a relatively flat region in the behavior of $d\rho/dT$ just below and just above the ordering temperature not unlike that found in $DyAl_2$.

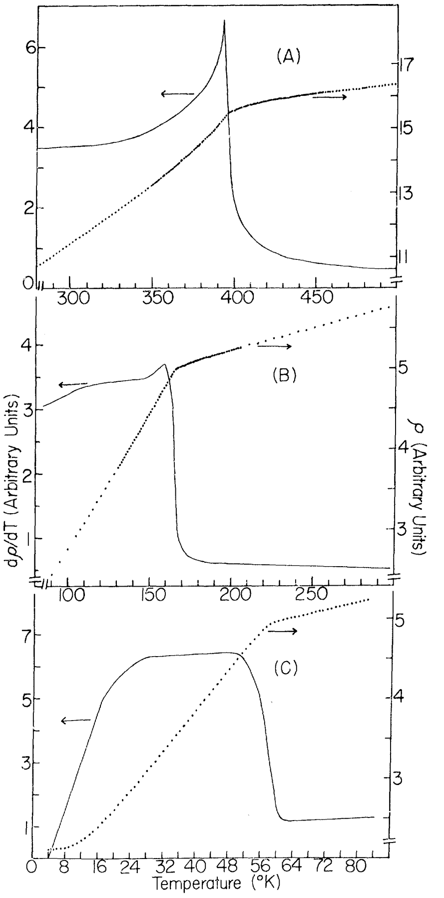

FIG. 2. The electrical resistivity $\rho(T)$ and its temperature coefficient $d\rho/dT$ of (A) $GdCo_2$, (B) $GdAl_2$, and (C) $DyAl_2$. Note that the temperature range for each sample is different; otherwise, as in Fig. 1.

The behavior for $GdCo_2$ is more distinctive than that of $DyAl_2$ or $GdAl_2$. The transition to the ordered state occurs sharply at 393°K with a clear peak. As we move from the $DyAl_2$ to the $GdAl_2$ on to the $GdCo_2$, we see a more pronounced peak in $d\rho/dT$. This behavior is traceable, in part, to the more linear temperature dependence of the phonon contribution, which we expect to have at higher ordering temperatures. The $d\rho/dT$ temperature dependence in $GdCo_2$ is much steeper for $T > T_c$ than for $T < T_c$ resulting in an asymmetric behavior about the ordering temperature. This ordering temperature, as has been previously noted,[1] is considerably above those found for the other compounds studied, but it is still lower than that of pure Co and higher than that of pure Gd.

Crangle and Ross[1] report in their magnetization study of many Laves phase compounds that the coupling mechanism for $GdCo_2$ is similar to that in $GdPt_2$, $GdRh_2$, and $GdNi_2$. They concluded this from the similar variations of T_c in moving across the rare-earth series, while holding the transition-metal ion constant. From our critical resistivity analysis we find wide ranges of behavior in $d\rho/dT$ for these compounds. This indicates that though the over-all interaction mechanism may be similar, the details of these mechanisms do show clear differences. The over-all behavior of $GdCo_2$ is ferrimagnetic (magnetization $\mu \approx 4.7\ \mu_B$ per formula unit) with antiferromagnetic coupling between the rare-earth sublattice ($\mu \approx 7\ \mu_B$) and the Co sublattice ($\mu \approx 1.1\ \mu_B$). Clearly in the case of $GdCo_2$, the Co-Co coupling plays a significant role. The moments on the Co atoms are not so well localized as they are on the Gd sites. This, in turn, causes relative differences in the spin disorder scattering thereby giving qualitative differences between the $d\rho/dT$ behavior of $GdCo_2$ and that of $GdNi_2$ and the others. According to Methfessel[23] and references therein, it is probable that in the ferromagnetic compound $GdCo_2$ the ordering at about 400°K occurs because of the 3-d spin alignment: the Gd moments ordering at a lower temperature.

In Table I, we give the ordering temperatures of all the compounds studied as determined from the $(d\rho/dT)_{\max}$ criterion; we compare these "Curie" temperatures with those obtained from the magnetization measurements of various other workers. Fundamental to a detailed comparison of these ordering temperatures are many questions about the characterization of the magnetic transitions in compounds and alloys in which the transition may be broadened because of stoichiometry or impurity factors. In all cases the T_c found from the $d\rho/dT$ maximum is consistently lower than those derived by extrapolating from high-field magnetization data. In the absence of a detailed quantitative theory of $d\rho/dT$ there may be some uncertainty in the precise value of T_c arrived at from our $d\rho/dT$ data. It should be added that the T_c determined from magnetization data depends strongly on the way the extrapolation procedure is carried out. No systematic critical analysis of the magnetization results has been done; therefore, we do not elaborate on the relatively small differences in T_c.

We have attempted to carry out a critical analysis for these samples by making plots on a log-log scale of $-d^2\rho/dT^2$ versus $\epsilon \equiv (T - T_c)/T_c$ for $T > T_c$, where T_c is determined from our $(d\rho/dT)_{\max}$ criterion. Here we may represent $d^2\rho/dT^2$ in the form[14]

$$d^2\rho/dT^2 = -(A/T_c)\,\epsilon^{-(\lambda+1)} ,$$

where A is a constant and λ is the critical expo-

TABLE I. Comparison of Curie temperatures as determined from $(d\rho/dT)_{\max}$ and magnetization measurements.

Compound	T_c from $(d\rho/dT)_{\max}$ (°K)	T_c (Ref. a) (°K)
$GdNi_2$	75	85, 77,[b] 90[c]
$GdPt_2$	22	36[b]
$GdRh_2$	66	77
$GdAl_2$	157[d]	171, 176, 182
$DyAl_2$	48	53, 62, 70
$GdCo_2$	393	404, 408

[a] Except for those temperatures marked with a superscript letter, see T. F. Connolly and Emily D. Copenhaver, *Bibliography of Magnetic Materials and Tabulation of Magnetic Transition Temperatures* (Clearinghouse for Federal Scientific and Technical Information, N. B. S. Springfield, Va., 1969).

[b] See Ref. 1.

[c] E. A. Skrabek and W. E. Wallace, J. Appl. Phys. 34, 1356 (1963).

[d] Note that this value of T_c is that obtained from the sample supplied by H. J. van Daal. The lower value of T_c previously reported by us (Ref. 12) is probably due to the presence of inhomogeneities in the original sample.

nent. In particular, $\lambda = 0$ corresponds to a logarithmic singularity. Because of the complex behavior just above T_c and the lack of fine-grain data in the steeply rising part of $d\rho/dT$ in $GdNi_2$, $GdPt_2$, and $GdRh_2$, this type of analysis is not directly possible. For the other samples ($GdAl_2$, $DyAl_2$, and $GdCo_2$), using the computed second derivatives of ρ, we find a complicated behavior with no simple divergence referred to the chosen T_c. At the large ϵ values λ is always greater than zero and changes sign as ϵ is reduced. There is no region where one may view the divergence in $d\rho/dT$ as logarithmic corresponding to $\lambda = 0$. Thus, we do not emphasize the application of this type of critical analysis to these data.[24]

At the present time no quantitative theoretical explanations exist concerning the varieties of behavior found for the resistivity and its derivatives at the critical region of a magnetic transition. However, the basic ground work leading to a unifying complete theory may be found in the approaches of Fisher and Langer,[17] Kim,[22] de Gennes and Friedel,[21] Mori,[25] and Suezaki and Mori.[18] It is hoped that the experimental results presented and discussed in this paper will stimulate further theoretical formulations for the behavior of transport properties at critical points in magnetic solids.

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PHYSICAL REVIEW B VOLUME 2, NUMBER 3 1 AUGUST 1970

Neutron and X-Ray Diffraction Study of the Low-Temperature Chemical and Magnetic Structure of Alpha-Manganese†*

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The low-temperature chemical and magnetic structures of antiferromagnetic α-manganese have been investigated. High-resolution neutron powder data indicate that the chemical and the magnetic unit cell are identical in size. An x-ray powder study of 60 reflections at 77 and 4 °K indicates no significant changes at low temperature in the atomic coordinates. Fourteen independent magnetic reflections measured at 77, 50, and 4 °K were used to determine the signs and magnitudes of the four magnetic moments for a collinear spin model. At 4 °K, they are 1.72 ± 0.19, 1.46 ± 0.08, 1.11 ± 0.04, and 0.02 ± 0.05 in units of μ_B for atoms I, II, III, and IV, respectively. The fit of the observed and calculated magnetic intensities indicates the need for a less restricted model for the magnetic structure. These results are compared with those of other experiments.

I. INTRODUCTION

The low-temperature allotrope of transition-metal manganese is α-manganese. Its chemical structure is bcc with 58 atoms per unit cell in the space group $T_d^3 - I\bar{4}3m$.[1] These atoms are distributed on four different sites. I: two atoms at 000 + bcc with point symmetry $\bar{4}3m$; II: eight atoms at x, x, x + bcc with point symmetry $3m$, III: 24 atoms at x, x, z + bcc with point symmetry m; IV: 24 atoms at x, x, z + bcc with point symmetry m, referred to subsequently as sites I, II, III, and IV, respectively.

In 1953, Shull and Wilkinson[2] established the existence of an ordered antiferromagnetic state in α-manganese below 100 °K from a neutron diffraction study of the powder. The complexity of the chemical structure and the paucity of measurable magnetic reflections made the determination of a magnetic structure impossible in that investigation. Magnetic diffuse scattering was observed above the Néel transition, providing evidence of residual short-range order between localized moments.

Kasper and Roberts,[3] in 1956, reported the results of another neutron diffraction study of α-manganese powder. They observed that new coherent diffraction peaks appeared below the transition temperature for odd values of $h+k+l$, while the nuclear diffraction peaks remained unchanged. They deduced that the antiferromagnetic ordering was associated with spin reversal under a body-centering translation. The intensities of seven magnetic reflections were then analyzed in terms of a restricted collinear spin model and found to be consistent with either of the two sets of magnetic moments for the four atom types listed in Table I. In their analysis, Kasper and Roberts