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Magnetic ordering and exchange interactions in the rare-earth gallium compounds $R\text{Ga}_2$

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Magnetic ordering and indirect exchange interactions were studied in the series of hexagonal rare-earth intermetallic compounds $R\text{Ga}_2$, where R denotes Ce, Pr, Nd, Gd, Tb, Dy, Ho, and Er. The magnetic susceptibility (χ) of polycrystalline samples was measured at low fields, from 1.5 K to about 300 K; the magnetization (M) versus applied magnetic field (H) of these polycrystalline samples and a HoGa_2 single crystal was measured up to 80 kOe at low temperatures; the resistivities (ρ) of CeGa_2, GdGa_2, DyGa_2, and HoGa_2 were measured from about 1.5 K to about 300 K, at zero field. The $\chi(T)$ and $M(H)$ results indicated that the samples order antiferromagnetically, with ordering temperatures T_N ranging from 6.4 K for DyGa_2 to 14.8 K for TbGa_2. $M(H)$ results for single-crystal HoGa_2 show that the [100] direction is the easy direction and the [001] direction is the hard direction. High-temperature $\chi(T)$ results were fitted to the Curie-Weiss formula and the resulting effective moments are in good agreement with those expected for trivalent rare-earth ions. The paramagnetic Weiss temperatures (Θ) are positive for all the samples measured except GdGa_2. The effects of magnetic ordering were observed below T_N in the resistivity results. Letting the spin-disorder part of the resistivity ρ_s be proportional to T^n, n was determined to be about 2.0, 4.3, and 3.3 for HoGa_2, GdGa_2, and DyGa_2, respectively. In view of the theory of Mannari for ρ_s, these differences may be connected with differences in the spin structures of these compounds. Ruderman-Kittel-Kasuya-Yosida (RKKY) sums have been calculated for these compounds. The negative sign of the sum is in agreement with the anitiferromagnetic properties observed in various measurements. For HoGa_2, the dressed moment, due to polarization of conduction electrons, has been estimated. The theory of Nagamiya was applied to three of the compounds which can be considered as a layer compound with long-range ferromagnetic intralayer coupling and antiferromagnetic coupling between a layer and its nearest-neighbor layers. With one possible exception, the data are consistent with the theory which contains three exchange coefficients. Crystal-field-induced anisotropy appears to be important in determining the spin structures of these compounds.

I. INTRODUCTION

In the last decade or two there has been a great deal of interest in the magnetic properties of rare-earth intermetallic compounds. In terms of gaining a fundamental understanding of magnetism in metallic conductors, rare-earth intermetallics are simpler to understand than $3d$ elemental metals and $3d$ metallic compounds because there is generally a clear distinction between well-defined $4f$ localized moments and conduction electrons in the former case, whereas in the latter the localized versus itinerant character of the $3d$ moments is difficult to understand. Although there have been quite a number of studies on the $R\text{Al}_2$ compounds, very little has been reported on the $R\text{Ga}_2$ series.[1] In the $R\text{Ga}_2$ compounds, since the Ga ion is nonmagnetic and contributes three conduction electrons, and since dipole-dipole interactions and direct exchange between $4f$ electrons of the rare earths is negligible,[1] one might expect the de Gennes–Ruderman-Kittel-Kasuya-Yosida (RKKY) theory[2,3] to be a reasonably good approximation for the series.

We have reported previously some $\chi(T)$ and $M(H)$ results; this earlier paper[4] will be referred to in this work as Paper I. Barbara *et al.*[5] and Asmat and Gignoux[6] performed neutron-diffraction measurements on several compounds in the series and determined a variety of stable antiferromagnetic or cycloidal spin structures. From the $\chi(T)$ and $M(H)$ results, we have determined Néel temperatures, parameters such as effective moments and Weiss temperatures in most of the $R\text{Ga}_2$ compounds. High-field magnetization and resistivity at low temperatures also have been studied.

II. EXPERIMENTAL RESULTS

$R\text{Ga}_2$ intermetallic compounds have the aluminum-boride (AlB_2) structure, which is hexagonal and belongs to the space group $P6/mmm\text{-}D_{6h}^1$ with R at (0,0,0) and Ga at $\pm(\frac{1}{3}a,\frac{2}{3}b,\frac{1}{2}c)$. The structure, shown in Fig. 1, has one formula unit per primitive cell. The c/a ratio ranges from 1.00 to 0.96 and

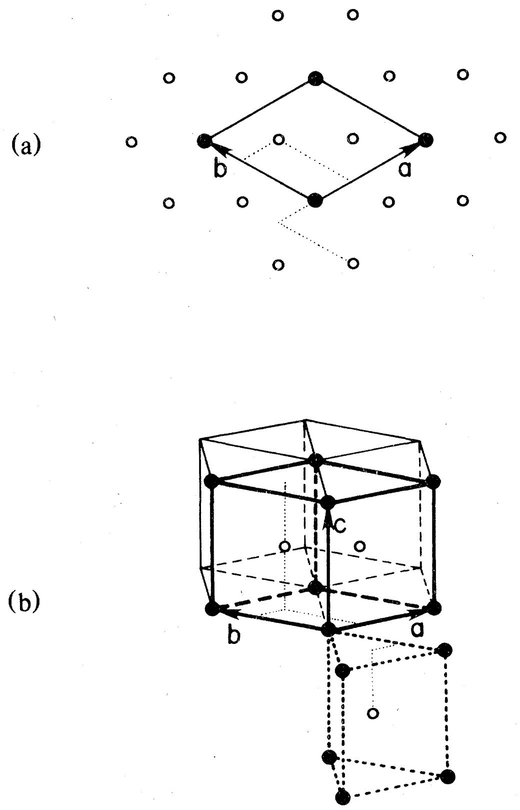

FIG. 1. (a) View of the basal plane from the c axis. (b) The side view of the hexagonal structure; for the RGa$_2$ compounds. The solid circles are rare-earth ions, the open circles are the gallium ions.

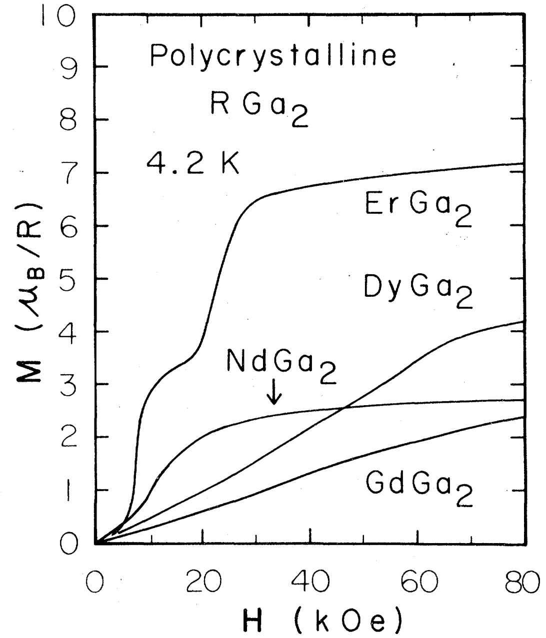

FIG. 2. Magnetization vs applied magnetic field at 4.2 K for the polycrystalline RGa$_2$ samples indicated.

the length of the lattice parameter a from 4.32 to 4.186 Å, in the sequence from CeGa$_2$ to ErGa$_2$.[7]

Sample preparation and some of the susceptibility and magnetization results were reported in Paper I. The $M(H)$ results at 4.2 K for NdGa$_2$, GdGa$_2$, DyGa$_2$, and ErGa$_2$ are shown in Fig. 2. It should be noted that fields even of 80 kOe do not saturate the magnetization to values close to the gJ values for the R^{3+} ions. Specifically the moment values per R at 80 kOe, normalized to the gJ values, are about 0.8 for Nd, Ho, and Er, 0.7 for Pr, 0.5 for Ce, 0.4 for Tb and Dy, and 0.3 for Gd. It is thus apparent that significant anisotropy exists in certain of these compounds, since the magnetization is far from saturated at fields much larger than the spin-flop fields.

The susceptibility and magnetization data for ErGa$_2$ exhibit some unusual features. For example $\chi(T)$ (Fig. 1 in Paper I) shows a second kink at 4 K which is below T_N (7.5 K). Presumably this is due to a change in the nature of the spin structure at this lower temperature. Figure 2 clearly shows the presence of two spin-flop fields of about 7.5 and 23 kOe. Figure 3 shows that these two spin flops exist at both 4.2 and 6 K, but disappear above T_N; it also shows that there was a strange hysteresis in the magnetization at 30 and 43 K, at fields of about 35 and 50 kOe, respectively. Except for the possibility of a crystallographic distortion at these temperatures, perhaps affected by magnetoelastic forces, we have no suggestion as to the origin of this anomaly.

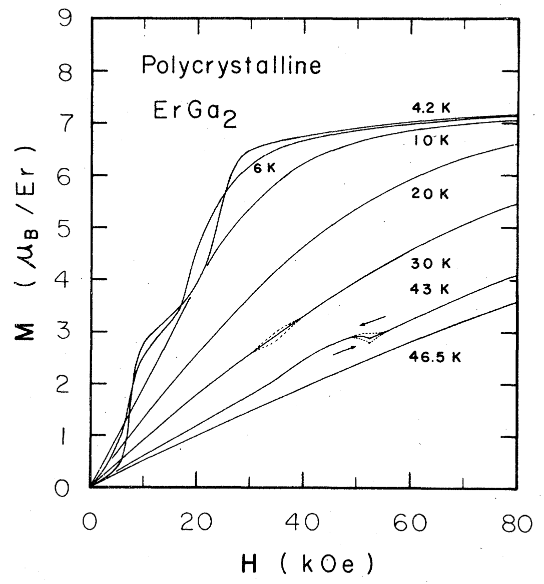

FIG. 3. M vs H at various temperatures for polycrystalline ErGa$_2$.

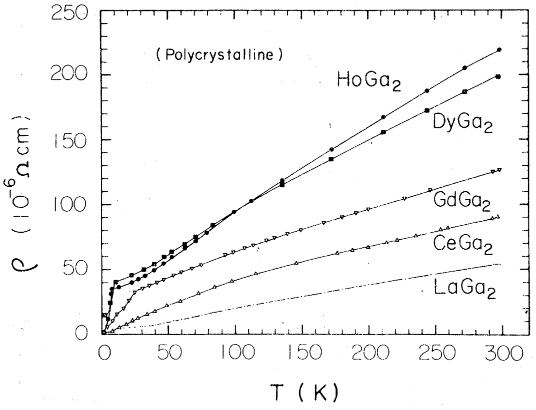

FIG. 4. Resistivity ρ vs temperature for selected polycrystalline $R\mathrm{Ga}_2$ samples.

The electrical resistivity for the polycrystalline samples was measured from about 1.5 to about 300 K at zero field in a cryogenic system. The data, recorded automatically by a digital-data-acquistion system, were taken at each temperature by running a constant current through the sample in both directions, and averaging the results to cancel thermal emf's. At various temperatures, resistance of the sample versus current was checked. The constant current was chosen in the region where the resistance was independent of current, and so that it did not heat the sample. The constant current used for all the samples measured was about 0.1 A. Typically, the current densitites were about 1.4–2.0 A/cm^2.

The electrical resistivity results for selected samples between 1.5 and 300 K are shown in Fig. 4. An expanded version showing details at low temperatures is shown in Fig. 5. Spin-disorder scattering below T_N is especially apparent in the results for HoGa_2 and DyGa_2. The "break" at 7.6 K for HoGa_2 is fairly close to the estimate of $T_N = 8.0$ K obtained from the $\chi(T)$ results. However, for DyGa_2 it is the kink at about 6.3 K rather than the "break" at about 10 K that signifies the Néel temperature. This structure at 10 K most likely has the same origin as the $\chi(T)$ bump centered on 11 K that is seen in Fig. 1 of Paper I, viz., a crystal-field effect. In order to understand these $\chi(T)$ and $\rho(T)$ results in detail, it will be necessary to have further information on specifics of the crystal-field splittings for Dy in this compound.

Although much less distinct, the $\rho(T)$ results for GdGa_2 and CeGa_2 also show the onset spin-disorder scattering below T_N. $\chi(T)$ for nonmagnetic LaGa_2 is relatively flat below 20 K except for an anomalous kink at 7.5 K. Probably this kink is due to a phase transition (superconductive or magnetic) in a small portion of second phase in the sample. The La metal used for making LaGa_2 contained the impurities: 0.02-at.% Ce, 0.02-at.% Pr, 0.003-at.% Nd, and 0.005-at.% Fe. In any case, the results for LaGa_2 clearly show, in the region below about 15 K, that the phonon scattering is negligible in comparison with the spin-disorder scattering in the magnetic compounds.

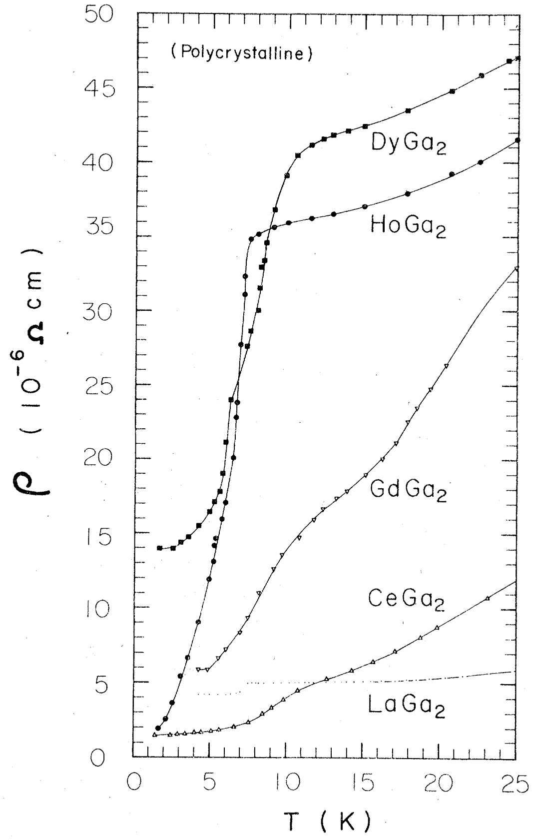

FIG. 5. Resistivity ρ vs temperatures for selected polycrystalline $R\mathrm{Ga}_2$ samples.

In order to determine the background susceptibility in a nonmagnetic $R\mathrm{Ga}_2$ compound, the susceptibility of polycrystalline LaGa_2 was measured between 4.2 and 300 K. The results could be well fitted in terms of a temperature-dependent term χ_0 due to Pauli paramagnetism (plus any diamagnetic core contribution), and a low-temperature Curie-like upturn due to the impurities mentioned above. The value of χ_0 obtained was 0.36×10^{-6} emu/g or 90.5×10^{-6} emu/mole. The local moment term due to the impurities was consistent with the impurity levels mentioned above.

III. DISCUSSION

The experimental effective moment p of the R ions, the paramagnetic Weiss temperature Θ, the estimated Néel temperature T_N, and the theoretical effective moment $g_J[J(J+1)]^{1/2}$ for R^{3+} ions for all the samples are listed in Table I. For most of the compounds the p values are in good agreement,

TABLE I. Experimental Néel temperature T_N, Weiss temperature Θ, experimental effective moment p, and theoretical effective moment $g[J(J+1)]^{1/2}$ for $R\text{Ga}_2$ compounds.

R	T_N(K)[a]	Θ(K)	p	$g[J(J+1)]^{1/2}$
Ce	9.5	12.5 ±2.0	2.51 ±0.20	2.54
Pr	7.3	10.6 ±0.1	3.50 ±0.02	3.58
Nd	9.2	12.4 ±0.1	3.63 ±0.02	3.62
Gd	12.1	−17.4 ±0.2	7.92 ±0.05	7.94
Tb	14.8	25.6 ±0.5	9.29 ±0.09	9.72
Dy	6.4	1.8 ±0.1	10.72 ±0.30	10.65
Ho	8.0	13.1 ±0.1	10.10 ±0.04	10.61
Er	7.5	1.7 ±0.2	9.41 ±0.55	9.58

[a] The uncertainties in T_N are: ±1 K for CeGa_2, ±0.5 K for TbGa_2 and GdGa_2, and ±0.1 K for the rest of the compounds.

TABLE II. The RKKY sum, $\sum_{i\neq 0} F(2k_F R_{i0})$ and $\sum_{i\neq 0} F(2k_F R_{i0}) e^{-R_{i0}/l}$ with $l = 10$ Å, for the $R\text{Ga}_2$ compounds. Up to nine nearest neighbors were summed over.

Compound	$\sum_{i\neq 0} F(2k_F R_{i0})$ (10^{-2} Å^{-3})	$\sum_{i\neq 0} F(2k_F R_{i0}) e^{-R_{i0}/10}$ (10^{-2} Å^{-3})
CeGa_2	−2.267	−1.277
PrGa_2	−2.341	−1.327
NdGa_2	−2.355	−1.333
GaGa_2	−2.445	−1.399
TbGa_2	−2.450	−1.412
DyGa_2	−2.467	−1.424
HoGa_2	−2.468	−1.432
ErGa_2	−2.477	−1.439

within experimental error, with those expected for R^{3+} ions. However, for Pr, Tb, and Ho the p values are clearly smaller than the theoretical ones. It is interesting to note that in other Ho intermetallics such as HoZn_2,[8] and HoAg,[9] p is also less than the expected theoretical value for Ho^{3+} ions. The reason for this is presumably conduction-electron spin polarization through the sf interaction[10]

$$H_{sf} = -\Gamma(q)\vec{S}\cdot\vec{s}\ , \tag{1}$$

where $\vec{s}$ is the conduction-electron spin operator, and $\Gamma(q)$ is the sf exchange parameter. The total moment m is given by Stewart as[11]

$$m = m^i[1 + 2\Gamma(0)\rho(g_J - 1)/g_J]\ , \tag{2}$$

where $m^i = g_J J \mu_B$ is the ionic moment, the second term is due to the conduction-electron polarization, ρ is the conduction-electron density of states per atom for one-spin direction, and the equation is valid when all the ionic moments are parallel. In terms of simple ideas we may write

$$m = m^i + m^c = g_J J \mu_B + 2S^c \mu_B\ , \tag{3}$$

where m^c and S^c are the effective conduction-electron moment and spin due to polarization by the Ho moments. If J' is defined to be the effective angular momentum quantum number of the Ho moment plus polarization cloud, then

$$p \equiv g_J[J'(J'+1)]^{1/2}\ . \tag{4}$$

In the easy ($\langle 100 \rangle$) direction (Fig. 4 in Paper I), $m \simeq 9.0$ and from $p = 10.1$, $J' = 7.6$. Assuming $J' \simeq J + S^c$ we have $S^c \simeq -0.4$, which is approximately in agreement with the value of S^c obtained from Eq. (3), viz., −0.5. If the diminished "saturation" moment and effective moments are indeed due to such conduction-electron polarization, then Eqs. (1) and (2) suggest that the sf interaction is antiferromagnetic in character, i.e., the exchange integral Γ is negative.[1] Microscopic measurements on this question would be of interest.

Table II gives the results of the RKKY sum [Eq. (4) in Paper I] for the $R\text{Ga}_2$ compounds; the sums are all negative with values (excluding the exponential factor which accounts for the electron mean free path l) ranging from -2.267×10^{-2} Å^{-3} for CeGa_2 to -2.477×10^{-2} Å^{-3} for ErGa_2. Antiferromagnetism is indicated by the negative sign of the RKKY sums, in agreement with the antiferromagnetic properties observed in the $\chi(T)$ and $M(H)$ results. By neutron diffraction, Barbara *et al.*,[5] and Asmat and Gignoux[6] have determined the spin structures for HoGa_2, DyGa_2, PrGa_2, and NdGa_2 at 4.2 K. The structures are all antiferromagnetic, with crystal-field-induced anisotropy playing an important role in determining the direction of spins.

Figures 6(a) and 6(b) show the magnetic unit cell for HoGa_2 and DyGa_2 in their ordered states. Both compounds can be regarded as layer compounds with ferromagnetic coupling in the ac planes. The spins are in the layers in the case of HoGa_2, and perpendicular to the layers for DyGa_2. The magnetic lattice for ErGa_2 is the same as for the Dy and Ho compounds; the only difference is that for the former compound the moments are aligned parallel with the c axis. In a qualitative sense, the structures make it clear how these compounds have positive Θ values because of the predominant ferromagnetic intralayer coupling, but overall antiferromagnetism because of the antiferromagnetic interlayer coupling.

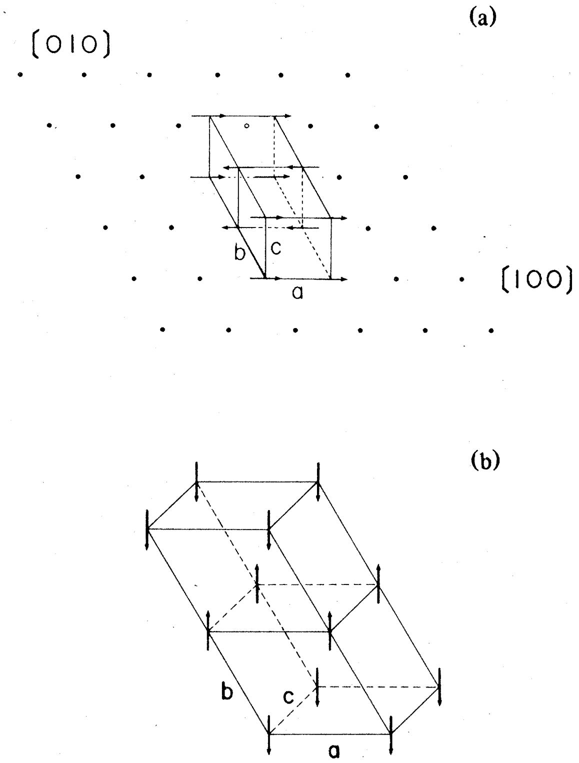

FIG. 6. (a) Antiferromagnetic structure of $HoGa_2$ as determined by neutron diffraction at 4.2 K. The dots represent the Ho ions in the basal plane. Only the spins of the magnetic unit cell are shown. (b) Antiferromagnetic spin arrangement for $DyGa_2$. The moments are on the *a-b* plane and perpendicular to the *a* axis.

Since the spin structures of the above three compounds can be considered as layered structures, and since the wave vector Q of the structures is perpendicular to the layers, the total exchange integral can be written as follows, according to the theory of Nagamiya[12]:

$$J(Q) = \sum_{\nu=-\infty}^{\infty} J_\nu \exp(-i\nu b'Q) \tag{5}$$

or

$$J(Q) \simeq J_0 + 2J_1\cos(b'Q) + 2J_2\cos(2b'Q) \ , \tag{6}$$

where $b' = (\frac{3}{2})^{1/2}b$ (b is the lattice constant), J_0 is the sum of exchange integrals coupling one spin to all the others in one layer, and J_1 and J_2 are similar sums coupling a spin in the initial layer with spins in the nearest and next-nearest layers, respectively. Since the compounds are antiferromagnetic, $Q = \pi/b'$ and $J_1 < 0$. It is possible to calculate the three exchange parameters J_0, J_1, and J_3 with the following results of the theory:

$$\begin{aligned} T_N &= (g_J-1)^2[J(J+1)/3k_B] \times 2J(Q) \ , \\ \Theta &= (g_J-1)^2[J(J+1)/3k_B] \times 2J(0) \ , \\ J_1/4J_2 &= -\cos(b'Q) = 1 \ . \end{aligned} \tag{7}$$

The experimental results for $DyGa_2$ are: $T_N = 6.4$ K, $\Theta = 1.8$ K with $J = 7.5$, $g_J = \frac{4}{3}$; for $HoGa_2$: $T_N = 8.0$ K, $\Theta = 13.1$ K with $J = 8$, $g_J = \frac{5}{4}$; and for $ErGa_2$: $T_N = 7.5$ K, $\Theta = 1.7$ K with $J = 7.5$, $g_J = \frac{6}{5}$. These results along with the theory lead to the following exchange parameters: for $DyGa_2$: $J_0 = 0.99$ K, $J_1 = -0.24$ K, and $J_2 = -0.06$ K; for $ErGa_2$: $J_0 = 3.13$ K, $J_1 = -0.85$ K, and $J_2 = -0.21$ K. Thus, although these results indicate that the initial layer is coupled antiferromagnetically to the *next*-nearest layer, the antiferromagnetic coupling between the intervening nearest layer and the next-nearest layer is sufficiently strong to overcome the fact that J_2 is negative.

On the other hand, in attempting to apply the Nagamiya theory to $HoGa_2$ a difficulty is encountered. This is that a straighforward application of Eqs. (7) to the experimental parameters leads to exchange coefficients J_0, J_1, and J_2, all of which are *positive*. This results because $\Theta > T_N$ and J_1 is given by

$$J_1 = \tfrac{1}{4}[J(0) - J(Q)] \ .$$

Clearly a positive J_1 is not consistent with the antiferromagnetic structure of Fig. 6(a). It is puzzling why the theory gives a consistent interpretation of the results for $DyGa_2$ and $ErGa_2$ but not for $HoGa_2$. The only conceivable explanation that occurs to us is that there is a systematic error in the determination of Θ for $HoGa_2$. If, for example, the $HoGa_2$ sample measured was in fact predominantly a single crystal rather than being truly polycrystalline, it might be the case that the Θ value measured could be shifted by crystal-field effects, as discussed by Wang.[13] Wang shows also that crystal-field effects will not shift the paramagnetic Weiss temperatures for a powdered (or polycrystalline) sample. As discussed in Paper I, the high-field magnetization for nominally polycrystalline $HoGa_2$ did exhibit behavior which is consistent with a preferred orientation of crystallites, perhaps as a result of directional solidification following arc melting.

Figure 7 shows a log-log plot of the spin-disorder part of the electrical resistivity below T_N for $HoGa_2$, $GdGa_2$, and $DyGa_2$. The results indicate that the resistivities are approximately proportional to $T^{2.03}$, $T^{4.25}$, and $T^{3.30}$ for the three compounds, respectively. Mannari[14] has shown, for a localized-moment ferromagnet for which the magnon energy ($\hbar\omega$) is quadratic in wave vector (q), that $\rho_s \propto T^2$; for an antiferromagnet for which $\hbar\omega \propto q$, $\rho_s \propto T^4$. These temperature dependences of spin-disorder resistivity need to

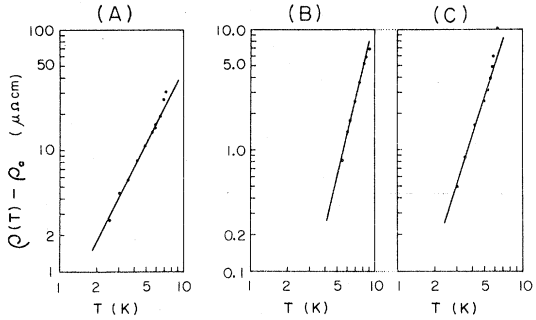

FIG. 7. Log-log plot of $\rho(T)-\rho_0$ against T. The slope of the straight line n is the power of temperature dependence T^n. (a) $HoGa_2$, $\rho_0 = 1.0$ $\mu\Omega$ cm, $n = 2.03$. (b) $GdGa_2$, $\rho_0 = 5.8$ $\mu\Omega$ cm, $n = 4.25$. (c) $DyGa_2$, $\rho_0 = 13.9$ $\mu\Omega$ cm, $n = 3.30$.

be modified by a factor $\exp(-\Delta/kT)$ if there is an energy gap Δ in the spin-wave spectrum due to anisotropy.[15] This experimental factor normally contributes fractional values to the power of the temperature dependence. It seems possible, but by no means certain, that the approximately T^2 result for $HoGa_2$ is related to the fact that, along the direction of the ordered spins, the spins of $HoGa_2$ are lined up ferromagnetically.

The spin-disorder resistivity result for $GdGa_2$ is close to T^4 which the theory would suggest is due to antiferromagnetic scattering. It should be noted that $GdGa_2$ has a negative Θ (-17.4 K) which is typical for an antiferromagnetic material. Furthermore, the ground state of Gd^{3+} is an S state ($^8S_{7/2}$), so one expects minimum anisotropy induced by crystal-field effects. Thus $GdGa_2$ is the crystal in the RGa$_2$ series which is most likely to follow the simple de Gennes–RKKY theory. Unfortunately, the magnetic structure of this compound is not known.

The temperature dependence of the susceptibility and resistivity in the neighborhood of T_N make it clear that a knowledge of the crystal-field levels will be required in order to understand the detailed behavior of $\chi(T)$ and $\rho(T)$. It is hoped that the present results will stimulate neutron or infrared studies aimed at determining these levels.

IV. SUMMARY AND CONCLUSIONS

The major results of this study can be summarized as follows. (i) All of the RGa$_2$ compounds exhibit predominant antiferromagnetic features, with ordering temperatures ranging from 6.4 to 14.8 K. Of the compounds studied, most of the effective paramagnetic moments are close to those expected for R^{+3} ions. Despite the general antiferromagnetic order, the paramagnetic Weiss temperatures were all positive with the exception of $GdGa_2$. This indicates the importance of close-neighbor ferromagnetic interactions. (ii) All of the compounds are metamagnetic in the sense that fields ranging from several hundred oersteds to about 40 kOe cause spin flops to ferromagnetic or canted spin states. In particular $ErGa_2$ shows two spin-flop fields, about 7.5 and 23 kOe at 4.2 K. Results on a single crystal for $HoGa_2$ show that the [100] direction is the easy direction and the [001] direction is the hard axis of magnetization. This is consistent with the spin structure determined with neutron diffraction by Barbara *et al.*, who showed that crystal-field interactions tend to align the spins perpendicular to the hexagonal axis. (iii) A detailed comparison was made between the experimental results and the de Gennes theory based on free-electon, RKKY coupling of the moments. The comparison shows that the sign of the paramagnetic Weiss temperature is incorrectly predicted (with the single exception of $GdGa_2$), and neither Θ nor $\rho_s^{\max}$ is proportional to the de Gennes factor. Presumably this results from the lack of free-electron behavior in the band structure, and/or the neglect of anisotropy in the theory. (iv) Attempts were made to apply the theory of Nagamiya to three of the compounds which can be considered as magnetic layer compounds. With one possible exception the signs and magnitudes of the three exchange constants determined were consistent with the data. (v) The temperature dependence of the spin-disorder scattering below T_N was studied for three of the compounds. Taking $\rho_s \propto T^n$, n ranged from about 2 to about 4. The differences seen in the three compounds presumably exist because of differences in short-range spin correlations and also because of differing effects of anisotropy on the spin-wave spectra.

In conclusion, this study points towards the need for further experimental measurements in these compounds to obtain information on crystal-field splittings. Since the structure of the RGa$_2$ compounds is simple, with only three atoms per primitive cell, this series would appear to be a fertile area for band-structure calculations as well as studies of crystal-field and exchange interactions of rare-earth ions in metallic, hexagonal environments.

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