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MAGNETIC PROPERTIES OF THE RARE-EARTH INTERMETALLICS RGa_2*

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ABSTRACT

The magnetic susceptibility (χ) of polycrystalline samples of RGa_2, R = Ce, Pr, Nd, Gd, Tb, Dy, Ho, and Er, has been measured at low field, from 1.5 K to 300 K. The magnetization of these samples also has been measured up to 80 kOe at low temperatures. Antiferromagnetic behavior was observed for all the samples with Neel Temperatures (T_N) ranging from about 4.1 K for $CeGa_2$ to about 14.8 K for $TbGa_2$. Curie-Weiss fits to the high-temperature χ(T) data led to effective moments in good agreement with those expected for R^{3+} ions. The paramagnetic Weiss temperatures cannot be reconciled with the de Gennes theory based on free electron coupling of the R spins via the RKKY interaction. Electrical resistivity of selected polycrystalline samples has been measured, and the effects of spin-disorder scattering observed below T_N. $CeGa_2$ shows no evidence of Kondo behavior. Magnetization measurements for the polycrystalline samples show metamagnetic phase transitions when the antiferromagnetic R-R interactions are overcome. Measurements on a $HoGa_2$ single crystal show that the [100] direction is the easy direction.

I. INTRODUCTION

Rare earth elements have electron configuration: [Xe] $4f^n5d^m6s^2$, where n, m, are integers, different for different elements. Since the 4f shell is imbedded deeply within 5s and 5p closed shells [1], it provides well-defined localized moments. These localized moments interact with each other via the conduction electrons and magnetic order is produced in many rare-earth metals and metallic compounds. The Ruderman-Kittel (RKKY) interaction based on free electrons suffices in some cases to understand the magnetic order but in other cases fails. The purpose of this work was to investigate whether plane-wave electron state plus RKKY theory would be adequate for the RGa_2 series of compounds. In addition we were interested in the question of whether $CeGa_2$ would exhibit "Kondo lattice" behavior as in compounds such as $CeAl_2$, $CeAl_3$ [2].

II. EXPERIMENTS AND RESULTS

The polycrystalline RGa_2 samples were prepared in an arc furnace. The purity of gallium is 99.9999% and of the rare earths is 99.9% or better. The crystal structure of the samples is hexagonal, AlB_2 type, with space group P6/mmm-D^1_{6h} [3]. The samples have been checked by x-ray diffraction to verify the structure and to ensure that no second phase was present.

Using the polycrystalline sample prepared as mentioned above, a $HoGa_2$ single crystal has been grown in a tungsten crucible by the Bridgman method. After being oriented by the Laue back reflection method, the crystal was cut into a cylinder by a spark cutter, with the c axis aligned along the cylindrical axis.

The magnetic susceptibility (χ) of polycrystalline RGa_2, R = Ce, Pr, Nd, Ga, Tb, Dy, Ho, and Er has been measured at low fields from 1.5 K to about 300 K, in a Faraday system. The χ(T) results from 1.5 K to 32 K are shown in Fig. 1.

High temperature χ(T) data were fit to the Curie-Weiss formula:

$$\chi = \chi_0 + \frac{C}{T-\theta} \quad . \qquad (1)$$

The fit for $HoGa_2$ is shown in Fig. 2. The effective moment of the R^{3+} ions p, the paramagnetic Weiss temperature θ, the estimated antiferromagnetic ordering temperature T_N, the theoretically calculated effective moment $g[J(J+1)]^{1/2}$, and the de Gennes factor $(g-1)^2J(J+1)$ for all the samples are listed in Table I.

Magnetization (M) versus applied magnetic field (H) at low temperatures has been measured up to 80 kOe by a vibrating sample magnetometer, with a superconducting coil providing the field. The results for $HoGa_2$, $TbGa_2$, $PrGa_2$, and $CeGa_2$ polycrystalline samples

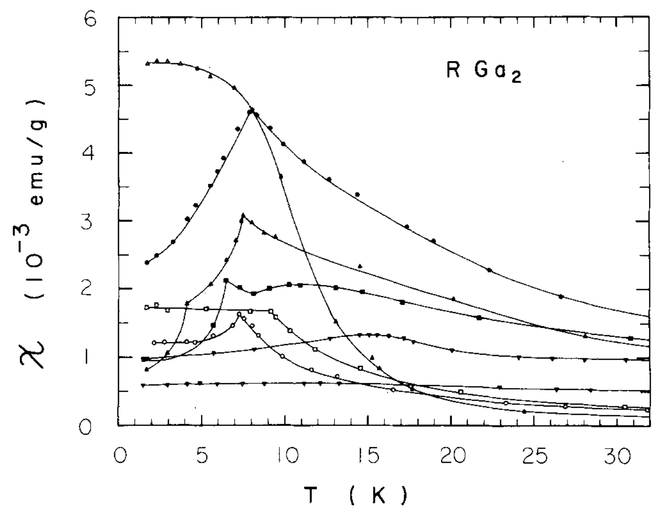

Fig. 1. χ(T) for Polycrystalline RGa_2. R: ▲ - Ce, ○ - Pr, ▽ - Gd, ▼ - Tb (H = 1.58 kOe); □ - Nd, ■ - Dy, ● - Ho, ▲ - Er, (H = 4.81 kOe).

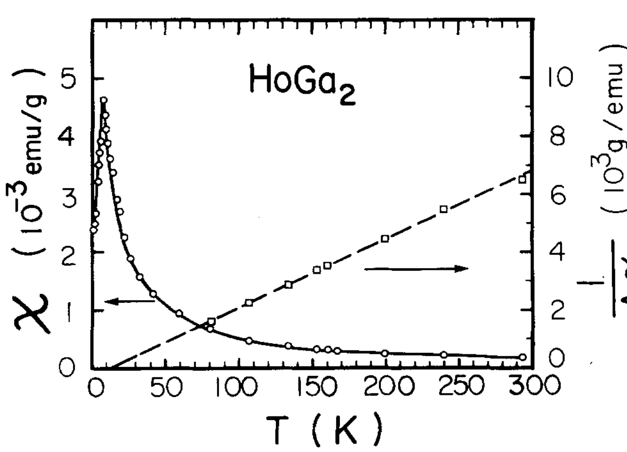

Fig. 2. Curie-Weiss fit for polycrystalline $HoGa_2$. $\Delta\chi(T) = \chi(T) - \chi_0$. Curie constant $C = 4.19 \times 10^{-2}$ emu-K/g, p = 10.10, θ = 13.1 K.

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at 4.2 K are shown in Fig. 3. The results for a $HoGa_2$ single crystal at 4.2 K measured along [100], [210], and [001] directions are shown in Fig. 4.

TABLE I

Experimental Neel temperature T_N, Curie-Weiss temperature θ, effective moment p; theoretical de Gennes factor $(g-1)^2 J(J+1)$, and effective moment $g[J(J+1)]^{1/2}$ for RGa_2 compounds.

R	T_N(K)	θ(K)	$(g-1)^2J(J+1)$	p	$g[J(J+1)]^{1/2}$
Ce	4.1	12.5	0.18	2.51	2.54
Pr	7.3	10.6	0.80	3.50	3.58
Nd	9.2	12.4	1.84	3.63	3.62
Gd	12.1	-17.4	15.75	7.92	7.94
Tb	14.8	25.6	10.5	9.29	9.72
Dy	6.4	1.8	7.08	10.72	10.65
Ho	8.0	13.1	4.50	10.10	10.61
Er	7.5	1.7	2.55	9.41	9.58

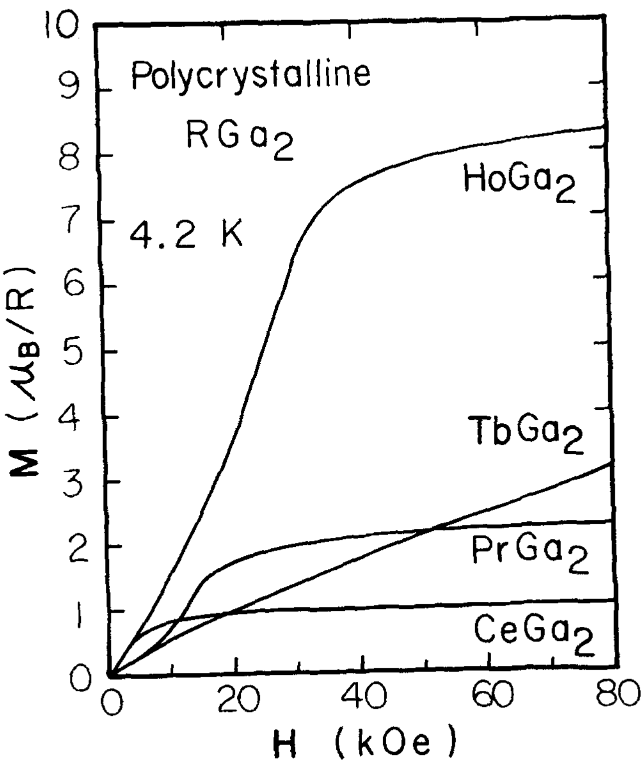

Fig. 3. Magnetization M versus applied magnetic field H at 4.2 K for polycrystalline $HoGa_2$, $TbGa_2$, $PrGa_2$, and $CeGa_2$.

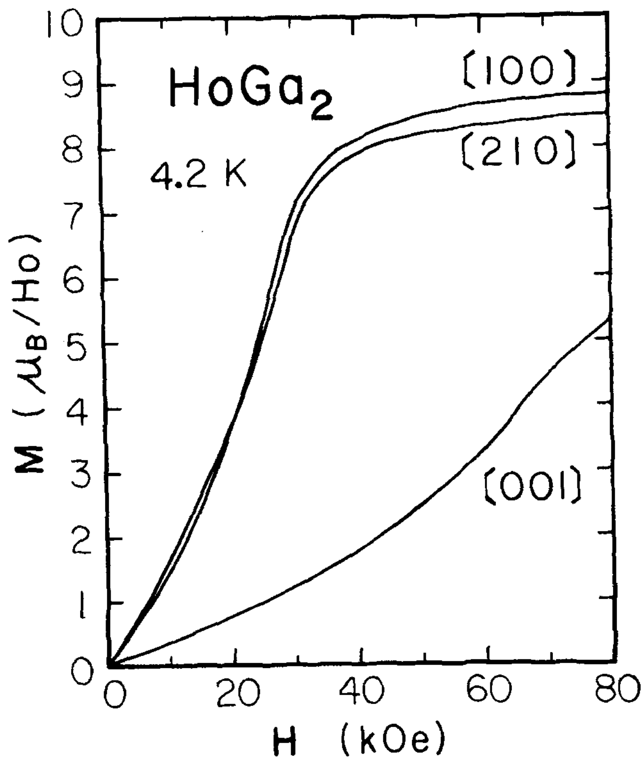

Fig. 4. Magnetization M versus applied magnetic field H for single crystal $HoGa_2$ along [100], [210], and [001] directions at 4.2 K.

III. DISCUSSION

By the RKKY theory with free electrons, the indirect exchange interaction, via the conduction electrons, between the localized rare earth moments in RGa_2 compounds can be written as [4]:

$$H = -2\Gamma^2 \sum_{i \neq j} F(\vec{r}_i - \vec{r}_j)\, \vec{S}_i \cdot \vec{S}_j \,, \tag{2}$$

where $\vec{S}_i$ is the ionic spin, $\vec{r}_i$ is the position of the ith ion, Γ is the s-f exchange integral, and F is the Lindhard function. In terms of the distance between localized moments $\vec{r}$, [5]

$$F(r) = \frac{m}{(2\pi)^3 \hbar^2} \left(\frac{\sin(2k_F r) - 2k_F r \cos(2k_F r)}{r^4} \right) . \tag{3}$$

The RKKY sum, [6]

$$\sum_{i \neq 0} F(2k_F r_{i0}) = \sum_{i \neq 0} \frac{\sin 2k_F r_{i0} - 2k_F r_{i0} \cos(2k_F r_{i0})}{5(2k_F r_{i0})\, r_{i0}^3} , \tag{4}$$

calculated for these RGa_2 compounds gives negative values, ranging from -2.267×10^{-2} Å^{-3} for $CeGa_2$ to -2.477×10^{-2} Å^{-3} for $ErGa_2$. From the negative sign of the RKKY sum [7], these compounds are predicted to be antiferromagnetic.

From Fig. 1, the $\chi(T)$ results for $HoGa_2$, $PrGa_2$, $ErGa_2$, and $DyGa_2$ show clear antiferromagnetic ordering. These samples show spin-flop transitions, i.e., metamagnetic phase transitions when the antiferromagnetic R-R couplings are overcome by the external magnetic field below T_N. This is shown in Fig. 3, most clearly for $PrGa_2$ and $HoGa_2$. The $\chi(T)$ results in

Fig. 1 for $CeGa_2$, $GdGa_2$, $NdGa_2$, and $TbGa_2$ do not exhibit typical susceptibility peaks indicating antiferromagnetic order. However, these samples also show metamagnetic phase transitions below their ordering temperatures, proving that they too are antiferromagnetic. The spin flop field for $CeGa_2$ at 4.2 K is only about 167 Oe, much too low to be observed in Fig. 3.

It should be noted also in Fig. 1 that $ErGa_2$ and $DyGa_2$ exhibit certain unusual features in their susceptibilities. For example, $\chi(T)$ for $ErGa_2$ shows a kink at 4 K which is below T_N. This may be due to a change in the nature of the order at 4 K. M(H) for $ErGa_2$ at both 4.2 K and 6 K shows evidence for spin flops at 6 kOe and 20 kOe. In addition, $\chi(T)$ for $DyGa_2$ shows a broad peak above T_N centered on about 11 K. Although this is suggestive of a crystal field effect, more measurements are needed to understand its origin.

The electrical resistivity measurements for a $CeGa_2$ polycrystalline sample show the effect of spin-disorder scattering below ~ 4 K, but it does not show any maximum or minimum around its ordering temperature, i.e., it does not show the Kondo lattice effect.

From Fig. 4, the M vs. H results for $HoGa_2$ single crystal, [100] is the easy direction and [001] is the hard direction. This result agrees with the conclusion of Barbara, et al. [8], whose neutron diffraction results showed that the direction of the moment is aligned parallel to the [100] direction. The magnetization along the [100] direction at 80 kOe, at 4.2 K, is about 8.81 μ_B/Ho.

$M(\mu_B/R)$ vs. H for polycrystalline $HoGa_2$, shown in Fig. 3, has values up to about 20 kOe which are approximately the same as the easy axis values of the single crystal $HoGa_2$ shown in Fig. 4. This is unusual if the polycrystal has random grains, which might not be the case for our samples. This is a possibility since a preferred orientation of relatively large grain may have occurred during solidification following the arc melting.

From the results in Table I, it can be seen that the de Gennes theory is inadequate for the RGa_2 compounds. That is, the experimental θ values are positive (with one exception) whereas the theory predicts them to be negative. It is also true that the θ values are not directly proportional to the de Gennes factor. Presumably this implies either that the s-f exchange integral is not constant throughout the series or that there is a more fundamental failure of the theory, for example, the free electron approximation. Since there are only three atoms per unit cell in this structure, band calculations would not be extraordinarily difficult, and they could be quite informative on this question.

Additional transport measurements and high field magnetization measurements on single crystals will be reported elsewhere.

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