Low-Temperature Magnetic Behavior of HoRh₄B₄

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Detailed measurements of the magnetization of HoRh₄B₄ and GdRh₄B₄ near the magnetic transition temperature are presented. In contrast to the ferromagnetic behavior of GdRh₄B₄, no spontaneous magnetization is found to develop below T_m in HoRh₄B₄. This raises doubts about the nature of the magnetic order in that material as well as in DyRh₄B₄ and TbRh₄B₄. Anisotropy and time dependence of the magnetization are found in these last three compounds. The behavior of the very small remanent magnetization and of the paramagnetic susceptibility of HoRh₄B₄ is shown. These results point to a complex magnetic order, possibly helical or sinusoidal antiferromagnetism with a long wavelength.

1. INTRODUCTION

Magnetic ordering is found in all the CeCo₄B₄-type rare-earth (R) rhodium borides RRh₄B₄ except for LuRh₄B₄ (in the case of NdRh₄B₄ and TmRh₄B₄ further confirmation is still needed).¹⁻³ Superconductivity occurs in RRh₄B₄ for R = Nd, Sm, Er, Tm, and Lu and appears to coexist with antiferromagnetic order in SmRh₄B₄, while it is destroyed by the onset of magnetic order in ErRh₄B₄. For R = Gd, Tb, Dy, and Ho, no superconductivity is observed and the magnetic order is thought to be ferromagnetic. The variation of the transition temperature T_m for these magnetic compounds deviates strongly from the behavior expected from the de Gennes curve; T_m is maximum (~11 K) for DyRh₄B₄. Other interesting and somewhat intriguing features are to be found in two recent papers⁴,⁵ dealing with the magnetic behavior of HoRh₄B₄. (i) The magnetization in high fields is unexpectedly small⁵ (M < 5 μ_B/Ho for H = 100 kOe). (ii) Above T = 50 K the inverse susceptibility follows closely a Curie–Weiss law with an effective moment \( \mu_{\text{eff}} = 10.55 \mu_B/\text{Ho} \) (the free ion value) and a negative intercept

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θ = −8 K; below 50 K, there is a marked downward curvature of χ^{-1}(T) and the magnetic transition is observed at 6.7 K.\(^5\) By contrast, there is practically no curvature in χ^{-1}(T) for the sample used in Ref. 4, and θ = +2 K; again the data above 50 K follow nearly perfectly a Curie–Weiss law with \(μ_{\text{eff}} = 10.5μ_B/Ho\). (iii) The magnetic contribution \(C_M(T)\) of the specific heat below \(T_m\) has a beautiful mean-field-like shape.\(^4\)

These features prompted us to study the magnetic behavior of HoRh₄B₄ in more detail. We followed the suggestion made in Ref. 5 to check whether the ideal mean field behavior observed for the magnetic specific heat reflects in the magnetization around \(T_m\). Data for GdRh₄B₄ were also taken for comparison.

### 2. EXPERIMENTAL DETAILS

The samples were synthesized by arc-melting the high-purity elements under Ar and were annealed in sealed quartz tubes (five days at 1150°C, followed by two weeks at 900°C, under Ar). Magnetic measurements were taken, on spherical and/or needle-shaped samples, in a vibrating sample magnetometer (\(ν = 22\) Hz), where the sample was held in thermal equilibrium with a large Cu block. The temperature of the sample was thus easily controlled within Δ\(T < 10^{-2}\) K near \(T_m\). Temperature measurements were done with a Ge resistance thermometer. Isothermal measurements of the magnetization were performed near \(T_m\) at temperature intervals of about 0.3 K, in fields between 0 and 13 kOe.

### 3. EXPERIMENTAL RESULTS

When the magnetic behavior of HoRh₄B₄ is compared with that of GdRh₄B₄, striking differences are found. Figure 1 shows the low-field data for spherical samples after correction for the demagnetization effect. GdRh₄B₄ behaves like a ferromagnet (\(T_m = 5.5\) K). The magnetization strongly depends on temperature and field; below \(T_m\) the initial susceptibility tends to infinity; at 1.61 K, the magnetization in a field of 150 Oe already reaches 50% of the expected saturation value of \(7μ_B/\text{Gd}\) and in 3 kOe the figure is 86% (not shown). By contrast, the initial susceptibility of a (zero-field-cooled) HoRh₄B₄ sample remains finite and appears to be independent of \(T\) below \(T_m\). This is even more apparent in an Arrott plot (\(M^2\) vs. \(H/M\)) of the magnetization (Fig. 2). For \(T > T_m\) the magnetic isotherms in moderate fields (\(H < 2\) kOe) define a set of parallel straight lines which intersect the \(H/M\) axis at reliable values of the zero-field inverse susceptibility \(χ_0^{-1}(T)\). Below 6.3 K the magnetic behavior is very peculiar. Instead of remaining parallel and cutting the \(M^2\) axis at positive values