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On the unusual magnetic behaviour of $CeRh_3B_2$

A Galatanu[1,3,4], E Yamamoto[1], T Okubo[2], M Yamada[2], A Thamizhavel[2], T Takeuchi[2], K Sugiyama[2], Y Inada[2] and Y Ōnuki[1,2]

[1] Advanced Science Research Centre, JAERI, Tokai, Ibaraki 319-1195, Japan
[2] Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

E-mail: gala@popx.tokai.jaeri.go.jp

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Abstract

$CeRh_3B_2$ is a ferromagnet with an unexpectedly high Curie temperature and low saturation magnetic moments. The origin of its unusual magnetic behaviour remains unelucidated despite various investigations performed in the last two decades. Here we present magnetic results obtained on high-quality single crystals over a wide temperature range. Some new magnetic features are uncovered and possible approaches to explaining its magnetism are briefly discussed.

1. Introduction

In the hexagonal rare-earth RT_3B_2 diborides (T = Rh, Ru, Os, Ir) [1], Ce compounds constitute anomalies with smaller volumes, due to smaller lattice constants c. This holds particularly for $CeRh_3B_2$ which is a ferromagnet with a Curie temperature reaching 120 K and exceeding that of $GdRh_3B_2$ (90 K) [2]. For $CeRh_3B_2$ the lattice parameter c (3.09 Å) is even smaller than in α-Ce and decreases further on decreasing temperature, while the lattice parameter a decreases upon increasing temperature [3]. Various investigations performed on this compound and its substituted alloys revealed a controversial localized magnetism picture in a Kondo-like scenario, however without evidence for a strong Kondo effect [3, 4]. Recently, we have started a systematic reinvestigation of $CeRh_3B_2$, performed on high-quality single crystals. Here we report detailed magnetic investigations over a wide temperature range, pointing to some previously neglected features.

2. Experimental details

High-quality single crystals of $CeRh_3B_2$ were grown by the Czochralski method in a tetra-arc furnace from polycrystals with stoichiometric composition. Laue patterns were used to

[3] Author to whom any correspondence should be addressed.
[4] On leave from: NIMP, 76900 Bucharest-Magurele, Romania.

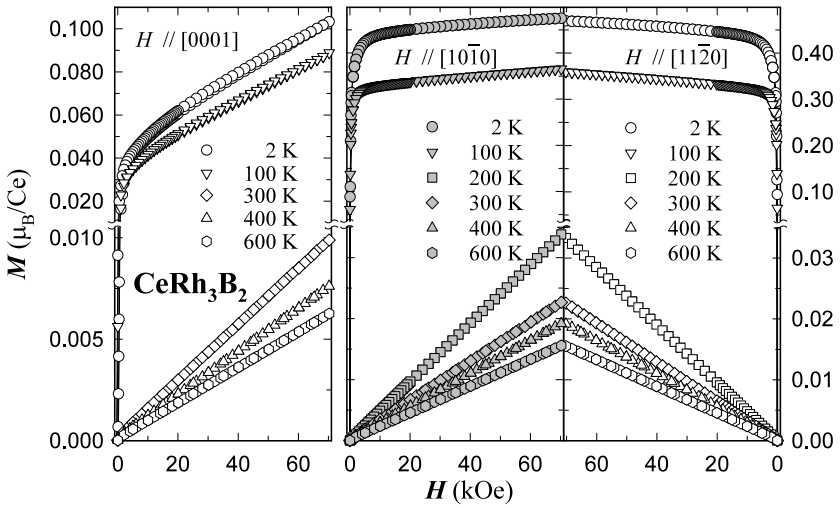

Figure 1. Magnetization isotherms for fields applied parallel to the principal axes in $CeRh_3B_2$ single crystal.

identify the crystallographic directions and to cut parallelepipedic-shaped samples. Magnetic measurements were performed using a commercial SQUID device in the temperature range 2–800 K and in fields up to 7 T. For measurements above room temperature, the samples were mounted using Cu (99.9%) wires, pre-annealed at 800 K. This procedure ensures an almost constant background for consequent measurements with estimated errors $\leqslant 1\%$.

3. Results and discussion

Figure 1 shows field-dependent magnetization isotherms obtained for both ferromagnetic and paramagnetic states. We note a strong anisotropy and ferromagnetic order with moments aligned in the basal plane, consistent with previous reports [5]. Arrott plots for $T < T_C$ evidenced the presence of spontaneous magnetic order for both directions in the basal plane in contrast to the case along the [0001] direction. The present results also revealed a difference between the saturation magnetization values in the basal plane (0.451 μ_B along [10$\bar{1}$0] and 0.447 μ_B along the [11$\bar{2}$0] direction). Along [0001] the saturation moment, taking into account the linear behaviour at higher fields, reaches only $\sim$0.04 μ_B at 2 K, less than 10% of the value in the basal plane. Since the differences in the basal plane were observed for two samples and in different measurement geometries, we considered this as an intrinsic feature and a detailed investigation of the hysteresis loops was performed at 2 K as shown in figure 2. For the basal plane the hysteresis loop width (i.e. twice the coercive force) is $\leqslant$100 Oe and no definite difference could be detected between [10$\bar{1}$0] and [11$\bar{2}$0] directions. However, for $H \parallel [10\bar{1}0]$ direction the remanent moments are slightly higher while the loop area is smaller (57.56 μ_B Oe) compared to that for $H \parallel [11\bar{2}0]$ direction (58.52 μ_B Oe), a difference of $\sim$2%, i.e. a value well above the experimental errors at this temperature. From these results it was concluded that [10$\bar{1}$0] is the easy magnetization direction (EMD) in this compound, as indicated in the structure sketch in figure 2. Here we note that Arrott plots at temperatures up to 120.5 K for $H \parallel [10\bar{1}0]$ indicated spontaneous magnetic order, while at 121 K the spontaneous order vanished (see the inset in figure 2). Thus T_C was determined as 120.8 K according to the low-field dc magnetic susceptibility.

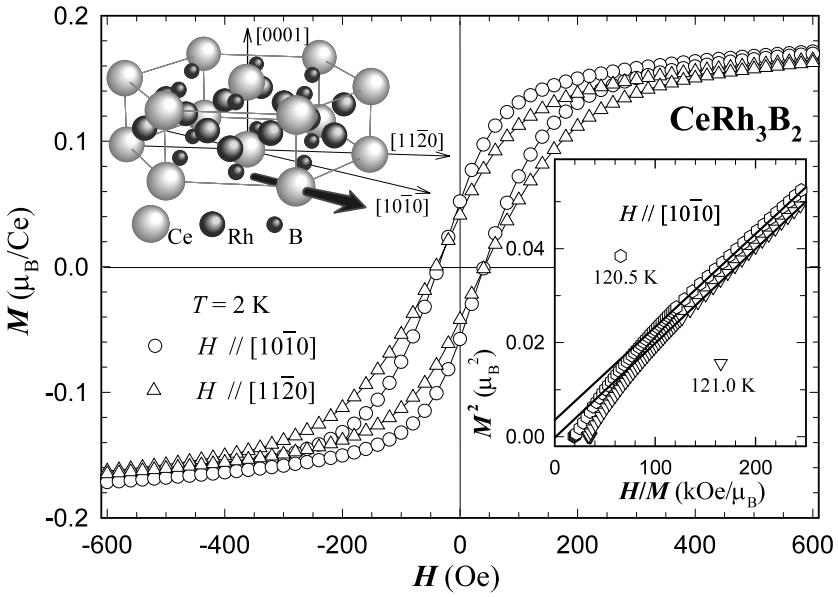

Figure 2. Magnetic hysteresis loops in the basal plane for $CeRh_3B_2$ at 2 K. The inset shows the Arrott plots at temperatures around T_C.

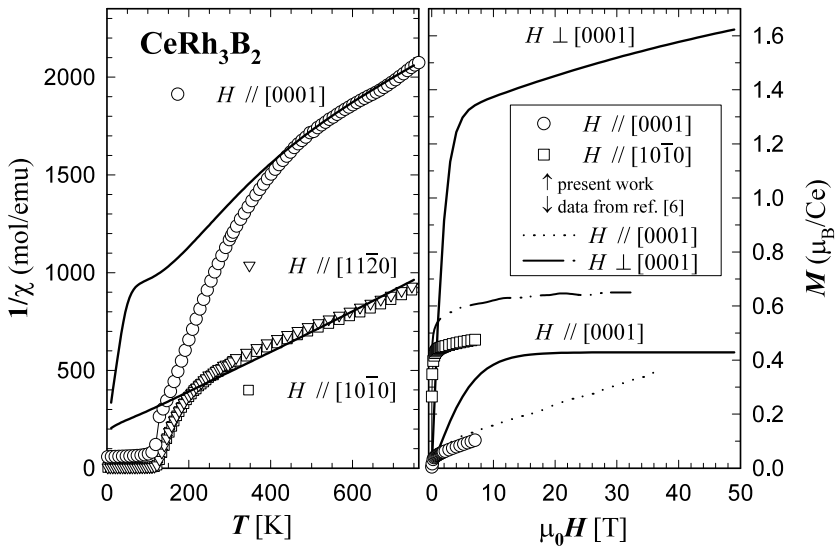

Figure 3. Analysis of the magnetic reciprocal susceptibility (left) and field-dependent magnetization (right) of $CeRh_3B_2$. The solid curves are fits described in the text.

Another interesting feature is that also in the basal plane, the high-temperature magnetic susceptibility (left panel in figure 3) is not temperature independent and follows a Curie–Weiss-like behaviour at temperatures over 600 K, yielding an effective magnetic moment $\mu_{\mathrm{eff}} = 2.9\ \mu_{\mathrm{B}}$ for $H \parallel [10\bar{1}0]$ and $[11\bar{2}0]$ ($\mu_{\mathrm{eff}} = 2.3\ \mu_{\mathrm{B}}$ for $H \parallel [0001]$ and $T \geqslant 650$ K). The associated paramagnetic Curie temperatures derived from the linear fits are large and negative. Also one can note the presence of a broad anomaly up to about 600 K along [0001]

and also in the basal plane, though less pronounced. The origin of this behaviour might be connected with a crystal field effect or with spin fluctuations as inferred also from electrical resistivity [3]. Nevertheless, the magnetic susceptibility resulting from the present data is far from linear at temperatures lower than 400 K and this result invalidates attempts to describe the magnetic behaviour based on low-temperature data.

Explaining the $CeRh_3B_2$ magnetism in a classic localized picture is at variance with the low moments in the ordered phase. In the absence of a strong Kondo effect, the magnetic moment reduction might be ascribed to strong crystalline electric field (CEF) effects, which are also a natural explanation for the anisotropic behaviour of the magnetic susceptibility. In the hexagonal symmetry, the sixfold-degenerate 4f level splits into three doublets. The susceptibility data at high temperature can be fitted using three well separated doublets at 0, 210 and 1554 K ($\lambda = -190$ mol emu^{-1}), as plotted with solid curves in figure 3, or with a mixed ground state composed from two doublets and a high-lying excited doublet $|\pm 5/2\rangle$ at 1512 K (not plotted). However, the corresponding magnetization data at 2 K cannot be described by these CEF schemes (right panel of figure 3). Here we also include, using dashed curves, the only available data obtained in higher fields [6], recorded for a different single crystal. In fact, such CEF schemes can hardly account for the magnetic behaviour of $CeRh_3B_2$ due to the value of the highest level splitting, which is of the order of the spin–orbit interaction.

Although itinerant magnetism might be able to explain the high Curie temperature and the possibility of its presence cannot be excluded for this compound [7, 8], we mention also another possible explanation. Starting from the failure of the de Gennes scaling for RRh_3B_2 compounds and considering scaling based on the quadrupolar factor, which was recently proven to hold for the ordering temperatures across the RPd_3S_4 series [9], we consider the possibility of an additional quadrupolar ordering mechanism. The quadrupolar scaling obviously favours the Ce compounds. Some other indirect features present in $CeRh_3B_2$ (e.g. a strong quadrupolar moment [10] and the anisotropic polarization of the Ce 5d electrons [11, 12]) are consistent with a quadrupolar ordering mechanism. Since the present magnetic data do not offer any evidence of such QP order, future investigations will be performed in an effort to understand the magnetism in this compound.

4. Summary

Magnetic investigations on high-quality $CeRh_3B_2$ single crystals revealed a single ferromagnetic phase with a Curie temperatures of 120.8 K and saturation magnetic moments of 0.45 μ_B at 2 K. The magnetic moments are aligned along the $[10\bar{1}0]$ direction in the basal plane. The present results rule out the possibility of a simple localized picture for the $CeRh_3B_2$ magnetism, even when CEF effects are taken into account. Other possible explanations remain to be explored.

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