Magnetic Anomalies in AlB2-type Hexagonal Ho2RhSi3 and Er2RhSi3

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Abstract. We report the results of initial magnetic measurements on the rare-earth (R) ternary compounds, R2RhSi3 (R= Ho and Er), crystallizing in a AlB2-derived hexagonal structure, hitherto not paid much attention in the past literature. While the results establish the existence of a magnetic transition around 5 K for both the compounds, dc magnetization (M) and heat-capacity behavior as a function of temperature and magnetic field based on these studies are already quite revealing. In the case of the Er compound, interestingly enough, the magnetic transition appears to be first-order-like and there is a 1/3 plateau in the isothermal magnetization curves in the magnetically ordered state, expected for ‘partially disordered antiferromagnetism (PDA)’ sometimes known for geometrically frustrated triangular magnetic insulators; there is also a bifurcation of zero-field-cooled (ZFC) and field-cooled (FC) low-field dc magnetic susceptibility (χ) curves in the magnetically ordered state – a characteristic feature of spin-glasses. The results overall reveal that there is a subtle competition between antiferromagnetism and ferromagnetism in this compound. In the case of the Ho compound, there is no indication for spin-glass anomalies and the isothermal M data reveal the existence of magnetization jumps, which are hysteretic.

INTRODUCTION

It is well-known that the Ce, Eu and Yb compounds exhibit exotic properties due to some degree of 4f electron delocalization. On the other hand, the compounds of heavy rare-earth (R) members have not drawn much attention due to essentially localized nature of the 4f electrons. However, some of heavy R intermetallic compounds were shown to exhibit Kondo-like anomalies (typical of Ce intermetallics) a few decades ago [1,2], which also led to new theoretical approaches in recent years [3]. In this respect, the rare-earth compounds of the type R2PdSi3, crystallizing in a AlB2-derived hexagonal crystal structure [4], turned out to be novel in many aspects. The most notable one is Gd2PdSi3, which was shown to exhibit unusual paramagnetic transport [1] as well as Hall anomalies across two metamagnetic transitions in the magnetically ordered state more than two decades ago [5], which is now called ‘Topological Hall effect’ in the current literature. In other words, this concept of great current interest in solid state physics branch is actually much older than what is believed to be [6]. Considering such recent upsurge in exploring interesting properties of such ternary heavy rare-earth compounds, it is important to investigate isostructural Rh based compounds. This family was not explored in depth for a long time after initial reports a few decades ago [4, 7], though Ce compound was studied in depth [8]. Recently, it was reported [9] that R=Gd, Tb and Dy members of R2RhSi3 family are characterized by magnetic and transport properties comparable to those of Gd3PdSi3. In this study, we report the magnetic properties of Ho2RhSi3 and Er2RhSi3 where the 4f states are further away from the Fermi level having relatively less contributions in the conduction band. The results bring out hitherto unknown magnetic anomalies for these compounds.

Before we present the results of our investigations, it is important to recall crystallographic features relevant to this article and the readers may find the details in earlier reports [4, 9]. Si and Rh atoms are ordered in planes perpendicular to c-axis, forming a honeycomb network. The rare-earth layer made up of triangular network alternate along c-axis with respect to Rh-Si layer. Due to the ordering of Pd and Si, the lattice parameters, a and c, are doubled with respect to that for AlB2 structure, as evidenced by weak superstructure lines at the low angle side in the x-ray diffraction pattern.
EXPERIMENTAL DETAILS

Polycrystalline samples have been prepared by melting together stoichiometric amounts of constituent elements in an arc furnace in an atmosphere of argon under partial pressure, followed by annealing at 1073 K for about a week in evacuated sealed quartz tubes. Experimental details are curtailed due to space limitations, but are available in the past literature [4,7,8,9]. An analysis of the x-ray diffraction patterns by Rietveld refinement methods confirms single phase nature of these compounds (not shown due to space limitations). De susceptibility ($\chi$), isothermal magnetization ($M$) and heat-capacity ($C$) measurements were performed as a function of temperature ($T$) and magnetic field ($H$), as described in Ref. 9.

RESULTS AND DISCUSSIONS

We first focus on the Er compound. In fig.1, we show the behavior of $\chi(T)$ measured with 100 Oe and 5 kOe in various ways. Inverse $\chi(T)$ measured in 5 kOe is found to be linear over a wide $T$-range (10 – 300 K) and the effective moment ($\mu_{\text{eff}}$) obtained from the slope of the plot (~9.69 $\mu_B$) is in excellent agreement with the theoretical value of 9.7$\mu_B$ for trivalent Er ions. The paramagnetic Curie temperature ($\theta_p$) is ~1.4 K with the positive sign suggesting the existence of ferromagnetic interactions. As the temperature is lowered (for the zero-field-cooled (ZFC) condition), there is a rather broad upturn around 5 K (when measured with 5 kOe) (see, left inset of fig.1a). In order to have a closer look of this upturn, we have obtained the $\chi(T)$ data below 20 K for the ZFC, field-cooled (FC) and field-cooled-warming (FCW) conditions with 100 Oe; the curves obtained are shown in fig. 1b in the $T$-region of interest, 2-7K. It is distinctly clear that there is a sharp jump in the ZFC curve, as though the virgin state exhibits a first-order-like transition; in support of this, the FCC and FCW curves, though broadened and shifted strangely to lower temperature marginally (by about 0.4 K, the reason for which is unclear), are found to be hysteretic. Additionally, unlike the curve in fig. 1a inset obtained in 5 kOe, there is a peak in ZFC curve at about 3.2 K and FCW curve deviates from ZFC curve at this temperature, increasing down to 2 K. This finding is a characteristic feature of cluster spin-glasses.

We now focus on the isothermal $M(H)$ behavior (measured with ZFC condition) in the magnetically ordered state, shown in fig. 1c for 1.8 and 4 K. The curves are essentially nonhysteretic. It is transparent that, following an initial sharp increase possibly due to the response from the magnetic domains to the application of $H$, there is a plateau till 20 kOe and then a sharp increase in the $M(H)$ plot of the data at 1.8 K; this step is a bit smoothened in the 4 K plot. The variation of $M$ with $H$ is very weak beyond 50 kOe (not shown), as though there is a tendency to saturate. The linear extrapolation of the high-field curve to zero field yields a value of about 6.5 $\mu_B$/Er, which is lower than the saturation value expected for fully degenerate trivalent Er ions (9 $\mu_B$); such a reduction could be due to crystal-field effects. The most intriguing observation is that the value of the magnetic moment by a similar extrapolation in the plateau region is ~2.15$\mu_B$/Er, which is essentially one-third of the extrapolated high-field saturation moment. Such a 1/3 magnetization plateau in triangular lattices is expected for a situation when the magnetic ions at one of the three vortices of the triangle is magnetically disordered, while the other two are antiferromagnetically coupled in zero field in the virgin state; when the compound is in a field in the plateau region, the magnetic moment of the ‘disordered’ Er ion gets oriented and becomes antiparallel to one of the two Er ions, leaving behind a net spontaneous moment from the third Er ion; at higher fields beyond 25 kOe, the magnetic moment from all the three Er ions get ferromagnetically aligned resulting in thrice of magnetic moment at the plateau. This kind of magnetism in zero-field for the virgin state is called ‘Partially Disordered Antiferromagnetism (PDA)’ due to geometrical frustration of the magnetic moments. Typical examples for such a PDA magnetism known in the past literature [10, 11] are CsCoCl$_3$ and Ca$_3$CoRhO$_6$, containing triangular magnetic lattices and such compounds are insulators. Such a PDA magnetism was not considered in the analysis of neutron diffraction pattern in the previous report (reported for 1.5 K only) [7].

We now bring out an interesting feature in the $C(T)$ data, measured also in the presence of external fields while warming after ZFC to 2 K. There is no worthwhile feature in the data above 8 K (measured till 150 K) and therefore we show the data in fig. 1d below 10 K only. As expected, in the zero-field data, there is a $\lambda$-anomaly well below 6 K, with the upturn and the sharp peak occurring very close to 5 K due to the onset of magnetic ordering; no additional prominent peak could be detectable down to 2 K, even in the $C/T$ curve.
The interesting finding is that, for $H = 10$ kOe, the $\lambda$-peak is shifted to a marginally higher temperature, which is usually a characteristic feature of ferromagnetism; further applications of $H$ suppresses the peak temperature, typical of antiferromagnets. Clearly, there is a subtle competition between ferromagnetism and antiferromagnetism with the application of $H$. The isothermal entropy change, defined as $\Delta S = S(H) - S(0)$, obtained by integrating $C/T$ curves, exhibits a negative peak typical of field-induced ferromagnetic alignment (fig. 1f). Surprisingly, it changes sign sharply at the onset of magnetic transition for 10 kOe, as though there are antiferromagnetic clusters before the onset of long range magnetic order; the peak values are reasonably large, say, for $0 \rightarrow 50$ kOe field-variation, spread over a wide $T$-range to enable possible applications at low temperatures, given that isothermal $M$ curves are nonhysteretic.

We now focus on the Ho compound. We show the results of $\chi$, isothermal $M$, and $C$ measurements in fig. 2. $\chi(T)$ exhibits Curie-Weiss behavior (fig. 2b, inset) above its magnetic transition temperature of 5.2 K, with the value of $\mu_{\text{eff}} (-10.63 \, \mu_B)$ in agreement with the theoretical value of Ho$^{3+}$ ion. The value of $\theta_p = -5.4$ K with the negative sign suggesting antiferromagnetic correlations; since the magnitude is in good agreement with the observed ordering temperature, we infer that there is negligible competition from ferromagnetic correlations, at least in low fields. There is a peak at 5.2 K, without any worthwhile bifurcation of ZFC-FC curves measured in 100 Oe (fig. 2a), thereby implying absence of glassiness of magnetism down to 2 K. An interesting finding in the ZFC curve measured with 100 Oe is that there is a drop of $\chi$ at 5.1K – that is, as soon as the compound enters magnetically ordered state – followed by a relatively broader peak; such a drop is not observed in FCC and FCW curves. Clearly, there is a subtle magnetic anomaly around 5 K sensitive to the way the measurements are done for this compound. Isothermal $M$ behavior is shown for 1.8 and 4 K in fig. 2b. The curve at 1.8 K is found to be hysteretic and a careful look of the
virgin curve reveals that there is an upward curvature around 5 kOe, attributable to spin-reorientation. There are additional sharp jumps at higher fields (35 and 47 kOe); while reversing the field, these jumps vanish, but the change of slope due to spin-reorientation persists. At 4 K, these steps are absent, though a careful look at the curve provides evidence for spin-reorientation in the low-field range.

In fig. 2c, we show the $C(T)$ data below 30 K (though measured till 60 K). There is a well-defined $\lambda$-anomaly with a sharp peak at 5 K in zero field due to magnetic ordering, which is suppressed to about 3 K for an application of 10 kOe; for higher fields, this feature is suppressed to a temperature below 2 K (as indicated by a weak upturn in $C/T$ below 4 K, not shown here). There is also a broad peak in zero-field in the range 5-20 K, possibly due to Schottky anomaly associated with crystal-field effects; this peak persists for 10 kOe, but it is smeared for higher fields. The $\Delta S$, shown in fig. 2d, below 60 K, exhibits a peak, shifting to higher temperatures with increasing (final) $H$ due to the depression of magnetic transition temperature. The fact that the sign is negative suggests field-induced ferromagnetic alignment. The values at the minimum of the curve are reasonably large, as in the case of Er compound, say, for a change of field 50 kOe.

**CONCLUSIONS**

We bring out interesting magnetic anomalies in Ho$_2$RhSi$_3$ and Er$_2$RhSi$_3$, based on initial magnetic investigations. In the case of Ho compound, there is an interesting magnetic transition around 5.2 K and the magnetic features are sensitive to the way measurements are done and isothermal $M$ is hysteretic showing spin reorientation effects. Jumps in the virgin isothermal magnetization data below 5 K are observed. With respect to Er compound, which is found to order at 5 K, it is notable that the Er compound exhibits an interesting magnetic transition around 5 K, in addition characterized by ‘partially disordered antiferromagnetic’ features due to geometrically frustrated magnetism – an observation unusual among intermetallics. In order to get better insight into this, ac $\chi$ and remnant magnetization studies will be carried out. Details will be published elsewhere. It would be rewarding to carry out neutron diffraction studies as a function of temperature and magnetic field.

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**REFERENCES**