Field-induced magnetic transitions in the quasi-two-dimensional heavy-fermion antiferromagnets Ce\textsubscript{n}RhIn\textsubscript{3n+2} (n = 1 or 2)

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Abstract

We have measured the field-dependent heat capacity in the tetragonal antiferromagnets CeRhIn\textsubscript{5} and Ce\textsubscript{2}RhIn\textsubscript{8}, both of which have an enhanced value of the electronic specific heat coefficient $\gamma \sim 400$ mJ/mol-Ce K$^2$ above $T_N$. For $T < T_N$, the specific heat data at zero applied magnetic field are consistent with the existence of an anisotropic spin-density wave opening a gap in the Fermi surface for CeRhIn\textsubscript{5}, while Ce\textsubscript{2}RhIn\textsubscript{8} shows behavior consistent with a simple antiferromagnetic magnon. From these results, the magnetic structure, in a manner similar to the crystal structure, appears more two-dimensional in CeRhIn\textsubscript{5} than in Ce\textsubscript{2}RhIn\textsubscript{8} where only about 12% of the Fermi surface remains ungapped relative to 92% for Ce\textsubscript{2}RhIn\textsubscript{8}. When $B||c$, both compounds behave in a manner expected for heavy fermion systems as both $T_N$ and the electronic heat capacity decrease as field is applied. When the field is applied in the tetragonal basal plane ($B||a$), CeRhIn\textsubscript{5} and Ce\textsubscript{2}RhIn\textsubscript{8} have very sim-
ilar phase diagrams which contain both first- and second-order field-induced magnetic transitions.

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I. INTRODUCTION

Ce$_n$RhIn$_{2n+3}$ ($n = 1$ or $2$) crystallize in the quasi-two-dimensional (quasi-2D) tetragonal structures Ho$_n$CoGa$_{2n+3}$, and both are moderately heavy-fermion antiferromagnets ($\gamma \sim 400$ mJ/mol-Ce K$^2$ for both systems above $T_N = 3.8$ K for $n = 1$ and 2.8 K for $n = 2$). The evolution of the ground states of CeRhIn$_5$ as a function of applied pressure, including a pressure-induced first-order superconducting transition at 2.1 K, is unlike any previously studied heavy-fermion system and is attributed to the quasi-2D crystal structure [1]. In a similar manner to CeRhIn$_5$, $T_N$ is seen to change only slightly with pressure and abruptly disappear in Ce$_2$RhIn$_8$, though superconductivity has not yet been observed [2].

A previous zero field heat capacity study on CeRhIn$_5$ revealed that the anisotropic crystal structure leads to a quasi-2D electronic and magnetic structure [3]. We have performed measurements of the heat capacity in applied magnetic fields for CeRhIn$_5$ and Ce$_2$RhIn$_8$ in an attempt to further understand the electronic and magnetic properties of these compounds. We find that for magnetic fields applied along the tetragonal $c$-axis, both systems behave like typical heavy-fermion compounds [4] as both $T_N$ and $\gamma_0$ decrease as field is increased. Very different behavior is seen when the field is directed along the $a$-axis as $T_N$ is found to increase and numerous field-induced transitions, both of first- and second-order, are observed. These transitions correspond to magnetic field-induced changes in the magnetic structure. In agreement with what one might expect, the magnetic properties seem less 2D as the crystal structure becomes less 2D going from single layer CeRhIn$_5$ to double layer Ce$_2$RhIn$_8$ (note that as $n \to \infty$, one gets the 3D cubic system CeIn$_3$).

II. RESULTS

Single crystals of Ce$_n$RhIn$_{2n+3}$ were grown using a flux technique described elsewhere [5]. The residual resistivity ratio between 2 K and 300 K using a standard 4-probe measurement and was found to be greater than 100 for all measured crystals, indicative of high-quality
samples. A clear kink was observed in the resistivity at $T_N = 3.8$ K for CeRhIn$_5$ and $T_N = 2.8$ K in Ce$_2$RhIn$_8$. Magnetization measurements have shown that the effective magnetic moment is slightly reduced from the value expected for Ce$^{3+}$ due to crystal fields [3]. The specific heat was measured on a small ($\sim 10$ mg) sample employing a standard thermal relaxation method.

The single crystals were typically rods with dimensions from 0.1 to 10 mm with the long axis of the rod found to be along the $\langle 100 \rangle$ axis of the tetragonal crystal. The samples were found to crystallize in the primitive tetragonal Ho$_n$CoGa$_{2n+3}$-type structure [1,4] with lattice parameters of $a = 0.4652(1)$ nm and $c = 0.7542(1)$ nm for $n = 1$ and $a = 0.4665(1)$ nm and $c = 1.2244(5)$ nm for $n = 2$ [1,2]. The crystal structure of Ce$_n$RhIn$_{2n+3}$ can be viewed as $(\text{CeIn}_3)_n(\text{RhIn}_2)$ with alternating $n$ cubic (CeIn$_3$) and one (RhIn$_2$) layers stacked along the $c$-axis. By looking at the crystal structure, we would expect that AF correlations will develop in the (CeIn$_3$) layers in a manner similar to bulk CeIn$_3$ [5]. The AF (CeIn$_3$) layers will then be weakly coupled by an interlayer exchange interaction through the (RhIn$_2$) layers which leads to a quasi-2D magnetic structure. This has been shown to be true as the moments are AF ordered within the tetragonal basal $a$-plane but display a modulation along the $c$-axis which is incommensurate with the lattice for CeRhIn$_5$ [1]. As $n$ is increased, the crystal structure should become more 3D ($n = \infty$ being the 3D cubic system CeIn$_3$) and the effects of the interlayer coupling should become less important causing the magnetic and electronic structure to be more 3D. Indeed, the magnetic structure in Ce$_2$RhIn$_8$ does not display an incommensurate spin density wave (SDW) [10].

The zero field data from specific heat measurements are shown in Fig. 1. A peak at $T_N$ is clearly seen for both samples, indicating the onset of magnetic order. The entropy associated with the magnetic transition is $\sim 0.3 R \ln 2$ with the remaining $0.7 R \ln 2$ recovered by 20 K for both $n = 1$ and 2. For $T > T_N$ the data could not be fit by simply using $C/T = \gamma + \beta_l T^2$, where $\gamma$ is the electronic specific heat coefficient and $\beta_l$ is the lattice Debye term. As found previously, one needs to use isostructural, nonmagnetic La$_n$RhIn$_{2n+3}$ to subtract the lattice contribution to $C$ [1]. After subtracting the lattice contribution, it is
still difficult to extract a value of $\gamma$ from the data. However, by performing a simple entropy balance construction, a value of $\gamma \approx 400 \text{ mJ/mol-Ce K}^2$ is found for both $n = 1$ and 2.

For temperatures below $T_N$, as found before on CeRhIn$_5$ [3], the magnetic heat capacity data, where the corresponding La compound is used to subtract the lattice contribution, can be fit using the equation

$$C_m/T = \gamma_0 + \beta_M T^2 + \beta_M' \left(e^{-E_g/k_BT}\right) T^2$$

where $\gamma_0$ is the zero temperature electronic term, $\beta_M T^2$ is the standard AF magnon term, and the last term is an activated AF magnon term. The need for an activated term to describe heat capacity data been seen before in other Ce and U compounds [3,11–13], and the term rises from an AF SDW with a gap in the excitation spectrum due to anisotropy. As discussed previously, the CeRhIn$_5$ magnetic structure indeed displays an anisotropic SDW with modulation vector $(1/2,1/3,0.297)$ [4] which is consistent with this picture. The inset to Fig. 1 shows the data for $T < T_N$ and the lines are fits to Eq. 1 for $T^2 < (0.85T_N)^2$. We find that the the activated term is NOT necessary to fit the Ce$_2$RhIn$_8$ data. Rather, there appears to be a small feature in the heat capacity centered around ~2 K whose origin is unknown. This feature is also observed in transport measurements and is known to persist as a function of pressure [14]. A summary of the fit parameters, along with data on CeIn$_3$ [15], is given in Table I. These results lead us to the conclusion that the magnetically ordered state in CeRhIn$_5$ consists of an anisotropic SDW that opens up a gap on the order of 8 K in the Fermi surface, while no such gap is seen in Ce$_2$RhIn$_8$, consistent with its commensurate structure. Note that the values for CeRhIn$_5$ are slightly different from a previous report where the lattice contribution from LaRhIn$_5$ was not subtracted from the raw data [3]. From the ratio of the electronic contribution for temperatures above and below $T_N$, we estimate that approximately $\gamma_0/\gamma \sim 0.12$ (12%) of the Fermi surface remains ungapped below $T_N$ for CeRhIn$_5$ while for Ce$_2$RhIn$_8$ 92% of the Fermi surface remains ungapped. The results on CeIn$_3$ of Berton et al. are also shown in Table I for comparison where $\gamma_0/\gamma \sim 0.056$ (5.6%). Clearly, the electronic structure, as evidenced by the ratio $\gamma_0/\gamma$, becomes more 3D in the
Ce$_n$RhIn$_{2n+3}$ series as $n$ is increased. 

Fig. 2 shows the heat capacity in applied magnetic fields for $B||c$ for both compounds (CeRhIn$_5$ on the top and Ce$_2$RhIn$_8$ on the bottom). As the magnetic moments are known to lie within the $a$-plane CeRhIn$_5$, the magnetic field is perpendicular to the magnetic moments in the ordered state in this orientation. The applied field is not sufficient to cause a field-induced magnetic transition in either compound. Rather, it is found that $T_N$ and $\gamma_0$ decrease as $B$ is increased as is usually observed in heavy fermion systems.

Fig. 3 shows the heat capacity in applied magnetic fields for $B||a$. For both samples, the Neel point (the onset of antiferromagnetic order), and magnetic field-induced transitions of both first- ($T_1$) and second-order ($T_2$) are clearly observed. The complete phase diagrams for both CeRhIn$_5$ and Ce$_2$RhIn$_8$ showing the various observed transitions are plotted in Fig. 4. The open symbols correspond to second order transitions ($T_N$ and $T_2$) and the filled symbols represent the first order transition ($T_1$). The dashed lines are merely guides for the eyes. Remarkably, the phase diagrams for both CeRhIn$_5$ and Ce$_2$RhIn$_8$ are extremely similar. Region I corresponds to the standard modulated spin density wave that is incommensurate with the lattice as reported previously in CeRhIn$_5$. The nature of the first- and second- order transitions going from Region I to Regions II and III are not known, and work is underway to determine the magnetic structures in Regions II and III. For Ce$_2$RhIn$_8$, Region II terminates at 70 kOe while for CeRhIn$_5$ it extends beyond the highest measured field of 90 kOe. The first order transition going from Region I or Region II to Region III is the same hysteretic field induced transition observed in magnetization measurements on CeRhIn$_5$ where an increase in the magnetization of $\lesssim 0.006 \mu_B/Ce$. These results clearly show that Ce$_2$RhIn$_8$ has some 2D electronic and magnetic character.

**III. CONCLUSION**

In summary, we have measured the anisotropic heat capacity in applied magnetic fields in the quasi-2D heavy-fermion antiferromagnets CeRhIn$_5$ and Ce$_2$RhIn$_8$. The magnetic and
electronic properties of CeRhIn$_5$ can be well explained by the formation of an anisotropic SDW, leading to a 2D electronic and magnetic structure. The phase diagram of magnetic field-induced magnetic transitions is remarkably similar for both systems as both a first- and second-order transition are observed in both compounds when the magnetic field is along the tetragonal $a$-axis. From the heat capacity measurements, we estimate that ~12% (92%) of the Fermi surface remains ungapped below the magnetic ordering temperature for CeRhIn$_5$ (Ce$_2$RhIn$_8$). The 2D nature of the electronic properties is a result of the tetragonal crystal structure of Ce$_n$RhIn$_{2n+3}$ which consists of $n$ cubic (CeIn$_3$) blocks which are weakly interacting along the $c$-axis through a (RhIn$_2$) layer. Not surprisingly, it appears that the case of $n = 1$ (CeRhIn$_5$) is more anisotropic than the double layered Ce$_2$RhIn$_8$ where the amount of ungapped Fermi surface below $T_N$ is greater than 90%, as is found for the 3D system ($n = \infty$) CeIn$_3$. The similarity of the field-induced transitions in both CeRhIn$_5$ and Ce$_2$RhIn$_8$ clearly shows that there is still some 2D character to the magnetic properties of Ce$_2$RhIn$_8$. The results here shed light on the unusual magnetic and electronic structure of CeRhIn$_5$ and Ce$_2$RhIn$_8$.

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[16] The software used to measure the heat capacity uses average values to fit the thermal relaxation data. From the raw traces, the transitions which we have assigned as first order are clearly first order in nature. Experience tells us the averaging method underestimates the value of the heat capacity near the peak of the first order transition by a factor of four or five.
TABLE I. Calculated zero field heat capacity fit parameters for Ce\textsubscript{n}RhIn\textsubscript{2n+3}. *Data for CeIn\textsubscript{3} is taken from Ref. 15. The various parameters are defined in the text. Units for $\gamma_0$ and $\gamma$ are mJ/mol Ce-K\textsuperscript{2} and for $\beta_M$ and $\beta'_M$ are mJ/mol Ce-K\textsuperscript{4}.

| \( n \) | \( T_N \) (K) | \( \gamma_0 \) | \( \gamma \) | \( \beta_M \) | \( \beta'_M \) | \( E_g/k_B \) (K) |
|---|---|---|---|---|---|---|
| CeRhIn\textsubscript{5} | 1 | 3.72 | 56 | 400 | 24.1 | 706 | 8.2 |
| Ce\textsubscript{2}RhIn\textsubscript{8} | 2 | 2.77 | 370 | 400 | 93.2 | - | - |
| *CeIn\textsubscript{3} | $\infty$ | 10 | 136 | 144 | 15 | - | - |
FIGURES

FIG. 1. The zero field magnetic specific heat $C_m$ divided by temperature $T$ versus $T^2$ for CeRhIn$_5$ and Ce$_2$RhIn$_8$. The inset displays the range $T^2 < 15$ K$^2$ corresponding to temperatures below $T_N$. The lines are fits described in the text.

FIG. 2. The magnetic specific heat $C_m$ divided by temperature $T$ versus $T$ for CeRhIn$_5$ (top) and Ce$_2$RhIn$_8$ (bottom) in various applied fields applied along the $c$-axis.

FIG. 3. The magnetic specific heat $C_m$ divided by temperature $T$ versus $T$ for CeRhIn$_5$ (top) and Ce$_2$RhIn$_8$ (bottom) in various applied fields applied along the $a$-axis. $T_N$ corresponds to the antiferromagnetic ordering temperature, and $T_1$ and $T_2$ correspond to field-induced first- and second-order transitions respectively. The curves are offset by 1 J/mol Ce-K$^2$ for each successive curve.

FIG. 4. The cumulative phase diagrams for CeRhIn$_5$ and Ce$_2$RhIn$_8$ for various applied fields applied along the $a$-axis. $T_N$ corresponds to the antiferromagnetic ordering temperature, and $T_1$ and $T_2$ correspond to field-induced first- and second-order transitions respectively. The dashed lines are guides to the eyes. The magnetic structure in Regions I and II is a spin density wave that is incommensurate with the lattice where Region II has a larger magnetic moment on each Ce atom. Region III corresponds to a spin density wave that is commensurate with the lattice.
