Magnetic structure of heavy fermion $\text{Ce}_2\text{RhIn}_8$

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The magnetic structure of the heavy fermion antiferromagnet $\text{Ce}_2\text{RhIn}_8$ is determined using neutron diffraction. It is a collinear antiferromagnet with a staggered moment of $0.55(6)\mu_B$ per Ce at 1.6 K, tilted $38^\circ$ from the tetragonal $c$ axis. In spite of its layered crystal structure, the phases for magnetic moments are the same as those in the cubic parent antiferromagnet CeIn$_3$. This suggests that the cubic CeIn$_3$ building blocks have a stronger influence on magnetic correlations than intervening layers, which gives the material its apparent two-dimensional lattice structure and renders CeRhIn$_5$ an incommensurate antiferromagnet.

Superconducting heavy fermion materials belong to a special class of correlated electron systems where unconventional superconductivity may be mediated by magnetic fluctuations. Until recently, there were only five U-based heavy fermion materials showing superconductivity at ambient pressure [2] in addition to the original heavy fermion superconductor CeCu$_2$Si$_2$ [3]. Three Ce-based heavy fermion materials isostructural to CeCu$_2$Si$_2$ and cubic CeIn$_3$ become superconductors under pressure. Recently, superconductivity has been discovered in a new structure class of heavy fermion materials with chemical formulas Ce$M$In$_5$. While the $M$=Rh member superconducts below 2.1 K under 17 kbar [4], the $M$=Ir and Co members superconduct below 0.4 K and 2.3 K, respectively, at ambient pressure [5,6]. The high superconducting transition temperatures of the new materials hold the record for heavy fermion superconductors. Thermodynamic and transport measurements at low temperature are consistent with unconventional superconductivity in which there are lines of nodes in the superconducting gap.

Because CeIn$_3$ and CeMIn$_5$ belong to the Ce$_nM_m$In$_{3n+2m}$ family of structures, they present a unique opportunity for investigating the influence of systematic structure modifications on the superconducting and magnetic properties [8]. In particular, it is interesting to compare CeIn$_3$ and CeRhIn$_5$, which are the $n=\infty$ and $n=1$ members of the Ce$_n$In$_{3n+2}$ family, and can be viewed as periodic stacking of $n$-layers of CeIn$_3$ on a layer of $M$In$_2$ [15,16]. Both are antiferromagnetic at ambient pressure with $T_N=10$ K for CeIn$_3$ [17] and $T_N=3.8$ K for CeRhIn$_5$ [16]. Both become superconductors when subjected to pressure [18], with the superconducting transition temperature of CeRhIn$_5$ being one order of magnitude higher than that for CeIn$_3$. This raises a fundamental question about the role of the intervening $M$In$_2$ layers on both the superconductivity and antiferromagnetism. Our study of Ce$_2$RhIn$_8$, which is the $n=2$ member of this heavy fermion sub-family, is intended to shed light on this question by changing the ratio of the CeIn$_3$ and RhIn$_2$ layers.

Antiferromagnetic structures for both CeIn$_3$ and CeRhIn$_5$ have been determined previously. Cubic CeIn$_3$ has a simple commensurate magnetic order with wave vector $(1/2,1/2,1/2)$ below its Néel temperature. The staggered magnetic moment is $0.55\mu_B$ per Ce in Ce$_2$RhIn$_8$ and it points $38^\circ$ from the $c$ axis. The solid circle denotes Ce, the shaded circle In, and the open circle Rh. The disk for CeRhIn$_5$ denotes the plane in which the ordered moment rotates.

![Figure 1](image-url) FIG. 1. The magnetic structure of Ce$_2$RhIn$_8$ ($n=2$) in a structural unit cell is shown together with CeRhIn$_5$ ($n=1$) [16] and CeIn$_3$ ($n=\infty$) [14,15]. The magnetic moment is $0.55\mu_B$ per Ce in Ce$_2$RhIn$_8$ and it points $38^\circ$ from the $c$ axis. The solid circle denotes Ce, the shaded circle In, and the open circle Rh. The disk for CeRhIn$_5$ denotes the plane in which the ordered moment rotates.
suggests a strong influence of the cubic CeIn$_3$ structural unit on magnetic correlations in this family of heavy fermion materials.

Single crystals of Ce$_2$RhIn$_8$ were grown from an In flux. They crystallize in the tetragonal Ho$_2$CoAs$_3$ structure (space group #123, P4/mmm), with lattice parameters $a = 4.665\,\text{Å}$ and $c = 12.244\,\text{Å}$ at room temperature. The sample used in this study was a well-faceted rectangular plate of dimension $\sim 4 \times 4 \times 0.7\,\text{mm}$ and weight of 88 mg. The largest surface is the (001) plane. Neutron diffraction experiments were performed at NIST using the thermal triple-axis spectrometer BT2 in a two-axis mode. The horizontal collimations were 60–40–40–open. Neutrons with incident energy $E = 35\,\text{meV}$ were selected using the (002) reflection of a pyrolytic graphite (PG) monochromator. The neutron penetration length at this energy is 1.8 mm, which is substantially longer than the thickness of the sample. No rocking-angle dependent absorption was noticed. PG filters of total 9 cm thickness were used to remove higher order neutrons. The sample temperature was regulated by a top loading pumped He cryostat.

Temperature-dependent magnetic Bragg peaks were found at $(m/2,n/2,l)$ with $m$ and $n$ odd integers and $l$ non-zero integers. This corresponds to a magnetic unit cell that doubles the structural unit cell in the basal plane and contains four magnetic Ce ions. Rocking scans at $(1/2,1/2,0)$ and $(1/2,1/2,-1)$, taken at 1.6 K, are shown in Fig. 2(a). The intensity of the $(1/2,1/2,1)$ peak is shown in Fig. 2(b) as the square of the order parameter of the magnetic phase transition. Integrated intensities of magnetic Bragg peaks from such rocking scans are normalized to structural Bragg peaks $(001), (002), (003), (005), (006)$ and $(220)$ to yield magnetic scattering cross sections, $\sigma(q) = I(q)\sin(\theta_d)$, in absolute units (see Table I). In such units, the magnetic cross section is [17]

| $q$ | $\sigma_{obs}$ | $\sigma_{calc}$ |
|-----|----------------|-----------------|
| $(0.5, 0.5, -1)$ | 52(1) | 46.2 |
| $(0.5, 0.5, 0)$  | 0.0(3) | 0.0  |
| $(0.5, 0.5, 1)$  | 49(1) | 46.2 |
| $(0.5, 0.5, 2)$  | 19(1) | 18.9 |
| $(0.5, 0.5, 3)$  | 6.4(4) | 5.5  |
| $(0.5, 0.5, 4)$  | 21(1) | 23.2 |
| $(0.5, 0.5, 5)$  | 7.5(7) | 7.6  |
| $(1.5, 1.5, 0)$  | 0.0(8) | 0.0  |
| $(1.5, 1.5, 1)$  | 18(1) | 24.8 |

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\[
\sigma(q) = 4 \left( \frac{\gamma r_0}{2} \right)^2 \langle M \rangle^2 |f(q)|^2 \sum_{\mu,\nu} (\delta_{\mu\nu} - \hat{q}_\mu \hat{q}_\nu) F_\mu(q) F_\nu(q),
\]

where $(\gamma r_0/2)^2 = 0.07265\,\text{barns}/\mu_B^2$, $M$ is the staggered moment of the Ce ion, $f(q)$ the Ce$^{3+}$ magnetic form factor [18], $\hat{q}$ the unit vector of $q$, and $F_\mu(q)$ the $\mu$th Cartesian component of magnetic structure factor per Ce$_2$RhIn$_8$.

Forbidden peaks at $(m/2,m/2,0)$ provide an important clue to the magnetic structure of Ce$_2$RhIn$_8$. They could be due to magnetic moments aligning along the $[110]$ direction. However, magnetic twinning in this tetragonal material will yield finite intensities at these reciprocal points. Another, more reasonable, cause is that the nearest-neighbor magnetic moments along the $c$ axis are antiparallel. The phase between the next nearest-neighbor magnetic moments along the $c$ axis and the phases of magnetic moments in a basal layer are already determined by the magnetic wave vector. This yields a collinear antiferromagnetic structure (refer to Fig. 3) with magnetic cross sections per Ce$_2$RhIn$_8$

\[
\sigma(q) = 4 \left( \frac{\gamma r_0}{2} \right)^2 \langle M \rangle^2 |f(q)|^2 (1 - (\hat{q} \cdot \hat{S})^2) \sin^2(\kappa l),
\]

where $2\epsilon = 0.38c$ is the separation between the nearest-neighbor Ce ions along the $c$ axis, $\hat{S}$ is the unit vector of the magnetic moment, and the average, $(1 - (\hat{q} \cdot \hat{S})^2)$, is over magnetic domains.

Fig. 3 shows $\sigma_{obs}(q)/|f(q)|^2 \sim (1 - (\hat{q} \cdot \hat{S})^2) \sin^2(\kappa l)$.
as a function of the $l$ index of $\mathbf{q}$. The structure factor, $4\sin^2(\ell \kappa)$, not only accounts for the forbidden $l = 0$ magnetic peaks, but it also accounts for the strong oscillation of $\sigma_{\text{obs}}$ as a function of $l$. The remaining, smooth $l$ dependence is to be accounted for by the polarization factor $(1 - (\mathbf{q} \cdot \mathbf{s})^2)$.

Denote the angle between $\mathbf{q}$ and the basal plane as $\alpha$, and the angle between the basal plane and the magnetic moment as $\beta$. Assuming equal occupations among magnetic twins, we have

$$\sigma_{\text{obs}}(l) \sim (1 - (\mathbf{q} \cdot \mathbf{s})^2) \cos^2 \alpha \cos^2 \beta + 2 \sin^2 \alpha \sin^2 \beta \alpha.$$  

For a magnetic moment lying in the basal plane, $\beta = 0$, which is the case for CeRhIn$_5$, the polarization factor varies too much. The resulting theoretical curve does not fit the data (refer to the dot-dashed line in Fig. 3). For $\beta = 35.26^\circ$, which corresponds to $\mathbf{s}$ in the (111) directions in a cubic system, the polarization factor averages to a constant, $2/3$. The resulting theoretical curve is a better fit (refer to the dotted line in Fig. 3) than that for $\beta = 0$, but it is still not satisfactory. The best least-squares fit (refer to the solid and dashed lines for $h = k = 1/2$ and $h = k = 3/2$ respectively) yields $\beta = 52(2)^\circ$. The staggered magnetic moment is determined at 1.6 K to be $M = 0.55(6) \mu_B$ per Ce.

Having determined the magnetic structure of Ce$_2$RhIn$_8$, now we consider the systematics relating the magnetic structure and lattice structure in Ce$_n$RhIn$_{3n+2}$ (see Fig. 3). In the $a$-$b$ plane, the magnetic moments of the Ce ions form a square lattice, surrounded by In ions, in all three materials. They all are simple, nearest-neighbor antiferromagnets in the basal plane. In CeRhIn$_5$, this Ce antiferromagnetic plane alternates with the RhIn$_2$ layer. Magnetic correlations across the RhIn$_2$ layer are incommensurate, with neighboring magnetic moments being rotated by 107$^\circ$ [16]. The local structure environment in the vertical $a$-$c$ or the $b$-$c$ plane within the CeIn$_3$ double layer in Ce$_n$RhIn$_8$ is very similar to that in the basal layer. The same nearest-neighbor antiferromagnetic correlations exist in the double layers. It is interesting that now across the RhIn$_2$ layer the Ce moments are antiparallel instead of rotated by 107$^\circ$. The insertion of the RhIn$_2$ layers between CeIn$_3$ bilayers, thus, does not modify the magnetic order relative to cubic CeIn$_3$. This suggests CeRhIn$_8$ as a unique member of the Ce$_n$RhIn$_{3n+2}$ family, and the $n \geq 2$ members are likely to be magnetically similar to cubic CeIn$_3$. Searching for heavy fermion materials with two-dimensional magnetism seems more profitable if one could find a Ce$_m$In$_{3+2m}$ structure family, where $m$ In$_2$ layers separate a single CeIn$_3$ layer.

Another interesting difference between the $n = 1$ and the $n = 2$ materials concerns the magnetic moment orientation. In CeRhIn$_5$, the moments rotate in the $a$-$b$ plane, indicating a $XY$ type magnetic anisotropy. In Ce$_2$RhIn$_8$, the magnetic moments point 52$^\circ$ from the basal plane. Different local anisotropic fields, together with isotropic exchange and crystal fields, likely contribute to the different magnetic structures in the two materials [19]. We also notice that the staggered moment of Ce$_2$RhIn$_8$ is comparable to that of CeIn$_3$, while $T_N$ of Ce$_2$RhIn$_8$ is closer to that of CeRhIn$_5$. The $T_N/|M|^2$ is the smallest for Ce$_2$RhIn$_8$ in the group. Antiferromagnetic transition has been studied in isostructural Nd$_n$MnIn$_{3n+2}$ ($M$=Rh,Ir and $n = 1,2$) and crystal field effects have been emphasized [21]. A detailed understanding of the magnetic interaction in these materials is essential in pursuing magnetic origin of unconventional superconductivity in these heavy fermion metals.

In conclusion, we find the magnetic structure of Ce$_2$RhIn$_8$ to be closely related to that of cubic CeIn$_3$. The staggered moment is $0.55(6) \mu_B$ per Ce at 1.6 K and it points 52$^\circ$ from the $a$-$b$ plane. Understanding the different magnetic structures in Ce$_n$RhIn$_8$ and CeRhIn$_5$ may help us understand the enormous enhancement in the superconducting transition temperature of CeRhIn$_5$ over its cubic relative.

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