Pressure-induced change in the magnetic modulation of $CeRhIn_5$

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We report the results of a high-pressure neutron-diffraction study of the heavy-fermion compound $CeRhIn_5$ down to 1.8 K. $CeRhIn_5$ is known to order magnetically below 3.8 K with an incommensurate structure. The application of hydrostatic pressure up to 8.6 kbar produces no change in the magnetic wave vector $\mathbf{q}_m$. At 10 kbar of pressure, however, a sudden change in the magnetic structure occurs. Although the magnetic transition temperature remains the same, $\mathbf{q}_m$ increases from (0.5, 0.5, 0.298) to (0.5, 0.5, 0.396). This change in the magnetic modulation may be the outcome of a change in the electronic character of this material at 9 kbar.

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There is considerable evidence that in heavy-fermion systems the existence of *partially Kondo compensated low moment magnetic order* is associated with the occurrence of pressure-induced superconductivity. A wealth of Ce based compounds with a magnetically ordered ground state, such as $CeCu_2Ge_2$, $CePd_2Si_2$, and $CeIn_3$,[1–5] have been found to show superconductivity under an applied hydrostatic pressure. The application of pressure gradually destroys the magnetic order of the system and eventually a superconducting state emerges beyond the critical pressure P_c. In contrast to normal BCS superconductors, it is believed that the magnetic spin fluctuations rather than the lattice vibrations effectively bind the electrons (more precisely, the heavy electrons) into Cooper pairs.[6] Many of these compounds also show non-Fermi-liquid behavior in the vicinity of the magnetic-nonmagnetic phase boundary.[7] In the last few years there has been considerable experimental as well as theoretical effort to understand the possible role of magnetic interactions in creating a superconducting ground state.[5,8]

The recently discovered heavy-fermion compound $CeRhIn_5$ provides a useful opportunity to investigate the coupling between magnetism and superconductivity because of its reasonably high transition temperature ($T_c \sim 2$ K) in an accessible pressure (16 kbar) range. This material is also different in that there appears to be a sharp boundary where superconductivity develops and then exists over an extended pressure range with little variation in T_C.

$CeRhIn_5$ crystallizes in a tetragonal $HoCoGa_5$ structure with alternating layers of $CeIn_3$ and $RhIn_2$ arranged along the c axis. At ambient pressure it orders magnetically at $T_N = 3.8$ K into a c-modulated incommensurate structure.[9] Neutron diffraction[10] and nuclear quadrapole resonance[11] experiments indicate that below T_N the magnetic moment at each Ce site is $0.26\mu_B$ with the spins lying in the basal plane. In the basal plane, the magnetic moments adopt a nearest-neighbor antiferromagnetic (AFM) structure, while along the c axis they form an incommensurate spiral with a pitch of $\delta = 0.298$, corresponding to a turn angle of 107° between the successive $CeIn_3$ layers. The resulting magnetic modulation vector is $\mathbf{q}_m = (0.5, 0.5, 0.298)$. The application of hydrostatic pressure up to 9 kbar has little effect on the value of T_N. Beyond 16 kbar of pressure, resistivity and heat-capacity data suggest that $CeRhIn_5$ undergoes a first-order-like transition to a superconducting ground state ($T_c \sim 2$ K) with the sudden disappearance of the 3.8 K magnetic phase.[9] Resistivity versus temperature data[9] indicate a small sharp drop around the temperature $T_? \approx 3$ K above 9 kbar of pressure, which continues to exist in the superconducting state (above 16 kbar of pressure). The signature of the long-range magnetic order in the the heat capacity versus temperature data above 3 kbar is indicated by a rather broad maximum (T_{max}), which is close to the ambient pressure value of T_N up to 9 kbar. Although, heat-capacity measurements[12] fail to resolve $T_?$ and T_N in the pressure range 9 to 15 kbar, they indicate a gradual shift of T_N (as indicated by T_{max}) to lower temperatures above 10 kbar of pressure. Above P_c, a very broad anomaly around 3 K, presumably of magnetic origin, continues to exist in the heat capacity versus temperature data, which matches closely with the value of $T_?$ obtained from the resistivity data.

The interpretation of both resistivity and heat-capacity experiments as to the onset of long-range order is not straightforward. In the case of resistivity, the very small change observed at $T_?$ could result from many causes. For heat-capacity data there is a smooth evolution in the shape of the peaked structure, which is clearly associated with T_N at atmospheric pressure but may have other origins at higher applied pressure. Neutron experiments definitively map the onset of an ordered moment through the appearance of Bragg reflections from the magnetic structure.

The temperature-pressure phase diagrams of Ce based heavy-fermion superconductors[5] such as $CeIn_3$ and $CePd_2Si_2$ show a gradual decrease of T_N with increasing pressure before the onset of superconductivity. In case of $CeRhIn_5$, T_N is quite robust with respect to the application pressure up to 10 kbar. The superconductivity also occurs with a sudden disappearance of T_N at P_c.

It is therefore essential to understand the nature of the magnetic phase of $CeRhIn_5$ which is stable at high pressure. Bao *et al.*[13] reported neutron-diffraction data of $CeRhIn_5$ at 3.8 kbar. They observed a magnetic modulation vector of $\mathbf{q}_m = (0.5, 0.5, 0.294)$, which is almost identical to the ambient pressure value. In this paper we report the results of a high-pressure (up to 13 kbar) neutron-diffraction study of the magnetic ordering of $CeRhIn_5$. Our investigation shows no variation in the magnetic structure up to a pressure of 8.6 kbar. Above 10 kbar of pressure we observed a marked change in $\mathbf{q}_m$ from (0.5, 0.5, 0.298) to (0.5, 0.5, 0.396). Despite this change in the $\mathbf{q}_m$, T_N remained constant up to 11 kbar of pressure. At 11 kbar, no anomaly was detected in the magnetic reflections in the vicinity of $T_? = 3$ K, the temperature at which an unidentified feature was seen in the resistivity data.[9] Surprisingly, at 13 kbar we failed to observe any magnetic reflections along the (0.5, 0.5, ℓ) direction for temperatures above our base of 1.8 K.

Single crystals of $CeRhIn_5$ were grown using the indium flux technique.[14] For this experiment, a bar shaped ($1.5 \times 1.5 \times 4$ mm^3) crystal was cut from a large single crystal. The crystal was mounted with the [110] direction vertical. This enabled us to reduce the absorption of neutrons by the indium nuclei for our diffraction scans along the direction of the reciprocal lattice vector (0.5, 0.5, ℓ) ($\ell = 0$ to 1). The high-pressure neutron-diffraction experiment was carried out using the thermal-neutron diffractometer D10, at the Institut Laue-Langevin, Grenoble, France, in the two-axis mode to accommodate a large He-flow cryostat. A clamp-type alumina pressure cell (inner diameter 6 mm and outer diameter 7 mm) was used for the generation of hydrostatic pressure with fluorinert as the pressure transmitting medium. The monochromatic neutron beam of wavelength 2.359 Å used for the diffraction experiment was obtained by reflection from a pyrolytic graphite monochromator. In order to reduce the background counts from the pressure cell and the cryostat, a pyrolytic graphite analyzer was included for most of the scans. A sodium chloride crystal was placed alongside the sample in the pressure cell. Its lattice parameters were determined independently, which enabled us to measure the actual pressure in the cell *in situ* by comparing the known pressure dependence of the lattice parameter of sodium chloride with these measurements.

In order to investigate the nature of the magnetic phase at high pressure, we performed the diffraction study at different hydrostatic pressures (8.6, 10, 11, and 13 kbars). Before applying each pressure, the sample was brought back to ambient pressure and room temperature to avoid any irreversibility arising from pressure and temperature cycling. Initially, the crystal was aligned at ambient pressure in the pressure cell and we observed a magnetic Bragg peak at $\mathbf{q}_m = (0.5, 0.5, 0.2983 \pm 0.0004)$ at 1.8 K, which is in good agreement with the value reported by Bao *et al.*[10] A hydrostatic pressure of 8.6 kbar was applied and the crystal was scanned along the (0.5, 0.5, ℓ) direction ($\ell = 0$ to 1) at 1.8 K. We observed a pair of magnetic peaks at $\ell = 0.2989$ and 0.6980 (see Fig. 1) corresponding to a modulation vector $\mathbf{q}_m = (0.5, 0.5, 0.298)$. We did not find any other magnetic peaks along this direction. This magnetic peak vanishes at 3.8 K, showing that the value of T_N remains unchanged at 8.6 kbar of pressure. The pressure was then increased to 11 kbar to observe any change in the magnetic structure. A scan along (0.5, 0.5, ℓ) failed to show any magnetic Bragg peaks around $\ell = 0.3$ or 0.7 at 1.8 K. Instead, we observed a pair of magnetic peaks at $\ell = 0.396$ and 0.604 (see Fig. 1). For a spiral magnetic structure along the *c* axis with a nearest-neighbor AFM arrangement in the basal plane, the expected magnetic reflections should occur at $\mathbf{q}_m = (h/2, k/2, \ell \pm \delta)$, where $h, k = \pm 1, \pm 3, \ldots, \ell = 0, \pm 1, \pm 2, \ldots$ and δ is the pitch of the spiral. Indeed, magnetic peaks were also observed at the positions (0.5, 0.5, 1.396), (1.5, 1.5, 0.396), (1.5, 1.5, 0.604), (1.5, 1.5, 1.396), and (1.5, 1.5, 1.604) with relative intensities that agree with a model of basal-plane moments that propagate in a simple helical spiral along the *c* direction. Within our experimental accuracy, no anomalous change in the position, width, or intensity of the magnetic peak with temperature was observed at this pressure. In Fig. 2, we have plotted the temperature dependence of the magnetic peak (0.5, 0.5, 0.396) at 11 kbar of pressure. The peak intensity decreases to the background value above 3.8 K. No magnetic reflection was seen at the commensurate positions (0.5, 0.5, 0) or (0.5, 0.5, 0.5). We also scanned the weak (110) nuclear peak at 1.8 and 5 K to detect any possible signature of ferromagnetic structure. Within the measurement limit, there was no change in the intensity of the (110) peak.

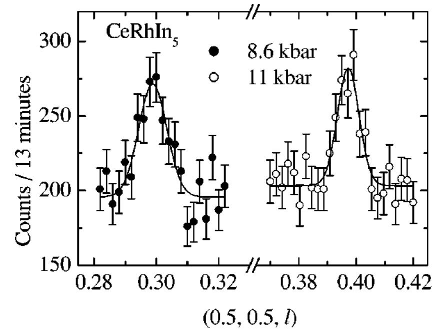

FIG. 1. Elastic neutron scans through the magnetic Bragg peaks of $CeRhIn_5$ at 8.6 kbar and 11 kbar at 1.8 K, showing the change in the value of the magnetic modulation vector with pressure.

In order to obtain a clearer picture of the functional dependence of $\mathbf{q}_m$ with pressure, we have also investigated the magnetic structure at 10 kbar and 13 kbar. The diffraction scan at 10 kbar also showed a modulation of (0.5, 0.5, 0.396), similar to that observed at 11 kbar. From this observation it appears that $\mathbf{q}_m$ does not change continuously in the pressure range 9 kbar $\leqslant P \leqslant 11$ kbar, rather it rapidly changes its value between 8.6 and 10 kbars. It should be

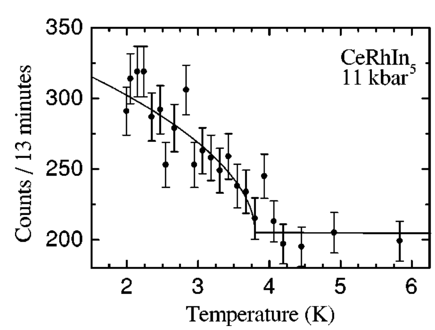

FIG. 2. Temperature dependence of the magnetic peak (0.5, 0.5, 0.396) at 11 kbar of pressure. The line through the data points is a guide to the eye.

noted at this point that the unidentified anomaly in the resistivity versus temperature data appears beyond a pressure value of 9 kbar. The T_N calculated from the heat capacity versus temperature data also shows a downward shift in temperature with an increase of pressure beyond 10 kbar. Therefore, the pressure at which we observed the change in $\mathbf{q}_m$ matches closely with the pressure values where the anomalies in resistivity and heat capacity were seen.

However, at 13 kbar of pressure, we were unable to detect any magnetic reflections in our diffraction scans (at 1.8 K) along the (0.5, 0.5, ℓ) direction. We have also searched for magnetic satellites in other directions, e.g., (1, 1, ℓ), (h, k, 0.296), (h, k, 0.396), (h, k, 1), where ℓ changes from 0 to 0.5 and h and k change from 0.45 to 0.55. No magnetic reflections were observed in these measurements.

Considering the fact that in $CeRhIn_5$ the quasi-two-dimensional $CeIn_3$ building blocks are essential for the magnetism, it is pertinent here to make a comparison with the bulk $CeIn_3$ compound. The compound $CeIn_3$ crystallizes in a cubic structure with a lattice parameter $a(300\ \mathrm{K}) = 4.73$ Å. It orders antiferromagnetically below 10 K with a $\mathbf{q}_m$ of (0.5, 0.5, 0.5) (Ref. 15) and a superconducting state emerges at 25.5 kbar[4] of pressure with the subsequent quenching of the antiferromagnetic state. The lattice parameter $a(300\ \mathrm{K})$ for $CeRhIn_5$ is 4.65 Å,[15] which implies that the $CeIn_3$ building blocks are in a compressed state as compared to the bulk $CeIn_3$ compound. Unlike $CeIn_3$, $CeRhIn_5$ shows a spiral magnetic structure. This kind of spiral magnetic structure is very common in the rare-earth systems and often occurs due to competing magnetic interactions.[16] The major factors responsible for the magnetic structure in rare-earth systems are: (i) the long-range oscillatory exchange interactions of the Rudermann-Kittel-Kasuya-Yosida (RKKY) type, (ii) the anisotropy energy resulting from crystalline electric field, and (iii) the magnetoelastic anisotropy.

The free-electron mediated RKKY exchange term $[\mathcal{J}(\mathbf{r})]$ is distance dependent and the sudden change of q_m in $CeRhIn_5$ above 9 kbar may occur because of a change in the interlayer spacing beyond a critical value $[c(1\ \mathrm{bar})/c(10\ \mathrm{kbar}) = 1.0052]$. Although measured in a high magnetic field, de Haas–van Alphen experiments in this compound indicate an anisotropic Fermi surface with some kind of spin-density wave transition occurring at 1.2 K.[17] From the Fermi surface point of view, the wave-vector dependent free-electron susceptibility, $\chi(q)$ is important in determining $\mathcal{J}(\mathbf{r})$ and for a stable spiral structure $\chi(q)$ should have a maximum at a nonzero value of q. It may be possible that the change in $\mathbf{q}_m$ is an outcome of the shift in $\chi(q)$ under pressure resulting from a change in the nesting characteristics of the Fermi surface. It is also interesting to note that both T_N and $\mathbf{q}_m$ are resistant to any change up to a pressure as high as 8.6 kbar. The crystal field and the magnetoelastic anisotropy may not have a direct involvement in changing $\mathbf{q}_m$, as no change in the ratio of the lattice parameters c/a was observed at different pressure values.

The change in $\mathbf{q}_m$ in the pressure range 8.6 to 10 kbar signifies a change in the turn angle of the spiral spin structure from 107° to 142.6° (see Fig. 3). The effective magnetic moments of $CeRhIn_5$ (μ_m) at 8.6 and 11 kbars were also calculated from the integrated intensity of the magnetic Bragg peaks. We observed a 20% reduction of the moment at 11 kbar as compared to the 8.6 kbar $[\mu_m(11\ \mathrm{kbar})/\mu_m(8.6\ \mathrm{kbar}) = 0.8 \pm 0.1]$ value. Earlier investigations[13] indicate no change in μ_m up to a pressure value of 3.8 kbar. The observed change in μ_m may be due to an increase in the Kondo coupling strength along with the increase of pressure. At 11 kbar we did not observe any anomaly in the magnetic scattering around 3 K. Therefore, the unidentified 3 K anomaly in the resistivity data[9] above 9 kbar may not be related to the observed change in the magnetic ordering mechanism. The heat-capacity measurements[12] did not show any anomaly at 3 K in this pressure range. Nevertheless, our experiment certainly signifies a change in the magnetic structure in the same pressure range where the unidentified 3 K anomaly develops in resistivity measurements.

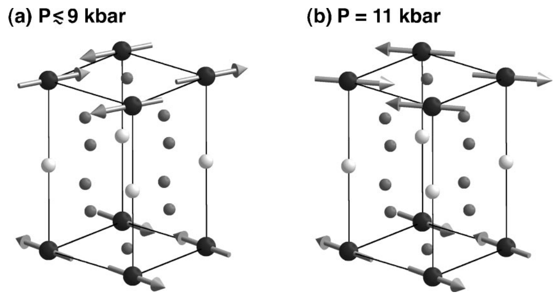

FIG. 3. An illustration of the magnetic structure of $CeRhIn_5$ (a) below 9 kbar and (b) at 11 kbar. The black spheres with spins attached are the Ce atoms, while the big white spheres and small gray spheres are Rh and In atoms, respectively.

The failure to observe any magnetic reflection at 13 kbar of pressure does not necessarily indicate that the system has attained a nonmagnetic ground state. It might be difficult to observe the magnetic reflection because: (i) the magnetic moment is reduced below the detectable limit, (ii) T_N has decreased below 1.8 K, (iii) the magnetic structure adopts a completely different modulation symmetry, or (iv) the short-range correlations dominate at all temperatures. Interestingly, the pressure dependence of entropy shows a second-order discontinuity around 12 kbar (Ref. 12) and this might be related to the absence of long-range magnetic order at 13 kbar. The high-pressure heat-capacity measurements[12] also indicate that the magnetic anomaly becomes broader and weaker with increasing pressure, which might be an indication of the loss of magnetic moment. Considering $CeRhIn_5$ is a layered compound, a broadening of the heat-capacity peak may also indicate that only anisotropic short-range order or two-dimensional magnetic correlations exist in the high-pressure state of the system.

Summarizing, this high-pressure neutron-diffraction study of $CeRhIn_5$ certainly supports the observed result by heat capacity and resistivity measurements that T_N remains unchanged up to a pressure value of 10 kbar. In addition, it indicates a change in the magnetic modulation vector in the pressure range 9–10 kbar. But, in contrast to the results of heat capacity or resistivity measurements, this experiment fails to find any ordered magnetic state at 13 kbar of pressure above 1.8 K. The nature of the magnetism in the superconducting state is of interest and a neutron-scattering experiment covering a wider pressure range (above P_c) is required to understand the coexistence of magnetism with superconductivity. Such an experiment is planned for the near future.

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