Anisotropic three-dimensional magnetic fluctuations in heavy fermion CeRhIn$_5$

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CeRhIn$_5$ is a heavy fermion antiferromagnet that orders at 3.8 K. The observation of pressure-induced superconductivity in CeRhIn$_5$ at a very high $T_C$ of 2.1 K for heavy fermion materials has led to speculations regarding to its magnetic fluctuation spectrum. Using magnetic neutron scattering, we report anisotropic three-dimensional antiferromagnetic fluctuations with an energy scale of less than 1.7 meV for temperatures as high as 3$T_C$. In addition, the effect of the magnetic fluctuations on electrical resistivity is well described by the Born approximation.

The discovery of superconductivity in heavy fermion materials by Steglich et al. just over two decades ago generated much of our current thinking on the relation between magnetism and superconductivity. In the intervening years these insights have been extended from the three-dimensional (3D) cerium and uranium-based heavy fermion materials, where the physics is thought to follow from the competition between the single site Kondo effect and the RKKY interaction between spins on different sites, to the layered cuprates in which the superconductivity and magnetism are believed to derive from a two-dimensional (2D) phenomenon. Anisotropic superconductivity arising via the exchange of antiferromagnetic spin fluctuations has been investigated theoretically, and it is predicted that 2D antiferromagnetic spin fluctuations are superior to 3D fluctuations in elevating the superconducting transition temperatures.

Very recently, a family of heavy fermion compounds with chemical formula CeMIn$_5$ ($M =$ Rh, Ir, Co) has been discovered. These materials, with the tetragonal HoCoGa$_5$ structure (space group #123, P4/mmm), consist of alternating layers of the cubic heavy fermion antiferromagnet CeIn$_3$ and the transition metal complex MIn$_2$. They display antiferromagnetism and superconductivity in close proximity. Their superconducting transition temperatures, $T_C$, are very high for heavy fermion systems. For example, $T_C$ is 2.1 K for CeRhIn$_5$ at 16 kbar, which is more than half of its Néel temperature at ambient pressure, while the maximum $T_C$ is 0.2 K for cubic CeIn$_3$ at 25 kbar is only 2% of its $T_N$ at ambient pressure. The $T_C$ of the ambient pressure superconductor, CeCoIn$_5$, $T_C$ = 2.3 K, is the highest among all heavy fermion superconductors.

The enhancement of $T_C$ in layered CeMIn$_5$ over CeIn$_3$ has been suggested to be due to the quasi-2D structure of the new materials, taking advantage of favorable coupling of 2-D antiferromagnetic fluctuations. Here we describe an inelastic neutron scattering and electrical transport study of the magnetic fluctuations for CeRhIn$_5$ in the vicinity of $T_N$ which demonstrate that this hypothesis, in its simplest form, does not seem applicable.

CeRhIn$_5$ ($a = 4.652\AA$, $c = 7.542\AA$ at 295 K) is an incommensurate antiferromagnet below $T_N = 3.8$ K. The small magnetic moments of the Ce ions, $0.26\mu_B$ at 1.4 K, form a helical spiral along the $c$ axis and are antiparallel for nearest-neighbor pairs in the tetragonal basal plane, resulting in magnetic Bragg peaks below $T_N$ at $q_M = (m/2, n/2, l \pm \delta)$ with $m$ and $n$ odd integers, $l$ integer and $\delta = 0.297$.

If CeRhIn$_5$ is magnetically 3-D, magnetic fluctuations are expected to be enhanced in the vicinity of $q_M$ near $T_N$. On the other hand, if CeRhIn$_5$ is 2-D, magnetic fluctuations are enhanced in rods which go through magnetic Bragg points along the $c^*$ axis of the same $m$ and $n$ indices.

Although single crystal samples of CeRhIn$_5$ with cm$^3$ size can be readily grown in our laboratory from an In flux, the high slow-neutron absorption coefficients of In and Rh forced us to employ a thin plate-like sample. To reduce absorption effects, we also used neutrons of incident energy $E_i = 35$ meV, selected with a pyrolytic graphite (PG)(002) monochromator, in our neutron scattering experiments at NIST. The instantaneous magnetic correlation function, $S(q)$, is usually measured with the two-axis method [14]. However, for technical reasons, we chose the three-axis method for most of this study. With PG (002) analyzers and horizontal collimations 40-40-40 and 60-40-40-open at the thermal neutron triple-axis spectrometers BT9 and BT2, respectively, the energy window, given by the half-width-at-half-maximum (HWHM) of incoherent scattering, is 1.7 meV. As will be shown below, this is much wider than the energy scale of magnetic fluctuations in CeRhIn$_5$ for the temperature range of interest. This three-axis configuration offers a better signal-to-noise ratio than the two-axis method for the correlated magnetic fluctuations in CeRhIn$_5$. No sample-angle dependent absorption is observed in our data. PG filters of 4 or 5 cm thickness were inserted in the incident neutron beam to remove higher order neutrons. The sample temperature was regulated using a heater installed in our top-loading pumped He cryostat.

The inset to Fig. 1(a) shows energy scans at the magnetic Bragg point $(1/2,1/2,0.297)$. Solid circles and
where \( f(q) \) is the magnetic form factor for a Ce\(^{3+} \) ion, and

\[
I \sim |f(q)|^2 \sum_{\mu,\nu} (\delta_{\mu\nu} - \hat{q}_\mu \hat{q}_\nu) S^{\mu\nu}(q),
\]

where \( S^{\mu\nu}(q) = \hbar \int d\omega S^{\mu\nu}(q, \omega) \) and

\[
S^{\mu\nu}(q) = \sum_{R \neq 0} e^{i\mathbf{q} \cdot \mathbf{R}} \langle M^{\mu}_0(t) M^{\nu}_0(t) \rangle.
\]

FIG. 1. (a) Intensity of magnetic fluctuations, \( S(q) - \langle M \rangle^2 \), at \( q = (1/2,1/2,0.297) \) (solid circles) as a function of temperature. Neutron scattering intensity measured at \( (0.7,0.7,0.297) \) (open circles) indicates the background. Temperature derivative of resistivity (open diamonds, scale on the right), measured with current along the \( a \) axis, closely follows magnetic fluctuations. Inset: Constant \( q = (1/2,1/2,0.297) \) scans at 7K. This implies that the energy of the scans at 4 and 7 K. This is larger than the elastic energy resolution so as to avoid the Bragg peak intensity below \( T_N = 3.8 \) K, but well inside the energy window for magnetic excitations [refer to the inset in Fig. 1(a)]. The peak at \( T_N \) reflects the divergent magnetic fluctuations, \( S(qM) - (M)^2 \), at a continuous phase transition, rounded by the finite resolution of the spectrometer. Open circles were measured at 1 meV and \( (0.7,0.7,0.297) \) which is away from any magnetic Bragg points. The contrast between the T-independent intensity here, which serves as a measure of the background, and the strongly T-dependent signal at \( (1/2,1/2,0.297) \) reflects the strong spatial modulation of \( S(q) \).

Magnetic resistivity, or angular integration of intensity of conduction electrons scattered by magnetic fluctuations, has been calculated by Fisher and Langer [16] using the Born approximation. The long mean-free-path for conduction electrons observed in our sample, \( l/a \sim 10^2 \) near \( T_N \), estimated from the measured resistivity [see Fig. 3(b)] and Hall coefficient [17], implies that the anomalies in \( d\rho/dT \) and in magnetic specific heat at \( T_N \) are directly related to the singularity in coherent scattering of conduction electrons by magnetic fluctuations, \( S(q) - (M)^2 \). Line-connected circles in Fig. 1(a) are experimental \( d\rho/dT \), and solid circles in Fig. 1(b) are magnetic specific heat, obtained by subtracting specific heat of LaRhIn\(_5\) from the total specific heat of CeRhIn\(_5\) (open circles) [3]. The similarity among these quantities indicates the prominence of the Fisher-Langer mechanism in CeRhIn\(_5\) at low temperature. This behavior has previously been observed in ferromagnetic metals, such as Ni [18], and antiferromagnets, such as PrB\(_6\) [19]. However, it is astonishing that the Born approximation is sufficient to account for the influence of magnetic fluctuations on the electrical transport in this heavy fermion material.

We now turn to the spatial dependence of magnetic correlations in CeRhIn\(_5\). A survey of \( S(q) \) along the \( c \) axis and in tetragonal basal plane intersecting Bragg points \( (1/2,1/2,0,\delta) \) and \( (1/2,1/2,1,\delta) \) is shown in Fig. 2. The solid circles were measured at 4 K. Strong modulation of \( S(q) \) with peaks at magnetic Bragg points, in scans along both the \( c \) axis [Fig. 2(a)] and in the basal plane [Fig. 2(b) and (c)], is apparent, indicating 3-D magnetic fluctuations. Fitting the data to an infinite sum of Lorentzians, centered at \( (1/2,1/2,1,\pm \delta) \), we obtain magnetic correlation lengths at 4 K of \( \xi_{\parallel} = 9.5(1)A \) along the \( c \) axis and \( \xi_{\parallel} = 23(1)A \) in the basal plane. While \( \xi_{\parallel} \) is about 5 times the nearest-neighbor distance, \( a \), of Ce ions in the basal plane, \( \xi_{\parallel} \) is only 1.3 times the inter-plane Ce distance, \( c \). However, this is different from a
FIG. 2. Instantaneous magnetic correlation function, $S(q)$, for (a) $q = (1/2, 1/2, l)$, and for $q$ along the (110) direction at (b) $l = 0.297$ and (c) $l = 1.297$ at various temperatures. Open circles in (a) are background measured along $(0.4, 0.4, l)$. Crosses indicate the magnetic Bragg peak positions below $T_N = 3.8$ K. The shaded peak is the magnetic Bragg peak measured at 1.7 K and the dashed line represents its intensity divided by 120.

2-D magnetic system, in which the intraplanar magnetic correlation length is orders of magnitude longer than the interplanar magnetic correlation length.

With increasing temperature (diamonds at 4.5 K, triangles at 5 K and squares at 7 K in Fig. 2), the magnetic peak intensity and correlation lengths are quickly reduced. At 7 K, little modulation in $S(q)$ can be detected either along the $c$ axis [Fig. 2(a)] or in the basal plane [Fig. 2(b)], indicating the loss of magnetic correlations above 7 K. Temperature dependences of inverse $\xi_\parallel$ and $\xi_c$ in units of their respective inter-Ce distances are shown in Fig. 3(a). That they vary with $T$ in the same way implies that the magnetic system in CeRhIn$_5$, although anisotropic, behaves three-dimensionally below 7 K, and no 3-D to 2-D crossover is found prior to the complete disappearance of intersite magnetic correlations.

To further examine the magnetic state at 7 K, we used the two-axis method to measure magnetic scattering in a wider range in reciprocal space than the scans shown in Fig. 2. Fig. 4(a) presents a quasielastic scan along $q = (h, h, 1)$ for $h$ ranging from 0 to 1. The extra intensity at 7 K derives from fluctuations of magnetic moments which condense into Bragg peaks at 1.4 K. Any multi-phonon contribution to the extra flat intensity should be minimal in view of the low temperatures and small wave numbers. Magnetic intensity at 7 K obtained in various scans with different symmetries is plotted in Fig. 4(b) as a function of $q$. The solid line through the data points is the square of the Ce$^{3+}$ form factor [20]. We notice that (i) polarization dependence is negligible, and (ii) there is little $q$ dependence in magnetic intensity beyond the form factor. This means that at 7 K, the magnetic moments in CeRhIn$_5$ fluctuate independently. The local anisotropic magnetic field, which aligns magnetic moments in the basal plane in the ordered state, has lost its effect on the moments at 7 K. This temperature is also the locus of the peak in the bulk magnetic susceptibility [5], which, thus, is related to development of short-range antiferromagnetic correlations. A similar correspondence between antiferromagnetic short-range order and the susceptibility maximum exists also in the heavy fermion superconductor UPt$_3$ [21], which orders antiferromagnetically below 5 K with a tiny magnetic moment 0.02(1) $\mu_B$ per U [22].

Even though from the point of view of statistical physics, CeRhIn$_5$ is clearly 3-D, at the microscopic level it still seems quite 2-D: the lattice structure is tetragonal with $c/a > 1$, and the anisotropic correlation length together with the pairwise appearance of magnetic Bragg peaks concentrate magnetic spectral weight in reciprocal space in ellipsoids which are elongated in the $c$ direction. While this still differs from the more ideally 2-D situation for the cuprate superconductors, the resemblance may be sufficient that phase-space arguments [4,7], favoring 2-D over 3-D superconductivity when the mediating bosons are antiferromagnetic fluctuations, may still be responsible for the unusually high superconducting $T_C$'s.
of the layered compounds based on CeIn$_3$. One classic way in which to decide the nature of the coupling to the bosons responsible for superconductivity is to examine the electrical resistivity $\rho$ above $T_C$. We have already shown that the T-dependence of $\rho$ at low T is due to scattering from the same magnetic fluctuations which would be needed to produce superconductivity. Should the substantial anisotropy of the fluctuations matter for the superconductivity, it would also appear in the resistance anisotropy above $T_C$. We have therefore measured the electrical resistivity for our samples using the standard 4-probe method, with currents along the $a$ and $c$ axes [see Fig. 3(b)]. The first point to notice is that in contrast to naive expectations and what is seen in the cuprates, $\rho_c$ is somewhat smaller than $\rho_a$ in this temperature range. This does mean, however, that the electrons scatter more, but not much more, strongly from in-plane rather than out-of-plane spin fluctuations. Second, $\rho_c$ and $\rho_a$ have very similar temperature dependences. The proportionality between them may be better seen in the inset to Fig. 3(b) and is very strict down to a temperature slightly above $T_N$. This expands our conclusion from refractory compounds, unless these effects are strongly compound or pressure dependent.

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