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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 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 $3T_C$. In addition, the effect of the magnetic fluctuations on electrical resistivity is well described by the Born approximation.

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The discovery of superconductivity in heavy fermion materials by Steglich *et al.*[1] just over two decades ago generated much of our current thinking on the relation between magnetism and superconductivity.[2] 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 Ruderman-Kittel-Kasuya-Yosida 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,[3] and it is predicted that 2D antiferromagnetic spin fluctuations are superior to 3D fluctuations in elevating the superconducting transition temperatures.[4]

Very recently, a family of heavy fermion compounds with chemical formula $CeMIn_5$ (M=Rh, Ir, Co) has been discovered.[5–7] These materials, with the tetragonal $HoCoGa_5$ structure[8] (space group No. 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 $CeRhIn_5$ above 16 kbar after suppressing the antiferromagnetic order,[5] which is more than half of its Néel temperature at ambient pressure, while the maximum T_C $=0.2$ K for cubic $CeIn_3$ at 25 kbar (Ref. 9) 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.[7] 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 2D antiferromagnetic fluctuations.[5–7,10–12] 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 may not be applicable.

$CeRhIn_5$ ($a=4.652$ Å, $c=7.542$ Å at 295 K) is an incommensurate antiferromagnet below $T_N=3.8$ K.[13,10,5] The small magnetic moments of the Ce ions, $0.37\mu_B$ at 1.4 K which is only a small fraction of paramagnetic moment $2.38\mu_B$, 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 $\mathbf{q}_M$ $=(m/2,n/2,l\pm\delta)$ with m and n odd integers, l integer and $\delta=0.297$.[13] If $CeRhIn_5$ is magnetically 3D, magnetic fluctuations are expected to be enhanced in the vicinity of $\mathbf{q}_M$ near T_N. On the other hand, if $CeRhIn_5$ is 2D, 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,[5] 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, $\mathcal{S}(\mathbf{q})$, 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 40-40-40-40 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 squares measure intensity of magnetic fluctuations at 4 and 7 K, respectively, and the shaded region represents the resolution-limited Bragg peak measured at 1.7 K. Open circles indicate

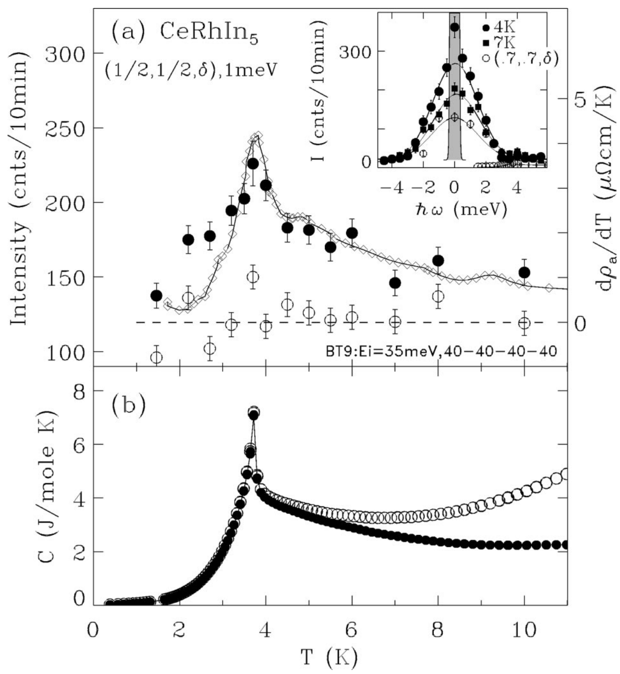

FIG. 1. (a) Intensity of magnetic fluctuations, $\mathcal{S}(\mathbf{q})-\langle M\rangle^2$, at $\mathbf{q}=(1/2,1/2,0.297)$ (solid circles) as a function of temperature. Open circles indicate neutron scattering intensity measured at (0.7,0.7,0.297), which is away from any magnetic Bragg points. Temperature derivative of in-plane resistivity (open diamonds, scale on the right) closely follows magnetic fluctuations. Inset: Constant $\mathbf{q}=(1/2,1/2,0.297)$ scans at 7 K, 4 K (solid symbols), and 1.7 K (shaded region). The constant $\mathbf{q}=(0.7,0.7,0.297)$ scan (open circles) measures the width of incoherent neutron scattering. (b) Total specific heat (open circles) and magnetic specific heat (solid circles) as a function of temperature.

incoherent nuclear scattering measured away from magnetic Bragg points. The width of the incoherent scattering, 1.7 meV HWHM, is the energy window for magnetic fluctuations and is the same as the width of the scans at 4 and 7 K. This implies that the energy scale for magnetic fluctuations, at least up to 7 K, is significantly smaller than 1.7 meV, and therefore a good energy integration of the magnetic excitation spectrum was achieved with our spectrometer configuration. In other words, the quasielastic condition of the two-axis method was realized in our experiments,[14] and the intensity of magnetic neutron scattering measures the *instantaneous* magnetic correlation function $\mathcal{S}(\mathbf{q})$:[15]

$$I\sim|f(\mathbf{q})|^2\sum_{\mu,\nu}(\delta_{\mu\nu}-\hat{\mathrm{q}}_\mu\hat{\mathrm{q}}_\nu)\mathcal{S}^{\mu\nu}(\mathbf{q}), \tag{1}$$

where $f(\mathbf{q})$ is the magnetic form factor for a Ce^{3+} ion, and

$$\mathcal{S}^{\mu\nu}(\mathbf{q})=\hbar\int d\omega\mathcal{S}^{\mu\nu}(\mathbf{q},\omega)=\sum_{\mathbf{R}}e^{i\mathbf{q}\cdot\mathbf{R}}\langle \mathrm{M}_0^\mu(t)\mathrm{M}_{\mathbf{R}}^\nu(t)\rangle. \tag{2}$$

Here $\langle\cdots\rangle$ denotes the thermodynamic average, and $\mathrm{M}_{\mathbf{R}}^\mu(t)$ is the μth Cartesian component of the magnetic moment of the Ce ion at position $\mathbf{R}$ at time t.

Solid circles in Fig. 1(a) are the temperature-dependent intensity, I, of neutrons scattered by magnetic fluctuations. They were measured at the magnetic Bragg point (1/2,1/2,0.297) and with a finite energy transfer of 1 meV. 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, $\mathcal{S}(\mathbf{q_M})-\langle M\rangle^2$, 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 intensity here, which serves as an upper bound of the background, and the strongly T-dependent signal at (1/2,1/2,0.297) reflects the strong spatial modulation of $\mathcal{S}(\mathbf{q})$ near T_N.

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, $l/a\sim10^2$ 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, $\mathcal{S}(\mathbf{q})-\langle M\rangle^2$.[16] Line-connected diamonds in Fig. 1(a) are experimental $d\rho_a/dT$, 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).[5] The similarity among these quantities indicates the prominence of the Fisher-Langer mechanism in $CeRhIn_5$ in the Kondo regime. The Fisher-Langer 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, for which the standard model is the Kondo lattice model.

We now turn to the spatial dependence of magnetic correlations in $CeRhIn_5$. A survey of $\mathcal{S}(\mathbf{q})$ along the c axis and in tetragonal basal plane intersecting Bragg points $(1/2,1/2,\delta)$ and $(1/2,1/2,1+\delta)$ is shown in Fig. 2. The solid circles were measured at 4 K. Strong modulation of $\mathcal{S}(\mathbf{q})$ 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 3D magnetic fluctuations. Fitting the data to an infinite sum of Lorentzians, centered at $(1/2,1/2,l\pm\delta)$, we obtain magnetic correlation lengths at 4 K of $\xi_c=9.5(1)$ Å along the c axis and $\xi_\parallel=23(1)$ Å in the basal plane. While $\xi_\parallel$ is about 5 times the nearest-neighbor distance, a, of Ce ions in the basal plane, ξ_c is only 1.3 times the interplane Ce distance, c. However, this is different from a 2D magnetic system, in which the intraplanar magnetic correlation length is orders of magnitude longer than the interplanar magnetic correlation length.[14]

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,

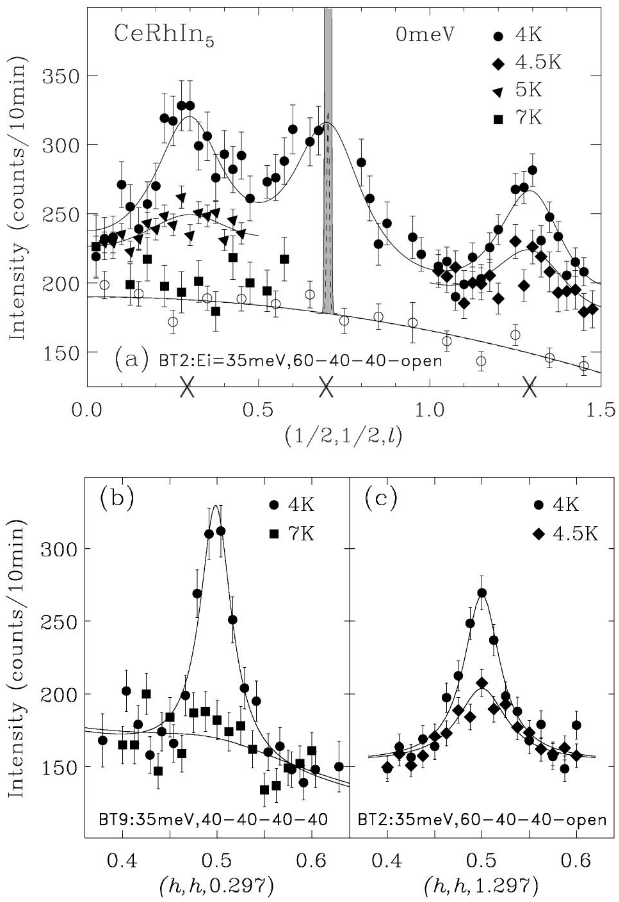

FIG. 2. Magnetic correlation function, $\mathcal{S}(\mathbf{q})$, for (a) $\mathbf{q}=(1/2,1/2,l)$, and for $\mathbf{q}$ along the (110) direction at (b) $l=0.297$ and (c) $l=1.297$ at various temperatures. Open circles in (a) are measured along $(0.4,0.4,l)$. Crosses indicate the magnetic Bragg peak positions below $T_N=3.8$ K. The shaded peak is a magnetic Bragg peak measured at 1.7 K and the dashed line represents its intensity divided by 120.

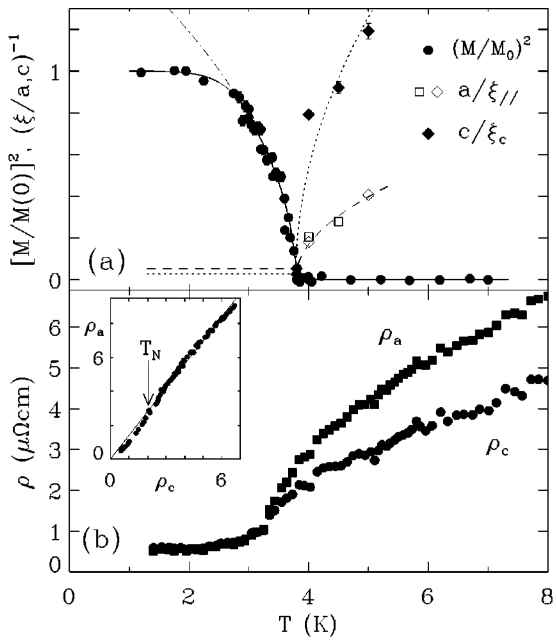

FIG. 3. (a) Solid circles represent the square of the order parameter. The dot-dashed line is $(1-T/T_N)^{0.5}$. Open symbols and solid diamonds are inverse magnetic correlation lengths in the tetragonal basal plane and along the c axis, respectively. The dashed and dotted line are guides to the eyes above T_N, and represent the Bragg peak widths below T_N which indicate the resolution limits. (b) Resistivity, measured with current parallel to the a axis (squares) and the c axis (circles) respectively, as a function of temperature. Inset: ρ_c vs ρ_a with temperature as the implicit variable. The arrow indicates the Néel temperature.

little modulation in $\mathcal{S}(\mathbf{q})$ 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 ξ_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 3D to 2D crossover is found prior to the 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 [refer to the inset to Fig. 4(b)]. Fig. 4(a) presents a quasielastic scan along $\mathbf{q}=(h,h,1)$ 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 multiphonon 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 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. 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 3D, at the microscopic level it still seems quite 2D: 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 2D situation for the cuprate superconductors, the resemblance may be sufficient that phase-space arguments,[4,7] favoring 2D over 3D 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 ρ above T_C. We have already shown

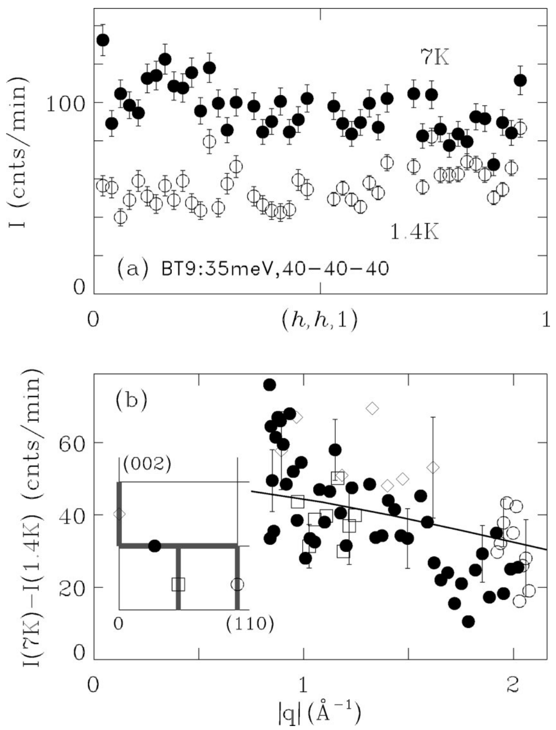

FIG. 4. (a) Quasielastic scan along $(h,\ h,1)$ at 1.4 and 7 K. The extra intensity at 7 K is due to magnetic fluctuations. (b) Magnetic intensity at 7 K as a function of $|\mathbf{q}|$. Solid circles were from the $(h,h,1)$ scan covering $h=0-1$, open diamonds from the $(0,0,l)$ scan with $l=1-2$, open squares from the $(0.5,0.5,l)$ scan and open circles from the $(1,1,l)$ scan with $l=0-1$, as shown by the shaded lines in the inset.

that the T dependence of ρ 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, ρ_c is somewhat smaller than ρ_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, ρ_c and ρ_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 the neutron scattering measurements to assert that as far as the conduction electrons and simple antiferromagnetic mechanisms for superconductivity are concerned, the magnetic correlations in $CeRhIn_5$ are 3D.

In summary, we observe the development of **q** dependent magnetic correlations below 7 K in $CeRhIn_5$. Although anisotropic, these correlations are 3D in nature and have a characteristic energy less than 1.7 meV. Conduction electron scattering in the vicinity of T_N is dominated by these antiferromagnetic fluctuations, and in a manner consistent with weak coupling (the Born approximation is valid) between renormalized, heavy conduction electrons and the f magnetic moments of the Ce ions uncompensated by the Kondo process. The electrical resistivity and magnetic fluctuations at ambient pressure in $CeRhIn_5$ are sufficiently isotropic that it is unlikely that a specifically 2D mechanism involving antiferromagnetic fluctuations is the explanation for the unusual superconductivity of the $CeIn_3$-based layered compounds, unless these effects are strongly compound or pressure dependent.

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