Hidden orbital currents and spin gap in the heavy fermion superconductor URu$_2$Si$_2$

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We have performed neutron scattering experiments on the heavy fermion superconductor URu$_2$Si$_2$ to search for the orbital currents predicted to exist in the ordered phase below $T_N = 17.5$ K. Elastic scans in the (H, K, 0) and (H, 0, L) planes revealed no such order parameter at low temperatures. This does not completely rule out orbital current formation, because our detection limit for a ring of scattering is 0.06(1) $\mu_B$, which is greater than the size of the predicted moment of 0.02 $\mu_B$. However, on heating, a ring of quasielastic scattering does exist in the (H, K, 0) plane centered at the (1, 0, 0) antiferromagnetic Bragg position and of incommensurate radius $\tau = 0.4$ r.l.u.. The intensity of this ring is thermally activated below $T_N$ with a characteristic energy scale of $\Delta = 110$ K: the coherence temperature. We believe that these incommensurate spin fluctuations compete with the AF spin fluctuations, and drive the transition to a disordered magnetic state above $T_N$. The significance of this higher energy scale with respect to $T_N$ suggests that these fluctuations also play a crucial role in the formation of the heavy fermion state.

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We have examined the prediction of Chandra et al. that orbital currents may exist in strongly correlated electron systems. High temperature superconductivity, superfluidity, the quantum Hall Effect, and heavy fermion metals are all examples of the fascinating behavior that arises from complicated interactions within systems of fermions. The interest in the latter example, heavy fermion metals, has been piqued due to the discovery of the coexistence of superconductivity and antiferromagnetism in several species such as UPt$_3$, UPd$_2$Al$_3$, UNi$_2$Al$_3$, and URu$_2$Si$_2$. A striking feature of these compounds is that they display conduction electron specific heats at low temperatures that are orders of magnitude greater than found in typical metals. This behavior differs markedly from the high temperature behavior, at which the value for the Sommerfeld constant is returned to normal free electron values. As evidenced from the resistivity, specific heat, and magnetic susceptibility, these compounds are well described at high temperatures as a set of weakly interacting conduction electrons and local moments. The crossover from this state to a low temperature state, in which the effective mass of the quasiparticles increases dramatically, is a gradual change that is characterized by a coherence temperature $\theta_C$.

The physics of the heavy fermion state is often posed as a competition between the Kondo effect, in which conduction electrons screen out local magnetic moments, and the RKKY interaction, which polarizes the conduction electrons and provides an oscillatory exchange constant as a function of distance between magnetic sites, thus enhancing local moment formation. The magnitudes of both effects increase as a function of the density of quasiparticle states at the Fermi energy $g(E_F)$, but they have different functional forms. At high values of $g(E_F)$, the Kondo interaction is stronger than the RKKY, and thus some heavy fermions do not order at all such as CeAl$_3$. But for low values of $g(E_F)$, there is a tendency for local moment formation, albeit of reduced size (such as UNi$_2$Al$_3$ and UPd$_2$Al$_3$, which have ordered U$^{4+}$ moments of 0.24 $\mu_B$ and 0.85 $\mu_B$ respectively).

In the case of UPt$_3$ and URu$_2$Si$_2$, the ordered moments in the Néel states are extremely small (0.02 $\mu_B$ and 0.03 $\mu_B$ respectively). UPt$_3$ has an exotic superconducting state involving multiple transitions as a function of applied pressure and magnetic fields. The scenario for URu$_2$Si$_2$ is intriguing as well: the transition at $T_N = 17.5$ K is accompanied by a large lambda anomaly in the specific heat, which, in light of the extremely small ordered moment, suggests that another order parameter is at play. There has been considerable interest in URu$_2$Si$_2$ over the last few years, with new developments providing hints that the ordered magnetic state is inhomogeneous, and somewhat parasitic to a so-called “hidden” ordered state. A complex phase diagram for this state has been elucidated based upon specific heat measurements at high magnetic fields. The character of this possible hidden order parameter is still unknown, but possibilities such as quadrupolar ordering and charge-density wave formation are still plausible. It has been shown, however, that none of the allowed quadrupolar or octupolar orderings can ac-
count for the weak moment. Recent NMR measurements have shown that small isotropic magnetic fields develop below \( T_N \) at the silicon sites, which have led Chandra et al. to develop a theory of orbital current formation. The signature for such currents which develop in the ordered phase below \( T_N \) would be a ring of incommensurate scattering in reciprocal space with a characteristic \( Q^{-3} \) form factor.

We have carried out a detailed search for this hidden order parameter in URu_2Si_2 using neutron scattering in the \((H, K, 0)\) and \((H, 0, L)\) planes. No such scattering is found to exist within experimental error. However, we have found an unusual ring of quasielastic scattering at an incommensurate radius from the \((1, 0, 0)\) antiferromagnetic zone center which decreases in spectral weight below \( T_N \). The spectral weight shifts from this ring of scattering, whose characteristic energy scale is the coherence temperature, to the antiferromagnetic spin correlation wavevector and suggests that competing interactions play a role in the ordering at \( T_N \).

Our experiments were performed at the DUALSPEC triple-axis spectrometer at Chalk River Laboratories with pyrolytic graphite as monochromator and analyzer set to a fixed energy of 3.52 THz. A graphite filter was used to remove higher order contamination. Elastic scans were performed, as well as quasielastic scans at an energy transfer of 0.25 THz. The collimation was chosen to be \( 0.40^\circ - 0.48^\circ \) at \( 0.56^\circ \). The crystals were the same two used earlier, one oriented in the \((H, K, 0)\) plane and the other in the \((H, 0, L)\) plane.

Figure 1 shows the \((1, 0, 0)\) magnetic Bragg signal arising from the 0.03 \( \mu_B \) ordered moment. The relative intensity of this peak as compared to background gives a measure of our sensitivity to ordered moments. Considering the signal to background ratio, our calculations indicate that the minimum size of a detectable signal for long-ranged local moment formation is 0.013(1) \( \mu_B \) for 3D order (resulting in a 3D Bragg peak). For a moment distribution which leads to a 2D ring structure with the 0.2 r.l.u. radius of Chandra et al., this limit changes to 0.06(1) \( \mu_B \) (one can calculate this by evaluating the ratios of the areas of a ring structure compared to a magnetic Bragg peak in Q-space, considering our instrumental resolution).

Elastic raster scans were made to look for hidden order with ranges \( 0.5 \leq H \leq 1.0 \) and \( 0 \leq K \leq 1.05 \) for the \((H, K, 0)\) plane and \( 0.175 \leq H \leq 1.075 \) and \( 0 \leq L \leq 1.05 \) for the \((H, 0, L)\) plane. The difference scans, \( I(8 \text{ K}) - I(22 \text{ K}) \), indicate that there are no new sources of magnetic Bragg scattering below \( T_N \). This, together with the results of Bull et al., indicate that no new magnetic Bragg peaks exist in the \((H, H, 0)\), \((H, 0, L)\), and \((H, 0, L)\) planes. However, this does not completely rule out orbital current formation, because our detection limit for a ring of scattering is 0.06(1) \( \mu_B \), which is greater than the size of the moment predicted by Chandra et al. of 0.03 \( \mu_B \).

![Figure 1: The antiferromagnetic Bragg peak at \((1, 0, 0)\) and \( T = 8 \text{ K} \) (fit is to a Gaussian). The inset shows the corresponding magnetic structure for the U^{4+} moments.](image)

Note that for body-centered tetragonal symmetry, the position for the predicted ring of scattering at \((\tau \cos \theta, \tau \sin \theta, 1)\) is equivalent to the wavevectors \((1 + \tau \cos \theta, \tau \sin \theta, 0)\) at which we made the search.

Since no new features were discovered in the elastic channel, we decided to look at the quasielastic spectra at \( \Delta E = 0.25 \) THz. This removes the large incoherent elastic peak and so increases the sensitivity to the formation of slow correlations modulated in Q. Our strategy was to further investigate an incommensurate ring of scattering which was discovered in the previous investigation of the inelastic spectrum near \((1.4, 0, 0)\). A broad feature centered at about 0.6 THz was reported above \( T_N \), indicative of heavily damped antiferromagnetic spin fluctuations. This feature sharpened to a nearly resolution limited peak in energy below \( T_N \) with a center at higher energies (\( \sim 1.1 \) THz). This results in an increase in scattering as one passes above \( T_N \). The structure of this scattering in reciprocal space at this energy transfer was not reported in the original paper, nor was its explicit temperature dependence. Subsequent work suggested that the structure could be a ring in reciprocal space, which is what Chandra et al. predicted for orbital current formation from the hidden order phase.

Figure 2 shows contours of the scattering in the \((H, K, 0)\) plane above and below \( T_N \) at 22 K and 16 K. We have folded the data about the line \( K = 0 \) because of the symmetry of reciprocal space. The scans at \( T = 22 \text{ K} \) show a ring of scattering centered in the \((H, K, 0)\) plane at AF zone centers such as \((1, 0, 0)\) and of radius \( \tau = 0.4 \) r.l.u.. These are not powder lines, which would be centered at \( Q = 0 \), but they are antiferromagnetic spin fluctuations centered about the AF points such as \((1, 0, 0)\) and \((2, 1, 0)\). Note the absence of this scattering about
FIG. 2: (Color online) Contour plot of scattering in the (H, K, 0) plane at T = 22 K and T = 16 K with ∆E = 0.25 THz energy transfer. Note the ring of scattering centered about (1, 0, 0), and the antiferromagnetic fluctuations at (1, 0, 0) associated with the order parameter. The dashed circles are guides to the eye.

(1, 1, 0), a ferromagnetic point. To confirm that these features are magnetic in origin, the form factor has been measured out to higher values of Q. Figure 3 shows the results of these scans, in which points at (0.6, 0, 0), (1.4, 0, 0), (2, 0.6, 0) and (2, 1.4, 0) were measured. The $U^{4+}$ magnetic form factor, plotted on the same curve, is in good agreement with our data. The scattering does not exhibit the predicted Q$^{-4}$ form factor that Chandra et al. predicted for orbital currents, although it does form a magnetic ring in reciprocal space.

The ring of scattering is thermally activated up to the transition at $T_N$ as shown in figure 4. This qualitatively agrees with the previous work of Broholm et al. who measured the inelastic spectra above and below the transition. What we are observing is the tail of an inelastic peak above $T_N$, which moves outside our energy window as the peak narrows and moves to a higher energy scale below $T_N$ (resulting in a suppression of intensity below the transition).

The fit in figure 4 is to a background plus a single activated intensity of the form:

$$I(T) = A \exp(-\Delta/T) \quad T < T_N$$  
(1)

$$I(T) = \text{constant} \quad T > T_N$$  
(2)

where $A$ is a constant, $T_N$ is the Néel temperature (17.5 K), and $\Delta = 110(10)$ K is the fitted activation energy. It is important to note the activation temperature is not that of the sampling energy, 0.25 THz $\sim$ 12 K, nor that of the 0.67 THz $\sim$ 30 K spin excitation above $T_N$. Instead it corresponds to that found by Palstra et al. for the specific heat anomaly below $T_N$. In the range of (2 K - 17.5 K), the specific heat could be fit to the following:

$$C(T) = \gamma T + \beta T^3 + \delta \exp^{-\Delta/T}$$  
(3)

with $\Delta \sim 115$ K. This suggested that a substantial gap opens in the density of states below $T_N$ (which when interpreted as a momentum space phenomenon, gaps $\sim 75$ % of the Fermi surface). This gap is similar to the maximum in the spin-wave density of states, and the gap seen in the optical reflectance measurements. The existence of two competing energy scales ($\Delta$ and $T_N$) has also been observed in DC resistivity experiments.

The quasielastic scattering observed in this experiment is reminiscent of the intimate connection between spin
fluctuations and the heavy fermion state. Gaulin et al. have studied this relationship in UNi$_2$Al$_3$, which has a $T_N$ of 4.6 K and $T_c$ of 1.2 K. The magnetic structure is incommensurate, with an ordering wavevector of $Q = (0.5 \pm \tau, 0, 0.5)$ ($\tau = 0.11$) and an ordered moment of 0.85 $\mu_B$ per U atom. The quasielastic spin fluctuations are of two kinds: those associated with the incommensurate wavevector, and with the commensurate $Q = (0, 0, 0.5)$ wavevector. The two modes compete with one another, with a shift in spectral weight from the commensurate to incommensurate fluctuations below $T_N$. Above the transition, the incommensurate fluctuations disappear, but the commensurate ones persist to nearly 80 K, the coherence temperature. This is strong evidence that these excitations are associated with the formation of the heavy fermion state. It is remarkable that both URu$_2$Si$_2$ and UNi$_2$Al$_3$ show the competition between two excitations associated with different energy scales ($T_N$ and $\Delta$). In the case of UNi$_2$Al$_3$, the commensurate fluctuations were noted to be at the same ordering wavevector as in the sister compound UPd$_2$Al$_3$, which orders at $T_N = 14.5$ K. A competition between the RKKY interaction and the Kondo effect is believed to explain the difference in the magnetic structures and spin excitation spectra of UNi$_2$Al$_3$ and UPd$_2$Al$_3$.

For URu$_2$Si$_2$, the situation is reversed: the commensurate fluctuations are associated with the ordering wavevector $(1, 0, 0)$, and the incommensurate excitations persist to high temperatures (and therefore can be identified with the formation of the heavy fermion state). This shift in spectral weight can be noted by the increase in scattering at $(1, 0, 0)$ below $T_N$ (see figure 2) and the corresponding decrease in intensity at $(1.4, 0, 0)$. Mason et al. have suggested that magnetic frustration plays a role in the unusual magnetic properties of URu$_2$Si$_2$, with the long range oscillatory nature of the RKKY interaction providing the mechanism. The incommensurate ring of scattering we observe may be a signature of such a RKKY interaction. However, the true nature of these fluctuations cannot be ascertained with this experiment alone. The fact that the activation energy is the same for the spin response and the specific heat anomaly strongly suggests that these are excitations out of the gapped "hidden order" state rather than the AF ordered phase. Further experiments are needed to develop a clearer picture of the origin of these excitations with respect to proposed scenarios of a phase separation occurring at $T_N$. It may be that this feature is linked electronic phase separation, as suggested by $\mu$SR and NMR measurements, since the spectral weight is too large to be explained by the 0.03 $\mu_B$ ordered moment.

In conclusion, our neutron scattering measurements in search of hidden order in the $(H, K, 0)$ and $(H, 0, L)$ planes have placed an upper limit of 0.013(1) $\mu_B$ for the presence of any long-ranged 3D ordered spin structure well-defined in $Q$. For a ring of scattering, the detectable moment is 0.06(1) $\mu_B$, which precludes orbital current formation that is somewhat larger than that predicted by Chandra et al. Quasielastic scattering experiments have revealed a connection between a ring of incommensurate scattering at a radius of $\tau = 0.4$ from the zone center and the heavy-fermion state. The exponential activation energy of this ring below $T_N$, comparable with the specific heat activation energy, suggests that a gap of 110 K is a feature of the "hidden order" phase.

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