Gapped itinerant spin excitations account for missing entropy in the hidden order state of URu$_2$Si$_2$

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One of the primary goals of modern condensed matter physics is to elucidate the nature of the ground state in various electronic systems. Many correlated electron materials, such as high temperature superconductors,1,2
gapently frustrated oxides,3,4 and low-dimensional magnets3,4 are still the objects of fruitful study because of the unique properties which arise due to poorly understood many-body effects. Heavy fermion metals5 - materials which have high effective electronic mass due to these effects - represent a class of materials with exotic properties, such as unusual magnetism, unconventional superconductivity, and “hidden order” parameters.6 The heavy fermion superconductor URu$_2$Si$_2$ has held the attention of physicists for the last two decades due to the presence of a “hidden order” phase below 17.5 K. Neutron scattering measurements indicate that the ordered moment is $0.03\, \mu_B$, much too small to account for the large heat capacity anomaly at 17.5 K. We present recent neutron scattering experiments which unveil a new piece of this puzzle - the spin excitation spectrum above 17.5 K exhibits well-correlated, itinerant-like spin excitations up to at least 10 meV emanating from incommensurate wavevectors. The gapping of these excitations corresponds to a large entropy release and explains the reduction in the electronic specific heat through the transition.

The central issue in URu$_2$Si$_2$ concerns the identification of the order parameter which explains the reduction in $\gamma$, and thus the change in entropy, through the transition at 17.5 K.6 Numerous speculations about the ground state have been advanced, from quadrupolar ordering,7 to spin-density wave formation,8 to “orbital currents” to account for the missing entropy.9 In this letter we present cold neutron time-of-flight spectroscopy results which shed some light on the “hidden order” in URu$_2$Si$_2$. We have performed experiments above and below the ordering temperature to measure how the spin excitations evolve. It is clear from our data that above $T_0$ the spectrum is dominated by fast, itinerant-like spin excitations emanating from incommensurate wavevectors at positions located 0.4a* about the antiferromagnetic (AF) points. From the group velocity and temperature dependence of these modes, we surmise that these are heavy quasiparticle excitations which form below the “coherence temperature” and play a crucial role in the formation of the heavy fermion and “hidden order” state. The gapping of these excitations, which corresponds to a loss of accessible states, accounts for the reduction in $\gamma$ through the transition at 17.5 K.

Figure 1 shows the excitation spectrum of URu$_2$Si$_2$ at 1.5 K in the H00 plane. The characteristic gaps at $\sim 2$ meV at the AF zone center (1,0,0) and $\sim 4$ meV at the incommensurate wavevectors (0.6,0,0) and (1.4,0,0) have been known for some time.10 The incommensurate wavevector corresponds to a displacement of $\sim 0.4\, a^*$ from the AF zone centers (i.e. where $h+k+l=an\, an\, an$ odd integer, and thus forbidden in the BCC chemical structure). A scenario for this mode-softening at the incommensurate position was previously described with a model based upon oscillatory exchange constants between near neighbors (not uncommon for RKKY type interactions).10,11

Figure 2 exhibits our new neutron results at 20 K in the same H00 plane. Above the phase transition, the sharp spin waves evolve into weak quasielastic spin fluctuations at the zone center (1,0,0) and strong excitations at the incommensurate positions (1±0.4,0,0). We have considered the possibility that these incommensurate excitations may be due to magnetovibrational scattering. This can arise from a shifting of a the phonon excitations at (2,0,0) to (1±0.4,0,0) as allowed through the neutron scattering cross-section for magnetoelastic coupling.12 However, with the small moment size of this system, it is improbable that such a scattering process is being observed. It was also originally reported in that the incommensurate excitations were just quasielastic fluctuations, as constant Q scans on a triple axis spectrometer resolved a peak at a finite energy of 0.6 THz $= 2.5$ meV and a decrease in intensity as a function of energy typical of an overdamped response. What was previously unknown was that the quasielastic fluctuations were only the lower limit of a band of high-velocity spin excitations that ex-
tend well above the upper limit of the sharp collective spin excitations of the ordered phase. As the temperature is increased, the (1,0,0) fluctuations decrease and the incommensurate fluctuations remain approximately constant to at least 25 K.\[13\]

The implications of this discovery are: (1) The incommensurate excitations have a well-defined structure as a function of Q, and thus the electrons are highly correlated above 17.5 K. This is completely unexpected for a system of localized moments in a paramagnetic state, but similar dynamics above $T_N$ (with the formation of paramagnons[14] for example) has been observed with neutron scattering experiments in other itinerant electron systems such as chromium and V$_2$O$_3$.\[13\], \[15\] (2)

The dispersion is such that the maximum group velocity is at least a factor of 2 larger than the maximum group velocity of the excitations in the ordered state. A significant restructuring of the Fermi surface must be responsible for the hidden order state. (3) The gapping of these strong spin fluctuations below 17.5 K must provide a considerable portion of the entropy removal at the transition.

These excitations have a structure in reciprocal space such that they occur at several symmetry-related wavevectors within the first Brillouin zone. The amount of phase space occupied by these excitations is greater than those at the (1,0,0) AF zone center. Figure 3 shows constant energy cuts of the excitations in the HOL plane to emphasize this. Note the weak AF fluctuations at (1,0,0) and (2,0,1) in comparison to the excitations at the incommensurate positions. Also note how a cone of scattering develops about the incommensurate positions, while the AF zone center simply decreases in intensity as energy transfer is increased. For comparison, a cut is also shown of the spin-wave spectrum at 1.5 K in the same energy window. The results at 20 K suggest a continuum of excitations within a cone of scattering (as expected for low-lying excitations at the Fermi surface), as opposed to the sharp excitations which are gapped below 17.5 K.

An estimate of the contribution to the electronic specific heat term from the removal of these low-energy spin fluctuations can be made through an analysis of the spectrum at 20 K. The specific heat of a cone of fast excitations within a constant energy window. The results at 20 K suggest a continuum of excitations within a cone of scattering (as expected for low-lying excitations at the Fermi surface), as opposed to the sharp excitations which are gapped below 17.5 K.

A characteristic correlation length and spin wave velocity need to be extracted from our data to complete this calculation. Our data has been fit to a standard formula for paramagnetic scattering from a system of correlated spins, also known as the Chou model.\[18\] This has been used to extract correlation lengths and spin wave velocities for excitations in the superconducting cuprates, for example.\[19\], \[20\], \[21\] With this model, $S(Q, \omega)$ is represented as

$$S(Q, \omega) = \frac{\hbar\omega}{1 - \frac{\hbar\omega}{\kappa^2 + q^2}} \frac{A}{(\hbar\omega \pm \hbar\omega_q)^2 + \Gamma^2}$$

where $\overrightarrow{Q} = \overrightarrow{Q} - \overrightarrow{\delta}$ ($\delta$ is the incommensurate wavevector), $\kappa$ is the inverse of the correlation length $\xi$, and the frequencies of the damped spin waves are $\omega_q = cq$, where $c$ is the spin wave velocity ($\Gamma = \kappa c$). Fitting with these parameters yields a correlation length of $\xi = 14(2)$ Angstroms, and $c = 45(10)$ meV Angstroms (see figure 2). This value is similar to the Fermi velocity, reported as $\sim 8.84 \times 10^3$ m/s (35 meV Angstroms)\[22\], illustrating the itinerant nature of the excitations. It is interesting to note that there is a curious intersection of cuprate and heavy fermion physics with this analysis - the fast excitations seen in the superconductor La$_{1.86}$Sr$_{0.14}$CuO$_4$ (LSCO), which have an effective mass near an electron mass, appear similar to the excitations in URu$_2$Si$_2$, albeit with a higher velocity because of their lighter effective mass.\[19\]

The contribution of the spin excitations to the specific heat of with these parameters for URu$_2$Si$_2$ is 220 +/- 70 mJ/(mol K$^2$) at 20 K. A comparison of this to the data, in Figure 5, shows a good agreement with a $\gamma$ of 155(5) mJ/(mol K$^2$). The gapping of these excitations leads

$$C_v = \frac{v_a \xi^{-2}}{3\pi^2 c} \times k_B^2 T$$

where $c$ is the characteristic spin wave velocity. Previous work has shown that the excitations lie within the HK0 plane around (0,6, 0, 0).\[16\] Note that due to the dispersion of the spin excitations, the contribution to the heat capacity is linear in temperature, which is exactly the same power law dependence for the electronic specific heat. This term arises due to these fast itinerant spin excitations in URu$_2$Si$_2$ and is the reason for the enhanced linear component to the specific heat above $T_0$ = 17.5 K. This calculation of a gap opening in the spin excitation spectrum is equivalent to a calculation of the gap opening in the Fermi surface. This is demonstrated by the similarity of the matrix-element-weighted density of spin-wave states and infrared spectroscopy measurements, which measure charge excitations.\[17\], \[11\] The spin and charge degrees of freedom are strongly coupled.

$$\frac{\partial C}{\partial T} = \frac{\hbar\omega}{1 - \frac{\hbar\omega}{\kappa^2 + q^2}} \frac{A}{(\hbar\omega \pm \hbar\omega_q)^2 + \Gamma^2}$$

where $\overrightarrow{Q} = \overrightarrow{Q} - \overrightarrow{\delta}$ ($\delta$ is the incommensurate wavevector), $\kappa$ is the inverse of the correlation length $\xi$, and the frequencies of the damped spin waves are $\omega_q = cq$, where $c$ is the spin wave velocity ($\Gamma = \kappa c$). Fitting with these parameters yields a correlation length of $\xi = 14(2)$ Angstroms, and $c = 45(10)$ meV Angstroms (see figure 2). This value is similar to the Fermi velocity, reported as $\sim 8.84 \times 10^3$ m/s (35 meV Angstroms)\[22\], illustrating the itinerant nature of the excitations. It is interesting to note that there is a curious intersection of cuprate and heavy fermion physics with this analysis - the fast excitations seen in the superconductor La$_{1.86}$Sr$_{0.14}$CuO$_4$ (LSCO), which have an effective mass near an electron mass, appear similar to the excitations in URu$_2$Si$_2$, albeit with a higher velocity because of their lighter effective mass.\[19\]

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to a dramatic reduction in $\gamma$ to 38(2) mJ/(mol K$^2$). This is naturally explained through the gap in the spin excitation spectrum, which has minima at 2 meV and 4 meV. The average thermal energy below 17.5 K is less than 2 meV, and thus most of the spin excitations are inaccessible. This is the reason for the reduction of $\gamma$ through the phase transition. The origin of the size of the specific heat jump at $T_0$ is still unresolved at this point although its low temperature limit arises from the removal of the spin contribution to leave a weak remanent ungapped electronic band. A gap opens in the spin excitation spectrum with a value of $\sim 110(10)$ K, but the identity of this ordered state is still a mystery. Through these measurements, we do now that the nature of the transition at $T_0$ seems to be dominated by itinerant electron physics rather than localized crystal field effects. This is the reason why URu$_2$Si$_2$ shows a transition between itinerant and localized behavior at $T_0$, as shown through thermal conductivity measurements. Indeed, we have seen no evidence of crystal field excitations up to at least 10 meV $\sim 110$ K. The complicated crystal field schemes which have been used to explain the heat capacity anomaly at 100 K, the hidden order state, and the anomalous thermal expansion at 60 K need to be re-examined within this framework. Our measurements show that the first excited state, if allowed by selection rules, must exist above 200 K.

The role that these excitations play with the formation of the heavy fermion state can be examined through their temperature dependence. Figure 5 shows recent heat capacity measurements, in comparison with inelastic neutron scattering scans at 100 K. Note that the incommensurate features have disappeared at this temperature, often called the "coherence temperature," which marks a cross-over from paramagnetic weakly correlated moments to correlated heavy electron behavior. They are intimately correlated with the coherence temperature and the formation of the heavy fermion state. This energy scale, as well, describes the temperature dependence of the heat capacity anomaly at 17.5 K through fits to an activation law, and the temperature dependence of the incommensurate excitations below 17.5 K. This is further evidence that the gapping of these excitations is directly related to the formation of the "hidden order" state.

In conclusion, our recent neutron scattering results in URu$_2$Si$_2$ unambiguously demonstrate that itinerant-style excitations exist above $T_0$ at incommensurate wavevectors $\sim 0.4$ a* from the antiferromagnetic zone center. The gapping of these excitations accounts for the change in $\gamma$, the electronic contribution of the specific heat, through $T_0$. The hidden order transition appears to be a rearrangement of electrons at the Fermi surface in an itinerant rather than localized electron picture. Excitations out of this state at these incommensurate wavevectors show a mode softening which can perhaps be reminiscent of another dynamic ground state which has no translational order but a prominent specific heat anomaly— the superfluid liquid helium transition. Correlations between the heavy quasiparticles in URu$_2$Si$_2$ build up below 100 K, and at 17.5 K, there is a transition to a new condensate which still remains a mystery. Even though cause of the change in the electronic specific heat has been identified, the true order parameter for this system has yet to be unveiled.

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**COMPETING FINANCIAL INTERESTS**

The authors declare that they have no competing financial interests.

**EXPERIMENTAL METHODS**

Our measurements were made with a single crystal grown at McMaster University grown by the Czochralski method using elemental U, Ru, and Si, followed by annealing in argon at 900 degrees C. The magnetic properties were confirmed through DC magnetometry measurements on a Quantum Design SQUID, and heat capacity and resistivity measurements on a Quantum Design PPMS. The neutron scattering measurements were performed at the Disk Chopper Spectrometer (DCS) at the NIST Center for Neutron Research in Gaithersburg, Maryland. The 11.5 g URu$_2$Si$_2$ crystal was aligned in the H0L plane and placed in a standard ILL cryostat and measurements were taken using cold neutrons of 2.5 Å and 5 Å. The identification of the equipment is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the equipment is necessarily the best available for the purpose.

Scattering experiments were completed by C. R. W., J. A. J., G. J. M., H. D. Z., Y. Q., J. R. D. C., Z. Y., and W. J. L. B. The crystals were grown by J. D. G. and G. M. L. Specific heat measurements were made by
Data analysis and writing of the paper was completed by C. R. W., J. A. J., G. J. M., G. M. L., L. B., and W. J. L. B.

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FIG. 1: Inelastic neutron scattering of URu$_2$Si$_2$ in the H00 plane at $T = 1.5$ K (where H and L are reciprocal lattice vectors $a^*$ and $c^*$, L is integrated from -0.12 to 0.12). Note the minima at the AF zone center (100) and incommensurate positions $(1+/-0.4,0,0)$. The feature at (200) is due to phonons. The inset shows how the incommensurate excitations become gapped through the transition by counting at the point $(0.6,0,0)$ at 0.25 meV transfer on a triple axis spectrometer. /cite(Wiebe)
FIG. 2: Itinerant spin excitations in URu$_2$Si$_2$ (a) Inelastic neutron scattering of URu$_2$Si$_2$ in the H00 plane at $T = 20$ K (L integrated from -0.12 to 0.12). Note the cone of ungapped excitations emanating from the incommensurate wavevectors $(1 \pm 0.4, 0, 0)$. (b) Fits to the Chou model, with parameters described in the text.
FIG. 3: Cuts along $\Delta E$ of scattering in the H0L plane at (a) 1.5 K and (b), (c), (d) 20 K. The rings about (200) correspond to phonons. Note the spin waves in (a) as rings centered on $(H+K+L)=\text{odd integers (AF zone center)}$ and rings centered about incommensurate points separated by 0.4 $a^*$ from the AF zone centers. At 20 K, there are weak overdamped AF fluctuations at the zone centers, but it is clear that a cone of highly correlated excitations develops as a function of energy. The energy of integration is denoted in meV for each plot.
FIG. 4: Reciprocal space map of the commensurate and incommensurate scattering in URu$_2$Si$_2$ (blue dots: incommensurate positions, red dots: antiferromagnetic lattice points, blue dots: nuclear lattice points)
FIG. 5: Cuts in the H0L plane integrated from 5-8 meV at (a) 20 K and (b) 100 K. Note how the incommensurate scattering at positions such as (1.6,0,1) disappears at 100 K (indicated by a circle). This is just above the “coherence” temperature where a signature of heavy quasiparticle formation is seen in the specific heat (inset). (c) The specific heat/temperature as a function of $T^2$, showing the anomaly at 17.5 K. The linear portion of the specific heat, $\gamma$, is calculated with the solid line to be 155(5) mJ/(mol K$^2$). The blue data point is the calculation of our $\gamma = 220(70)$ mJ/(mol K$^2$) from the spin fluctuations observed at 20 K. The errors bars have been calculated from equation (2) in the text.