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FAST TRACK COMMUNICATION

Itinerant spin excitations near the hidden order transition in URu₂Si₂

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Abstract

By means of neutron scattering we show that the high temperature precursor to the hidden order state of the heavy fermion superconductor URu₂Si₂ exhibits heavily damped incommensurate paramagnons whose strong energy dispersion is very similar to that of the long-lived longitudinal f spin excitations that appear below T_0 . This suggests that there is a strongly hybridized character to the itinerant excitations observed previously above the hidden order transition. Here we present evidence that the itinerant excitations, like those in chromium, are due to Fermi surface nesting of hole and electron pockets; hence the hidden order phase probably originates from a Fermi surface instability. We identify wavevectors that span nested regions of a f-d hybridized band calculation and that match the neutron spin crossover from incommensurate to commensurate on approach to the hidden order phase.

(Some figures in this article are in colour only in the electronic version)

The heavy fermion superconductor URu₂Si₂ has puzzled physicists for more than two decades [1–5] yet, despite recent progress [6], theoretical and experimental work is still required to unravel its mysteries. The large change in entropy across the lambda-like anomaly at $T_0 = 17.5$ K in URu₂Si₂ denotes a phase transition has taken place, but unlike other second order

transitions, we can find no large order parameter to accompany that change. Neutron scattering measurements of the staggered magnetic moment [4, 5, 7], show it is too small to explain the specific heat jump and hidden order, but incommensurate spin density waves have been detected that might explain the entropy loss [6]. Experiment finds that spin excitations are strong and longitudinal and belong to the bulk hidden order phase [5]. Theoretical perspectives on URu₂Si₂ generally

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fall into either localized or itinerant scenarios. In the first, f electrons on the uranium sites must have a quadrupolar or even octupolar character [8, 9]. This scenario is not supported by the absence of observable crystal field excitations [4–6]. In the second, a Fermi surface instability leads to a restructuring and the subsequent change in entropy [3, 10, 11].

Our recent work on URu_2Si_2 [6] revealed that spin fluctuations emanate from incommensurate wavevectors with a relatively large velocity, that they can account for the large specific heat, and can explain its reduction below T_0 by the gapping of the spin excitations at all wavevectors. In an earlier search for orbital currents, as rings of magnetic incommensurate scattering in reciprocal space with a Q^{-4} form factor [2], Wiebe *et al* [12] found that any such ring scattering was undetectable. Instead, they found enhanced thermally activated scattering at the $(0.6, 0, 0)$ position. From our high resolution NIST work [6] on the Disk Chopper Spectrometer (DCS) instrument above and below the T_0 transition, we found dynamic incommensurate scattering around the forbidden $(1 \pm \delta, 0, 0)$ position with $\delta = 0.4$ at $T = 20$ K and concluded the following: (1) these highly correlated and gapless excitations have a well defined Q-structure and their incommensurability with the lattice suggests itinerant electronic behavior as opposed to localized electron physics¹¹, (2) a restructuring of the Fermi surface must be responsible for the hidden order transition, and (3) the gapping of the incommensurate excitations below T_0 in the hidden order state removes thermally accessible excitations and accounts for the missing entropy. The entropy was calculated by fitting the over-damped modes to a spin density wave model [13, 14], extracting a correlation length, and using a linear DOS counting argument. Despite not knowing precisely what the hidden ordered state is, we believe that [6] identified the collective modes associated with the hidden order. Broholm *et al* [5] used a model of two singlets coupled by seven exchange constants to explain the excitation spectrum. We will demonstrate in this letter how nesting of the Fermi surface leads to the incommensurate wavevector of the over-damped collective modes. Our findings dramatically simplify the understanding of the excitations in the system.

High purity U, Ru, and Si were melted in a mono-arc furnace into URu_2Si_2 buttons. Three large single crystals, 20 g in total, were then grown via a modified czochralski method in a titanium gettered argon atmosphere tri-arc furnace followed by annealing in argon at 900 °C. Three crystals were co-aligned in the (HOL) scattering plane, two grown at Los Alamos National Laboratory and one at McMaster University. Each sample was confirmed to be a single crystal in a 2π survey at the E3 spectrometer at Chalk River. We performed extensive inelastic neutron scattering (INS) studies on URu_2Si_2 near the hidden order transition temperature. In the first experiment the setup was the same as in [6]. In the second experiment with the C5 Triple Axis Spectrometer at the NRU reactor in Chalk River, Canada, we followed the excitations to larger energies. The instrument setup and

¹¹ Severe broadening of the crystal field excitations can occur for strong f - d hybridizations, but the dominant excitations in our system seem to have a strong itinerant character.

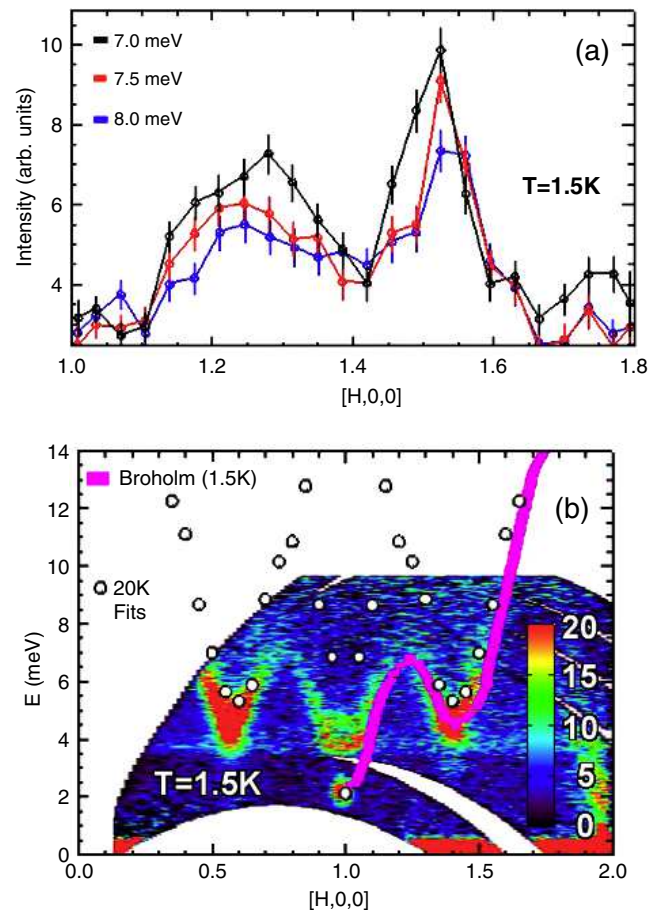


Figure 1. High resolution DCS data (b) integrated in the L direction from $[-0.15$ to $0.15]$, at $T = 1.5$ K deep in the hidden ordered state. We can see the excitations at the commensurate (100) position and the modes previously thought of as a ‘mode softening’ centered on the incommensurate positions $(1 \pm 0.4, 0, 0)$. Overlaid in magenta is the 1.5 K dispersion relation from the model of Broholm *et al* [5] from their triple axis work. In panel (a), one can see the excitations extend to higher energy than previously suggested. The cuts are energy integrated from $[6.5, 7.5]$, $[7, 8]$, $[7.5, 8.5]$ meV (with the same L integration as (b)) in arbitrary units for the black, red, and blue, traces respectively. In (b) we overlay on the 1.5 K data the resonant paramagnon energy at 20 K from fits to the damped simple harmonic oscillator spectrum to the thermal Chalk River data.

collimation were set to 0.5° -PG- 0.80° -S- 0.85° -PG- 2.4° with final scattering energy $E_f = 14.6$ meV. Two graphite filters in the scattered beam eliminated higher-order feed through. Constant-Q scans were performed up to 20 meV at wavevector transfers $Q = (1 + h, 0, 0)$ for $h = 0-1$ and compared to measurements at $(3-h, 0, 0)$ to detect the presence of phonons. For $H < 1.65$ the scattering is entirely magnetic. Constant energy scans up to 16.5 meV were performed to determine the momentum width of the cones of incommensurate scattering.

In figure 1(b), we reproduce our high resolution NIST data at $T = 1.5$ K, well into the hidden order state (the Q-independent spurion at 3.9 meV is due to fast fission processes in uranium). We can see the excitations above the commensurate (100) position and the modes previously thought of as a ‘mode softening’ centered on the incommensurate position $(1 \pm 0.4, 0, 0)$. Overlaid in figure 1(b)

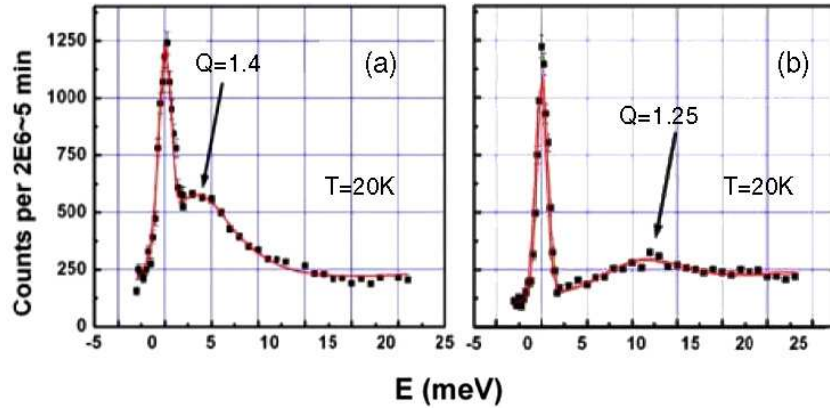


Figure 2. The triple axis constant-Q scans with final energy 14.6 meV at $T = 20$ K for $\mathbf{Q} = (H, 0, 0)$ in (a) and (b) show how the energy of the damped paramagnon resonance disperses with wavevectors. The lines are a damped simple harmonic oscillator fit whose resonant energy is plotted in figure 1(b). Note the presence of damped ungapged spin fluctuations existing up to high energies.

in magenta is the 1.5 K dispersion measured by Broholm *et al* earlier [5]. The subtraction routine used to remove the spurion [6] also removes intensity above $H = 1.3$ which is clearly visible figure 1(b). One can see in figure 1(a) 1 meV wide constant energy cuts along $[H00]$. These cuts near the local maxima of the Broholm data show increased scattering above the single peak of their model. Thus the excitations extend higher in energy than previously thought. The broad tails to higher energy suggest that the sharp spin peak is the onset of a continuum, rather than a long-lived spin wave.

In figures 2(a) and (b) we present examples of constant-Q scans with thermal neutrons at NRU above the hidden order phase at $T = 20$ K. The broad paramagnetic scattering shows a peak whose energy rises to a maximum between minima at $H = 1.0$ and $H = 1.4$, the wave vectors whose susceptibility is largest. We fit the inelastic spectrum to a damped simple harmonic oscillator model and to an elastic Gaussian plus a sloping background. The inelastic spectrum is given by

$$S(\omega, Q) = (n(\omega) + 1) \frac{A\omega\omega_c\Gamma}{(\omega^2 - \omega_c^2)^2 + \omega^2\Gamma^2} \quad (1)$$

where $n(\omega) = (\exp(\frac{\hbar\omega}{k_B T}) - 1)^{-1}$ is the Bose factor, Γ is the damping, and ω_c is the resonant energy. We found that a Q-independent damping rate of $\Gamma = 10 \pm 1$ meV best describes the data. In figure 1(b) the resonant energy of the paramagnetic scattering is plotted as open circles and is folded about (100). The paramagnon dispersion at 20 K is found to be surprisingly similar to that of the long-lived longitudinal spin excitations of the hidden order phase at 1.4 K. The main difference is that the paramagnons are heavily damped. However, there remain energy minima at $H = 1.0$ and the incommensurate 1.4 position that generate two maxima in the susceptibility $A/\omega_c(Q)$. The existence of a precursor phase of strongly damped excitations is reminiscent of other itinerant electron systems near magnetic ordering transitions such as chromium and MnSi and can be represented as a general result of self-consistent renormalization theory [15]. In addition to the broad spectrum of damped paramagnons we also found that a commensurate central mode appeared for $\mathbf{Q} \rightarrow (1, 0, 0)$

in scans such as figure 2 consistent with the commensurate precursor at 20 K [5, 16]. This indicates that slow fluctuations of the hidden order phase take place on approach to T_0 .

We now present a new interpretation of the spin wave spectrum above and below T_0 in terms of nesting at the Fermi surface. The Fermi surface of URu₂Si₂ from recent band structure calculations [17] is reproduced in figure 4. We see that the incommensurate wavevectors of $0.6a^*$ and $1.4a^*$ can come from the nesting of the electron jack at Γ with the two hole pockets at Z. We can eliminate several other possibilities based upon the topology of the Fermi surface (which must have nearly parallel sheets for nesting), and also with the knowledge that the excitations are along the a^* and b^* directions. While wavevector matching does not prove nesting (the velocity at Z is larger than at Γ) it is at least suggestive that the incommensurate excitation mode is represented by an electron ‘jack’—hole pocket excitation above T_0 . Below T_0 , the electron ellipse nesting vectors $X-X'$ may lead to the commensurate excitation at the (100) position. The commensurate X to X' magnetic transition involves an X-point ARPES density that grows rapidly on cooling (see [17, figure 18]) and might explain the crossover from incommensurate to commensurate susceptibility [16] on cooling below 23 K. The ARPES data [17] was measured at high temperatures ($T = 200$ K). To confirm our nesting hypothesis requires low-temperature measurements. There is not a perfect nesting condition for the electron pockets, but this may be an artifact of the calculation, and the pockets could actually be more symmetric in shape. However, the matching of these wavevectors is quite good and given the arguments for itinerant magnetism stated above, it is reasonable to assume that nesting plays an important role in this transition. A recent theory paper has also identified the role of these Fermi surface ‘hot spots’ in the formation of the hidden order state [19]. What is unclear is why the nesting wave vectors associated with $\mathbf{Q} = (1, 0, 0)$ dominate in the hidden order state. We speculate that on cooling the f weight may move to a lower hybrid band with commensurate nesting, whereas above the hidden order phase the valence fluctuations of the thermally broadened upper hybrid bands are nested incommensurately. This view is supported by the recent observation in ARPES at 13 K of

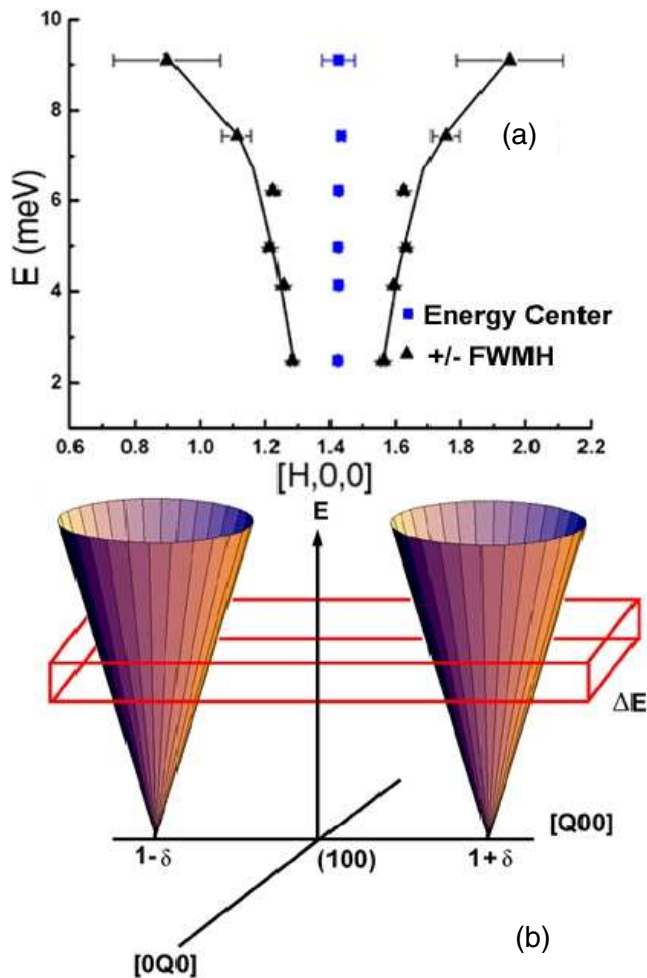


Figure 3. In the upper panel (a) from constant energy scans we show the spreading with energy of the width in wavevector of the incommensurate dynamic spin density wave at $T = 20$ K. The lower panel (b) shows a cartoon of the incommensurate fluctuations in the H-K plane.

an f band that drops below E_F [23]. A recent experimental paper suggests that the existence of commensurate (1, 0, 0) excitations is a requirement for the formation of Cooper pairs at low temperatures [20, 22].

Our data for URu_2Si_2 shows some similarities to the itinerant system chromium in that a SDW forms at a nesting wavevector [18]. In URu_2Si_2 , however, the incommensurate fluctuations are dynamic as opposed to the static SDW of chromium. Only under pressure does a static spin order of $0.3 \mu_B$ occur, and then only at the commensurate (100) wavevector [21]. In figure 3(a) we show growth with energy of the FWHH in wavevector of the incommensurate filled cones of scattering at $T = 20$ K and (b) a cartoon depicting how the filled cones evolve with energy. The cones of scattering observed in URu_2Si_2 are very similar to the fast itinerant excitations in Cr apart from the fact that the incommensurate scattering in URu_2Si_2 forms a solid cone [6].

While a full explanation of the URu_2Si_2 hidden order state still remains to be found, we have laid out a consistent explanation for how the excitation spectrum relates to the Fermi surface of itinerant electrons. We find that the

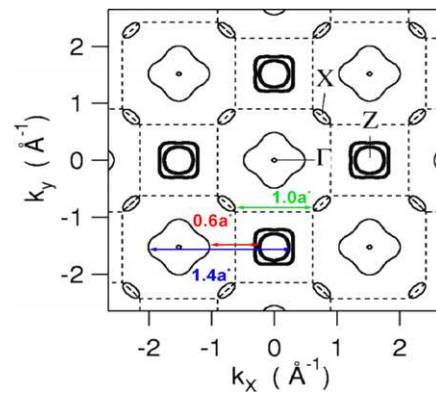


Figure 4. The calculated Fermi surface of URu_2Si_2 [17] and possible nesting vectors which may produce the commensurate and incommensurate itinerant excitations.

paramagnon dispersion above T_0 follows the spin wave dispersion for $T \leq T_0$, so it follows that the exchange interaction between uranium moments does not change across T_0 . Varma and Zhu [11] claim that with helical or Pomeranchuk order, there will be a gap opening with a reduction in the low energy transverse spectral weight. Although they are non-specific about the nature of the change in spectrum, our experiment shows that the response arises only from longitudinal fluctuations. The main characteristic of the phase transition is that the electronic damping of longitudinal fluctuations decreases dramatically across T_0 .

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