Incommensurate charge modulation in the hidden-order analogue UPt$_2$Si$_2$

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In 5f-electron systems, interactions with comparable strength compete due to the extended nature of the wavefunction leading to rich variety of exotic ground states$^1$. URu$_2$Si$_2$, one of the most researched materials in uranium intermetallic family shows heat capacity anomaly at $T_{HO} = 17.5$ K, a precursor to an unconventional superconductivity at $T_c = 1.5$ K$^2$. The nature of the “hidden order” parameter responsible for the anomaly is still debated, more than 30 years after its discovery. In the absence of an obvious magnetic order, a strong and coherent magnetic excitation is observed around two distinct wavevectors$^3$, the antiferromagnetic wavevector, $Q_0 = (1 0 0)$, and the incommensurate wavevector, $Q_1 = (0.4 0 0)$$^4$. UPt$_2$Si$_2$, on
the other hand, order antiferromagnetically with $Q_{\text{AFM}} = (1 0 0)$ at $T_N = 35$ K$^5$ and has long been considered a rare and plain example of uranium intermetallic with strongly localized $f$-electrons where magnetism can be explained within the crystal level scheme. Nevertheless, around room temperature it too shows a heat capacity anomaly, which has been a long-standing and puzzling mystery$^7$. Here, using single crystal neutron and x-ray diffraction we discover that a crystal lattice modulation develops in UPt$_2$Si$_2$ below $\approx 320$ K. The modulation wavevector, $Q_{\text{mod}} \approx (0.4 0 0)$, corresponds to the Fermi surface nesting wavevector of URu$_2$Si$_2$,$^8$ suggesting that similar Fermi surface driven physics exists in these seemingly different systems. Furthermore, the superlattice Bragg peaks show an anomaly around 200 K where the wavevector is pinned down to an incommensurate position, suggesting a transition from local to Kondo behavior. We thus stipulate the same underlying physics in both materials, where only a subtle perturbation of nearly degenerate competing energy scales leads to different ground states.

In strong contrast with the hidden order (HO) and itinerant magnetism in URu$_2$Si$_2$, its sister material UPt$_2$Si$_2$ shows antiferromagnetic (AFM) order below $T_N$, with a large ordered local magnetic moment of $\approx 2 \mu_B$, where ferromagnetic (FM) ab-planes are stacked antiferromagnetically along c-axis$^5$. It was suggested that a simple crystal field scheme can account for the anisotropy, temperature and field dependence of magnetization$^6$. Recent studies$^{10-13}$, however, questioned this assessment and the degree of electron localization in this system, suggesting that phase transitions under applied magnetic field should be understood in terms of Fermi surface effects$^{10}$. This approach has been further supported by a density functional theory (DFT) calculation$^{11}$. Very re-
cent inelastic neutron scattering study revealed dual nature, both itinerant and local, in magnetic excitation of UPt$_2$Si$_2$. The crystal structure of UPt$_2$Si$_2$ and its relationship with electronic properties have been another important longstanding scientific controversy. The x-ray diffraction measurements found large in-plane anisotropic thermal factors on Pt(2) and Si(2) sites, which were interpreted as an indication of strong crystallographic disorder. This purported disorder was thought to be responsible for the highly anisotropic resistivity, which showed Anderson localization along the c-axis while being metallic in the ab-plane.

The origin and the nature of the disorder in this heavy fermion system, however, has long been open to questions. Most UT$_2$M$_2$ (T: transition metal, M: Si or Ge) compounds crystallize into the ThCr$_2$Si$_2$ type structures and only a few, including UPt$_2$Si$_2$, have been reported to adopt the CaBe$_2$Ge$_2$ type structure. Therefore, when a specific heat anomaly around room temperature was found, researchers anticipated that there could be a structural transition between the two closely related crystal structures leading to disorder. However, x-ray diffraction measurements on a polycrystalline sample did not find any indication of a symmetry change. Moreover, a comparative transport study of as-grown and annealed samples shows that the effect from the presumed disorder does not depend on synthesis conditions, or impurity level.

In this Letter, we present neutron and synchrotron x-ray diffraction measurements on a single crystal of UPt$_2$Si$_2$. We find that the system develops an incommensurate charge modulation, which is responsible for the heat capacity anomaly and which was previously interpreted as a putative
disorder. Figure 1 (a) shows single crystal neutron diffraction data obtained at WAND neutron diffractometer at $T = 50 \text{ K}$, which is above $T_N = 35 \text{ K}$ but well below the heat capacity anomaly temperature of $\approx 310 \text{ K}$. We observe a pattern of satellite peaks offset from the original lattice Bragg reflections, with a wavevector $Q_{\text{mod}} = (\tau 0 0)$, $(0 \tau 0)$ where $\tau \approx 0.4$. We investigated the origin of these superlattice reflections by performing neutron polarization analysis on the $(2 0.4 0)$ and $(2 1.6 0)$ satellite peaks at $50 \text{ K}$ at HB1 triple axis spectrometer, which is shown in Fig. 1 (e) and (f), respectively. Non-spin-flip scattering dominates the scattering intensity of both reflections, confirming that the incommensurate order is in the non-magnetic charge/lattice degrees of freedom. Our findings thus correct the previous reports on the atomic disorder, which in fact was due to the mis-assignment of the observed suppression of the lattice principal Bragg reflections to disorder without considering the charge modulation.

This finding is worthy of special note in that the modulation wavevector leading to the charge density wave (CDW) in $\text{UPt}_2\text{Si}_2$ corresponds to the incommensurate wave vector $Q_1$, responsible for the heat capacity reduction in the hidden order phase in $\text{URu}_2\text{Si}_2$, which is believed to originate from Fermi surface nesting (FSN). Hence, the observed CDW precursor to AFM in $\text{UPt}_2\text{Si}_2$ can be considered an analogue of HO precursor to superconductivity in $\text{URu}_2\text{Si}_2$.

By virtue of WAND diffraction patterns covering wide reciprocal space area, Fig. 1 (a), we can clearly identify the polarization of the observed atomic displacements. As shown in Fig. 1 (b) and (c), the intensities of longitudinal superlattice peaks, $(H \pm \tau 0 0)$, or, equivalently, $(0 K \pm \tau 0)$, are absent, or very weak compared to those of transverse superlattice peaks, $(\pm \tau K 0)$ and $(H$
±τ 0). For example, the integrated intensity of a longitudinal (4.4 0 0) peak is about 240 times smaller compared to the corresponding transverse (4 0.4 0) peak. For lattice Bragg peaks near the diagonal, (H H 0) direction we find all 4 satellite reflections with comparable intensity, Fig. 1 (d).

The intensity of a satellite peak at a momentum transfer \( q \) can be expressed as\cite{17, 18},

\[
I(q) \approx \left| \sum_\nu (q \cdot \epsilon_\nu) f_\nu(q \pm Q_{\text{mod}}) \right|^2 \delta(q - G \pm Q_{\text{mod}}),
\]

e.g. using Taylor expansion of Jacobi-Anger identity. Here, \( \epsilon_\nu \) is the displacement of atom \( \nu \) in the unit cell and \( G \) is the reciprocal lattice vector. The \((q \cdot \epsilon_\nu)\) term in the satellite reflections is responsible for the observed polarization dependence of the diffraction pattern, indicating that the atomic displacements, \( \epsilon_\nu \), are almost exclusively transverse, either along a axis for \( Q_{\text{mod}} \approx (0 0.4 0) \), or along b axis for \( Q_{\text{mod}} \approx (0.4 0 0) \). While the observed pattern of superlattice reflections can be regarded as overlaid patterns from two \( Q \)-domains, \( Q_{\text{mod}} \approx (0 0.4 0) \) and \( Q_{\text{mod}} \approx (0.4 0 0) \), within our resolution we were not able to detect any clear sign of tetragonal symmetry breaking that would correspond to this \( Q \)-domain structure.

To understand the atomic modulation pattern in real space, we performed DFT calculation by relaxing atomic positions in a \( 5 \times 1 \times 1 \) supercell and we find a reduction of total energy by 15.3 meV per unit cell when CDW develops. As shown in Fig. 2 (a), there are two kinds of Pt-Si layers in UPt2Si2. In the (Pt1-Si1-Pt1) layer, a square lattice of Si atoms is sandwiched between \( \sqrt{2} \times \sqrt{2} \) square lattice of Pt atoms, while in the (Si2-Pt2-Si2) layer the situation is inverted. Our calculation shows that more than 90 % of displacement is in the ab-plane of the Si2-Pt2-Si2 layer, in agreement with where the putative disorder was previously reported\cite{15}. It is interesting to note
that the lattice modulation of sandwiched metallic layers is quite ubiquitous as found in Pt-based layered superconductors SrPt$_2$As$_2$ and LaPt$_2$Si$_2$\textsuperscript{19,20} or layered transition-metal dichalcogenides 2H-NbSe$_2$, 2H-TaSe$_2$, and 1T-TaS$_2$\textsuperscript{21}.

Figure 3 (a) shows the evolution of the superlattice Bragg peak at $q = (2 0.4 0)$ with temperature. The intensity keeps increasing as sample is cooled down, indicating gradual development of the superlattice modulation, Fig. 3 (c). We find a second-order-like transition with a transition temperature $T_{\text{CDW}} = 319(8)$ K, consistent with the previous heat capacity anomaly temperature\textsuperscript{7} and critical exponent consistent with the mean field behavior. Surprisingly, as shown in Fig. 3 (b), the peak moves away from the nearly commensurate position of $\tau = 0.40(1)$ observed at 300 K and becomes pinned at an incommensurate value 0.418(2) below about 200 K, thus indicating an anomaly around $T_K = 200$ K. Comparison with previous bulk measurements shows that $T_K$ is where magnetic susceptibility deviates from the Curie-Weiss dependence\textsuperscript{6} and also where the a-axis resistivity shows broad maximum\textsuperscript{22}. This strongly suggests that $T_K$ is the Kondo coherence temperature, which marks a cross-over from paramagnetic, weakly correlated electrons to correlated, heavy-electron behavior. The gradual change of $Q_{\text{mod}}$ from nearly commensurate to incommensurate is, therefore, likely to be the result of a transition from local-moment to Kondo-like system. Though there are a few non-5$f$ systems which show coexistence of CDW and Kondo states\textsuperscript{23,24}, UPt$_2$Si$_2$ is the first demonstration of direct coupling between CDW and Kondo effect and could serve as a test bed to study the interplay of these two phenomena.

The nature of the CDW have been investigated in more detail with a synchrotron x-ray.
A careful inspection of the diffraction pattern reveals that besides the first harmonic superlattice reflections with $Q_{\text{mod}}$, there are also weak superlattice reflections indexable as the second and the third harmonics located around $(4 3 - \delta K 2)$, Fig. 4 (a). At high temperature, though weak, the second and third harmonics nearly overlap at $q = (4 3.8 2)$ as higher harmonics will be found at $(4 3+2Q_{\text{mod}} 2)$ and $(4 5-3Q_{\text{mod}} 2)$ while $Q_{\text{mod}}$ is approximately commensurate with $\tau = 0.4$. As the temperature decreases from $T_{\text{CDW}}$, the second and third harmonic peaks get stronger in intensity and begin to split as the incommensurability increases. Their positions are locked in below $T_K$ at the corresponding incommensurate $2\tau$ and $3\tau$ positions, confirming their higher harmonics origin.

We show the temperature dependence of the integrated intensities from all harmonics in Fig. 4 (b). The principal harmonics increase around $T_{\text{CDW}}$ follows a long tail of critical fluctuation precursor to CDW at $T > T_{\text{CDW}}$. We find an indication of the second phase transition around 270 K, where the intensities of higher harmonics begin to increase and the CDW order parameter becomes two-component. The existence of such higher harmonics suggests that the atomic CDW modulation is not ideally sinusoidal, as also noted in DFT discussion. Lastly, the intensities show also an anomaly around $T_K$, indicating transition from the local to Kondo regime, below which the atomic modulation gets “squared” and becomes commensurate with the lattice.

It is provocative to compare our UPt$_2$Si$_2$ with URu$_2$Si$_2$. While these systems seem to be very different at the first glance, one with strongly itinerant and the other with strongly local character, they indeed share many common features. They have very closely related crystal structures and the same magnetic structures, FM in-plane and AFM along the c-axis, although the magnetic order in
URu$_2$Si$_2$ seems to hover in the background, ready to appear under pressure or magnetic field\textsuperscript{25,26}. Their magnetic susceptibilities, $\chi$, show deviation from Curie-Weiss behavior around 200 K. At temperatrures below the $\chi$ maximum, URu$_2$Si$_2$ shows a formation of coherent Fermi liquid leading to the hidden order and superconductivity\textsuperscript{2}. UPt$_2$Si$_2$, as we find here, forms an incommensurate CDW, which leads to an electronic ground state of an antiferromagnet\textsuperscript{5}. At the same time, thermoelectric measurements\textsuperscript{27} suggest that both systems show Fermi surface reconstruction with a partial gap opening upon hidden order, or antiferromagnetic transition, respectively.

Here, we show that both systems have same two important wavevectors at play. Considering these shared wavevectors characterizing their respective ground states, it is very likely that the same underlying physics exists in both materials while a subtle perturbation onto nearly degenerate competing energy scales leads to different ground states. From our Bader charge analysis\textsuperscript{28}, the net charge transfer from Uranium atom to nearby metallic layer (Pt-Si and Ru-Si layer for UPt$_2$Si$_2$ and URu$_2$Si$_2$, respectively) is the same, with 1.6 electron for both compounds, providing a similar electronic configuration. While UPt$_2$Si$_2$ has been considered a classical local-moment system and URu$_2$Si$_2$ an itinerant Kondo material, our findings suggest that both systems are at the boundary between local and itinerant\textsuperscript{13} and share common physics, which therefore calls for a unified theoretical understanding.

**Methods**

Neutron diffraction measurements were carried out at Wide Angle Neutron Diffractometer (WAND) and HB1 Polarized Triple-Axis Spectrometer (PTAX) at High Flux Isotope Reactor (HFIR), ORNL.
The incident wavelength was 1.486 Å for WAND measurements and 2.462 Å for PTAX experiment. Data were collected in the temperature range between 2 K and 330 K using closed cycle refrigerator (CCR) with sample loaded into an aluminum can. For PTAX measurement, the neutron spin was aligned parallel to wave vector transfer, \( \mathbf{Q} \), so that Non-Spin-Flip (NSF) scattering is nuclear and Spin-Flip (SF) is magnetic. The flipping ratio of PTAX was approximately 15. Additional neutron diffraction experiment was performed on the E4 two-axis diffractometer at the Helmholtz-Zentrum Berlin. E4 uses a vertically focused PG monochromator that selects 2.4 Å neutrons and a PG filter placed in the incident beam to reduce higher-order wavelength contamination. A radial collimator is inserted after the sample and a 2-D position-sensitive detector records the diffracted neutrons.

Synchrotron x-ray experiments were carried out at A1 beam line at Cornell High Energy Synchrotron Source (CHESS). Fixed incident energy of 19.9 keV and an area detector were used. The sample temperature was controlled using cryostream in the range from 100 K to 400 K.

All x-ray measurements were in reflection geometry, while neutron measurements were performed in transmission geometry. Samples used for x-ray and neutron diffraction are previously described single crystals.

We carried out density-functional theory (DFT) calculations via the Generalized Gradient Approximation (GGA) using Perdew-Burke-Ernzerhof (PBE) functional, as implemented in the VASP package. The local nature of Uranium f-electrons was described by Hubbard-type onsite and nearest Coulomb repulsions \( U = 2.0 \) eV and \( J = 0.5 \) eV. The experimental lattice parameters,
a = 4.188 Å and c = 9.918 Å were used. The cutoff energy of plane wave basis was set at 400 eV and k-points were sampled by $20 \times 20 \times 10$ Monkhorst-Pack grid including gamma point for a unit cell. We compared the total energy of the optimized structures for $4 \times 1 \times 1$, $5 \times 1 \times 1$, $5 \times 5 \times 1$ and $6 \times 1 \times 1$ supercells and confirmed that $5 \times 1 \times 1$ superstructure is the most stable among these.

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Figure 1: Neutron diffraction measured at 50 K with Wide Angle Neutron Diffractometer, WAND. (a) a full map in (H K 0) plane and zoomed-in views around (b) (0 4 0), (c) (4 0 0), and (d) (2 2 0) showing peak extinction that reveals the polarization of atomic displacements. Spin-Flip(SF) and Non-Spin-Flip(NSF) intensities from polarized triple-axis spectrometer, PTAX, measured at 50 K at (e) $q = (2 0.4 0)$ and (f) (2 1.6 0).
Figure 2: DFT simulation of the atomic displacements pattern. (a) crystal structure of UPt$_2$Si$_2$ with bond between Pt and Si atoms demonstrating two different kinds of Pt-Si layers in a CaBe$_2$Ge$_2$-type structure. The atomic labels follow the convention which reports disorder in UPt$_2$Si$_2$. (b) simulated atomic modulation of Pt$_2$ atom positions in the ab-plane associated with the CDW.
Figure 3: Temperature dependence of superlattice peak. (a) gradual change of \( q = (2 \tau 0) \) satellite Bragg peak, and (b) temperature dependent peak position of \( \tau \) which locks in around \( T_K = 200 \) K. (c) integrated intensity of \( (\tau 0 0) \) measured up to 317 K. The data presented in (a) and (b) are measured from 4 K to 300 K measured at PTAX in NSF configuration, and therefore are of charge/lattice origin in nature. The data in (c) were measured with a thermal neutron diffractometer E4.
Figure 4: Temperature dependence of the diffraction pattern measured using synchrotron x-ray at A1, CHESS. (a) temperature dependence of harmonics reflections around \( q = (4 - 3\delta K 2) \). (b) the integrated intensity of principal harmonic (black square), second harmonic (red circle), and third harmonic (blue triangle). The scale axis for the principle harmonic is separately shown on the right.