Mysterious hidden order, superconductivity and magnetism of URu_2Si_2

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Abstract

The term "Hidden Order" (HO), brought up ten years ago, has been used to describe unknown ordered state whose origin could not explained by conventional solid state probes and is able to prove superconductivity at lower temperature ($T_c \approx 1.5\ K$). This essay reviews the 25 years' efforts to understand the continuous (second-order) and mean-field-like phase transition occurring at 17.5 K in URu_2Si_2 , known as HO, mainly through experimental phenomena, in particular, construction of the Fermi Surface (FS) and destroying HO via large magnetic field and Rh doping. Recent theoretical models will also be discussed at the end.

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1 Introduction to U-based compounds

The observed unusual behaviors of U-based compounds can be explained by open $5f$ shell of Uranium since the exchange interaction, the $5f$ bandwidth, the spin-orbital interaction and the intra-atom f-f Coulomb interaction are all within the same energy scale. As a result, (a) U displays the properties ranging from transition metals to rare earths and it has largest spin-orbital interaction leading to symmetry breaking effects[1]; (b) the Wigner-Seitz radius R_{WS}[1] of comparative elements places U near the minimum between metallic and atomic $5f$ wave functions; and (c) ionic U can adopt six different valences when combined with other elements: one usually finds U^{4+} ($5f^3 6d^1 7s^2 \rightarrow 5f^2$) or U^{3+} ($5f^3 6d^1 7s^2 \rightarrow 5f^3$). However, it is difficult to distinguish or separate these two valences in metallic systems with strong hybridized f states which play a major role in the ground state properties[1].

According to Hill plot, one can observe superconducting compound should be dominant for small U-U distances, and a significant magnetic transitions can be found for distances larger than 3.5 $\mathring{A}$ (Fig. 1). There are nine unconventional U-based compounds that combine superconductivity with magnetism (ferromagnetism and antiferromagnetism) or nearly magnetic behav-

[1]The WignerSeitz radius is the radius of a sphere whose volume is equal to the mean volume per atom in a solid defined as $R_{WS} = (3/4\pi n)^{1/3}$ where n is the particle density.

iors, among which URu_2Si_2 has been the only continuing and intense interest for the past 25 years[2].

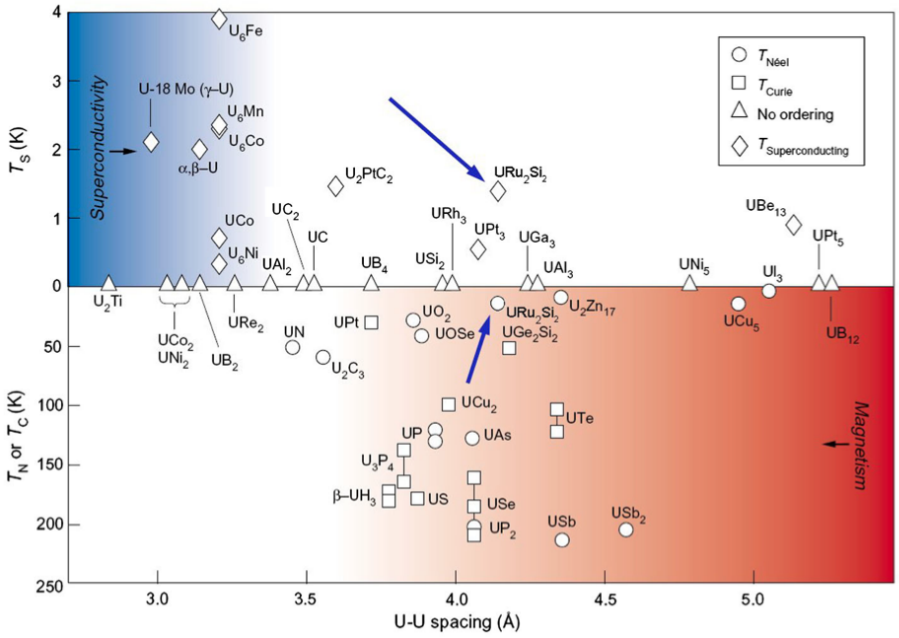
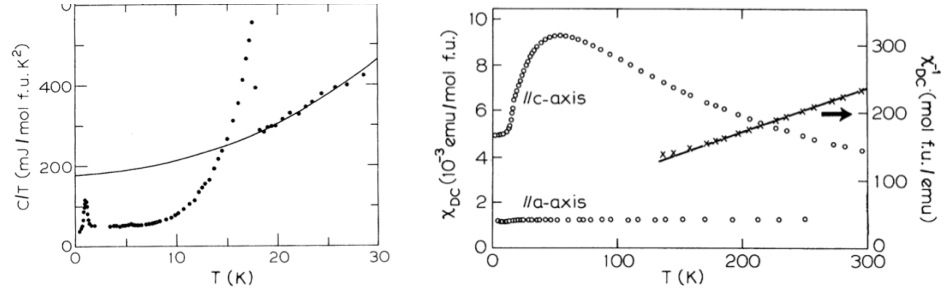


Figure 1: The hill plot for U-based compounds. The bottom arrows indicates the hidden order transition temperature at T_o, while the top one indicates the superconducting transition temperature at T_c.

1.1 The continuing interest in URu_2Si_2

A poster in 1984 by Schlabitz *et al.* (unpublished) showing two transitions: one superconducting and the other antiferromagnetic, which followed by a later publication[3]. Both agreed on the superconducting transition at $\approx 1.0\ K$ while differed on the origin of magnetic transition at $17.5\ K$. We now understand the transition is not due to long-range-ordered magnetism and there is no measurable lattice modulation relating to static Charge Density Wave (CDW) or Spin Density Wave (SDW) formation. Without well defined Order Parameter (OP) for the emergent phase or for its characteristic elementary excitations, the term "Hidden Order" (HO) was adopted to describe the mysterious phase occurring at $T_o = 17.5\ K$[2].

Fig. 2(a) implies two mean-field-like phase transitions in the specific heat. Large amount of entropy S forms at T_o then S is approximately $0.2Rln2$ given by $\int_0^{T_o}(\Delta C/T)dT$. The linear magnetic susceptibility $\chi_{dc} = M/H$ with applied field $H = 2\ T$ (magnetic response) is strong Ising like, as there is a magnetic signal only along c which begins to deviate from a local-moment Curie-Weiss dependence below 150 K (Fig. 2(b))[3]. Evidently, the maximum of χ_{dc} at $\approx 60\ K$ indicates that URu_2Si_2 is not a conventional bulk antiferromagnet[2] but corresponding to a coherence temperature T^* and the formation of Fermi liquid.



(a) (b)

Figure 2: (a) Specific heat (C/T) of URu_2Si_2 as a function of T showing the entropy balance and superconducting transition. (b) dc susceptibility χ_{dc} of URu_2Si_2 with applied field $H = 2\ T$ along the a and c axes. Note the deviation from the Curie-Weiss law ($\mu_{eff} = 3.51\ \mu_B/U$; $\theta_{CW} = -65\ K$) along the c axis below 150 K[3].

Although the HO transition occurring at 17.5 K is sample quality independent, the low-temperature properties are sample dependent. As the heavy-fermion state is created when the temperature decreases, the spin (fluctuation) scattering is removed and a coherent low-carrier state without significant scattering is formed. The superconducting transition temperature T_c varies significantly (between 0.8 K and 1.5 K) with sample quality and purity and coexists with the HO on a microscopic scale without disturbing it[3].

Transport and thermodynamic measurements indicated a considerable Fermi Surface (FS) reconstruction occurs at the HO transition[3]. The ob-

[2]The magnetic susceptibility of a conventional antiferromagnetic material typically reaches maximum at the Néel temperature which is $T_o = 17.5\ K$ of URu_2Si_2.

served electronic specific heat in the HO state and the jump in the resistivity at the transition were consistent with the opening of an energy gap of the FS given by $C_e(T) \propto exp(-\Delta/k_BT)$ where Δ is the charge gap opening in the electronic spectrum below T_o. The thermal expansion coefficients exhibited a large in-plane positive peak with a small negative one along the c axis leading to a net increase of volume, which implied a significant coupling of the HO to the lattice. The optical conductivity measurement also supported the reconstruction of the FS in the transition to the HO state. Hence the HO state possesses both a spin and a charge gap combined the transport, thermodynamic and optical data. This is the first clue of microscopic HO behavior[2].

1.2 The Hidden Order state in URu_2Si_2

The HO involves a continuous (second-order) and mean-field-like transition to a unknown novel phase at $T_o = 17.5\ K$, of which OP and elementary excitations could not be understood by microscopic experiments. Up until now there is no comprehensive mechanism of generic HO state and its relation to quantum criticality. In the URu_2Si_2 case, the HO state is identified as a staggered alignment of magnetic multipoles[2].

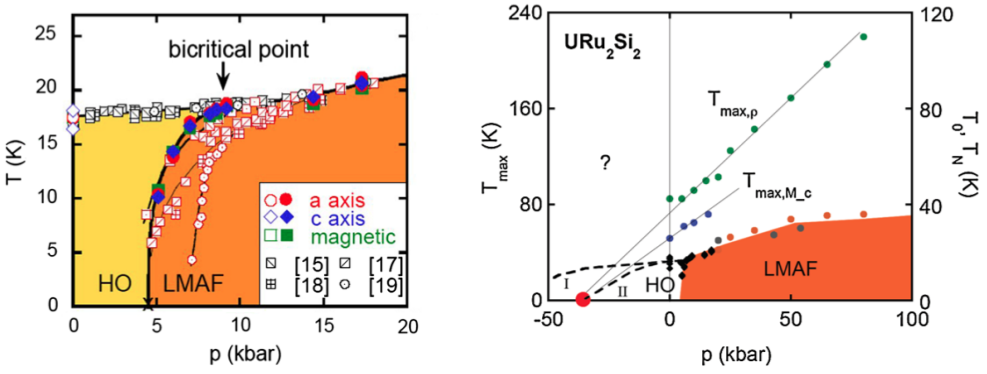
Based on previous experimental results, the principle properties of the HO in URu_2Si_2 are summarized: (a) a large entropy forming at T_o; (b) greatly reduced carrier concentration; (c) the opening of charge and spin gaps; (d) a clear coupling to the lattice; and (e) an electronically ordered state that can by destroyed by pressure, magnetic field and Rh doping.

2 Experimental Survey

The very recent measurements of phase diagram of URu_2Si_2 (Fig. 3) indicated that the HO and the Large Moment AntiFerromagnetic (LMAF) phase are intrinsically separated by a phase boundary which has to be first-order with a bicritical point at 9 $kbar$ and 18 K, since three second-order phase transition lines cannot meet in one point[3]. Therefore the HO and LMAF could not have the same symmetry. This suggested unconventional

[3]The phase boundaries from the HO and LMAF to the disordered high-temperature phase are already known to be of second order from qualitative heat capacity measurements

properties of the HO such as incommensurate orbital currents, helicity order, or multipolar order[4].



(a) (b)

Figure 3: (a) Phase diagram based on Larmor diffraction and conventional magnetic diffraction data. The onset of LMAF and HO is marked by full and empty symbols respectively, where symbols with bright contours refer to T_N and ones with dark contours to T_o. (b) Extended phase diagram of URu_2Si_2: $T_{max,\ \rho}$ and $T_{max,\ M_c}$ denote coherence maxima in the resistivity and magnetization respectively. The HO might either mask a Quantum Critical Point (QCP) (I) or replace LMAF (II) near quantum criticality[4].

2.1 Establishment of the Fermi Surface of URu_2Si_2

An interesting investigation of the FS of HO as well as LMAF phase comes from de Haas-van Alphen (dHvA)[4] and Shubnikov-de Haas (SdH)[5] measurements under pressure (Fig. 4). The early angular dependent dHvA measurements of Ohkuni *et al.* (1999) revealed three, rather small, closed FS pockets deep in the HO state, which is consistent with a substantial FS gapping occurring in the HO state. The recent angle dependent SdH

[4]The de Haas-van Alphen (dHvA), discovered in 1930 by Wander Johannes de Haas and his student P. M. van Alphen, is a quantum mechanical effect in which the magnetic moment of a pure metal crystal oscillates as the applied field H is increased.

[5]The Shubnikovde Haas effect (SdH) is the oscillation in the conductivity of a material that occurs at low temperatures in the presence of very intense magnetic fields, used to determine the effective mass of charge carriers (electrons and electron holes)

measurements revealed a previously undetected splitting of β branch as well as a new η branch which coincided with previously observed γ branch (Fig. 4)[5].

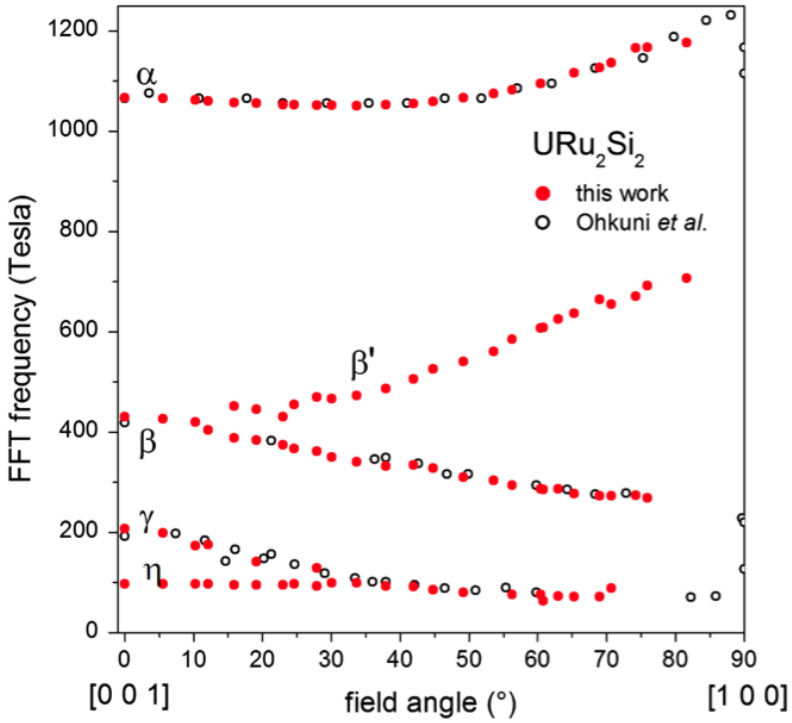
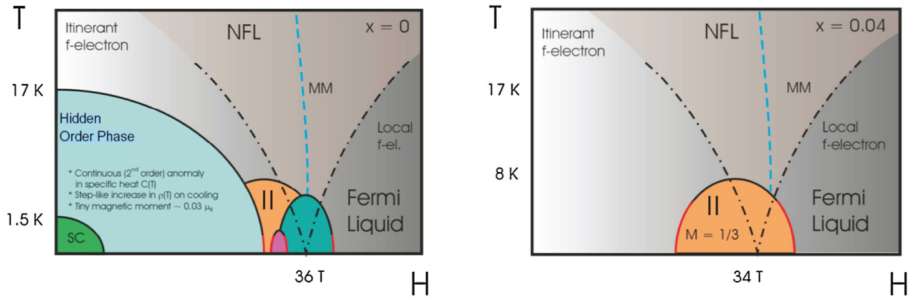


Figure 4: Angular dependence of the Fast Fourior Transform (FFT) frequencies obtained from SdH measurements in URu_2Si_2, reflecting the cross sectional areas of the different FS branches perpendicular to the magnetic field[5].

By increasing the pressure the LMAF was reached (Fig. 3) and the quantum oscillations were measured and compared to those in the HO state. There was little changes in the FS between these two phases. This is unexpected since the HO and LMAF phases are considered to be separated by first-order phase transition[4]. The new SdH result supports the assumption that the lattice doubling and modified Brillouin zone of the LMAF are already present in the HO state. This also suggests the HO transition breaks bct translational symmetry, which agrees to the various angular dependent FS branches[5].

2.2 High magnetic fields and Rh doping behavior

The very recent high field SdH experiments discovered the reconstruction of the FS changes when leaving the HO phase to phase II scanning the applied field to 35.9 T (H_o) and appearance of a novel phase between 36.1 T (H_1) and 39.7 T (H_2) without additional phase transition above 40 T (Fig. 5(a)). Here the high-field phases determined via the resistivity imply qualitatively the same number and T-H shapes as the ambient-pressure phases[6]. This means that pressurizing the material from HO to LMAF does not alter the resulting high field phase formations, again indicating the similar FS in HO and LMAF.



(a) (b)

Figure 5: The high magnetic field T-H phase diagrams for URu_2Si_2 and $U(Ru_{0.96}Rh_{0.04})_2Si_2$. MM represents the metamagnetic transitions and NFL is the Non Fermi Liquid[6].

The Rh doping helps us to simplify the high field phase diagram (Fig. 5(b)). Above 4 % Rh substitution on the Ru site, five complicated phase were reduced to single field-induced phase: the surviving symmetric domelike shape of phase II spreading the field from 26 to 37 T with a peak at 9 K. A single point of the resistivity and magnetization data of heavy fermion liquid was reached at 34 T and 0 K, supporting that field-induced QCP is suppressed by the formation of a field-induced novel phase[2]. A recent theoretical study showed the phase diagram of Rh doped URu_2Si_2 could be explained as competition between two coupling OPs[7].

3 Theoretical models

There have been a large amount of theoretical contributions to the HO state in the past 25 years while some of them are still in their initial phases requiring more detailed modifications compared with the present more comprehensive experimental developments[2].

3.1 The Crystalline Electric Field model

The earlier CEF model focused on explaining the extraordinarily small and its Ising like behavior. The first CEF model considered U^{4+} $(5f^2)$ ion with total angular moment $J = 4$ in a tetragonal crystal field and obtained an agreement with experimental susceptibility data when the lowest three CEF levels (Eq. (2) to (4)) satisfying the Hamiltonian (Eq. (1)) were singlets with a splitting of around 35 K between the ground and first excited states Eq. (2) and Eq. (3) respectively[8]. Correspondingly the predicted ordered moment was, however, 10 times larger than the measured one.

$$\mathcal{H}_{CF} = B_2^0\hat{O}_2^0 + B_4^0\hat{O}_4^0 + B_4^4\hat{O}_4^4 + B_6^0\hat{O}_6^0 + B_6^4\hat{O}_6^4 \tag{1}$$

where $\hat{O}_j^k$ are Stevens operators and B_m^n are the coefficients for URu_2Si_2.

$$\left|\Gamma_1^{(1)}\right\rangle = \epsilon(|4\rangle + |-4\rangle) + \gamma\,|0\rangle \tag{2}$$

$$|\Gamma_2\rangle = (|4\rangle - |-4\rangle)/\sqrt{2} \tag{3}$$

$$\left|\Gamma_1^{(2)}\right\rangle = (\gamma/\sqrt{2})(|4\rangle + |-4\rangle) + \sqrt{2}\epsilon\,|0\rangle \tag{4}$$

where $2\epsilon^2 + \gamma^2 = 1$.

A later extensive CEF study was developed by considering the same previous U^{4+} $(5f^2)$ CEF Hamiltonian, but analyzing the possible ordering of nine CEF states in more details and considering the different angular momentum operators that could support multipolar OPs[9]. They discovered three possible variants for the three lowest singlet states: A, considered previously, supported dipole order $[J_z]$ simultaneously with hexadecapole order $[J_xJ_y(J_x^2 - J_y^2)]$ but was rejected because it predicted a higher contribution to the specific heat; B and C, both sustained quadrupole order $[J_xJ_y$ or $J_x^2 - J_y^2]$ and octupole order $[J_z(J_x^2 - J_y^2)$ or $J_zJ_xJ_y]$. Compared with previous experiment results, they favored electric quadrupolar order of localized f electrons

for the HO state where the dipole matrix would be zero.

Unfortunately, there is no up-to-date experimental evidence for the localized CEF levels and splitting, causing the exclusion of quadrupole orders. Although CEF models provide insight of possible multipole symmetries on a single U^{4+} $(5f^2)$ ion, they are not sufficient to achieve collective long-range order, which requires exchange couplings of multipoles[2].

3.2 The two Order Parameters of Hidden Order

Two interacting OPs was introduced in Ginzburg-Landau free energy expansion in order to describe the HO phase transition: ψ, the primary unknown OP of the HO state and m, the magnetization of the LMAF state determined by neutron scattering and diffraction[10]. They predicted magnetic field dependence of both ψ and m and determined the symmetry of the coupling between these two OPs: $g\psi \cdot m$ (bilnear) or $g\psi^2 \cdot m^2$ (biquadratic). The comparison with measured magnetic field dependence of the neutron moment favored linear coupling mechanism, thereby indicating that ψ breaks time-reversal symmetry.

Based on combination of the DFT and temperature-dependent Density Microscopic Functional Theory (DMFT) applied to a U^{4+} $(5f^2)$ localized configuration, a CEF mechanism consisting of two lowest CEF levels $\Gamma_1^{(1)}$ and Γ_2 with a splitting of $\approx 35\ K$ was derived[11]. This mechanism supports a complex OP, consisting of both a dipolar order and a hexadecapolar order.

As the temperature is lowered, the system evolves from a local $5f^2$ configuration into a multichannel Kondo state, which "arrested" by the CEF splitting at temperature lower than $35\ K$. The two phases, HO and LMAF, thereby are treated as real and imaginary parts of the same OP respectively, due to collective CEF excitations. Accordingly, the HO phase transition is the formation of the real part of the nonmagnetic hexadecapole OP which breaks rotational symmetry but preserves time-reversal symmetry. Upon critical pressure, the imaginary part of the OP dominates and corresponds to the LMAF phase with its time-reversal symmetry breaking. Therefore this complex OP exists in both phases with different symmetry breaking properties[11].

4 Conclusion

After 25 years' developments aiming at understanding the HO state of URu_2Si_2, the whole story still remained unknown. Yet the reconstruction of FS has been well established and other quantities such as the FS gap and dynamical magnetic susceptibility were proven to be mean-field-like OP behavior, which provided motivation of further searching for OPs[2]$\sim$[5]. The high field and doping behavior indicated the fragility of the HO state and its uniqueness among all the heavy fermion materials[6]. Surprising small antiferromagnetic moment in the HO state would continuously trigger intensive interests since the up-to-date theories could not be able to explain those experimental phenomena completely. In addition, at least Spontaneous Symmetry Breaking (SSB) was partially studied, and they are now clear implication of unit cell doubling along the c axis and a first observation of fourfold rotational symmetry breaking in the basal plane[10],[11]. The mysterious problem of URu_2Si_2 will improve our understanding of the novel ordered phases.

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