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Thermodynamic evidence for broken fourfold rotational symmetry in the hidden-order phase of URu_2Si_2

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ABSTRACT

Despite more than a quarter century of research, the nature of the second-order phase transition in the heavy-fermion metal URu_2Si_2 remains enigmatic. The key question is which symmetry is being broken below this "hidden order" transition. We review the recent progress on this issue, particularly focusing on the thermodynamic evidence from very sensitive micro-cantilever magnetic torque measurements that the fourfold rotational symmetry of the underlying tetragonal crystal is broken. The angle dependence of the torque under in-plane field rotation exhibits the twofold oscillation term, which sets in just below the transition temperature. This observation restricts the symmetry of the hidden order parameter to the E^+- or E^--type, depending on whether the time reversal symmetry is preserved or not.

1. Introduction

At T_h = 17.5 K, URu_2Si_2 undergoes a second-order phase transition accompanied by the large anomalies in thermodynamic and transport properties [1–4]. Several remarkable features have been reported in the lower-temperature hidden order phase [5]. URu_2Si_2 exhibits the body-centered tetragonal crystal structure (see inset of Fig. 1), and to date no structural phase transition is observed at T_h. Tiny magnetic moment appears ($M_0 \sim 0.03\mu_B$) below T_h [6], but it is by far too small to explain the large entropy released during the transition and seems to have an extrinsic origin [7–9]. The electronic excitation gap is formed at a large portion of the Fermi surface and most of the carriers (~90%) disappears [10–12]. The gap is also formed in the incommensurate magnetic excitations below the hidden order transition [13]. With application of pressure, the large-moment antiferromagnetic state with the wave vector $\mathbf{Q}_c$ = (0,0,1) = (1,0,0) appears, which is separated from the hidden-order phase by the first-order phase transition (see Fig. 1) [5,9]. The quantum oscillation experiments [14] show that the Fermi surface of the hidden-order phase is quite similar to that of the pressure-induced antiferromagnetic phase. These results suggest that the hidden order and antiferromagnetism are nearly degenerate.

In general, a second-order phase transition causes a change in various type of symmetries, such as crystal, rotational, gauge and time reversal symmetries. An order parameter is introduced to describe the low-temperature ordered phase with reduced symmetries. Therefore the key to the nature of hidden order lies in understanding which symmetry is being broken. Several microscopic models, including multipole ordering [15–19], spin-density-wave formation [20–22], orbital currents [23] and helicity order [24], have been proposed. However, in spite of the intensive experimental and theoretical studies, what is the genuine order parameter in the hidden order phase is still an open question.

2. Searching for fourfold rotational symmetry breaking

2.1. Information from the superconducting state embedded in the hidden-order phase

Inside the hidden-order phase, the unconventional superconductivity appears below T_c = 1.4 K. When the antiferromagnetic phase is induced by the pressure, the superconductivity disappears [see Fig. 1]: i.e. the superconductivity can coexist with the hidden order, but not with the antiferromagnetic order. Therefore the information on the superconducting state is also important for understanding the hidden order. The thermal conductivity [12,25] and specific heat [26] measurements using very clean single crystals, which become available recently [27], consistently show that the field dependence of these quantities has strong anisotropy. From these results, both suggest the existence of point nodes along the c-axis in the superconducting order parameter. The symmetry analysis for the spin-singlet states leads to the chiral d-wave symmetry with the form [12]:

$$\sin\frac{k_z}{2}c\left(\sin\frac{k_x+k_y}{2}a \pm i\sin\frac{k_x-k_y}{2}a\right) \tag{1}$$

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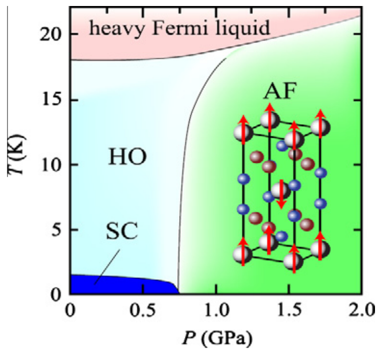

Fig. 1. Schematic temperature–pressure phase diagram of URu_2Si_2. At ambient pressure, the second-order phase transition from the paramagnetic, heavy Fermi liquid metallic state to the hidden order (HO) state occurs at T_h = 17.5 K. At lower temperature below T_c = 1.4 K superconducting (SC) state appears. Above the critical pressure (∼0.75 GPa), the large-moment antiferromagnetic (AF) state sets in through the first-order phase transition. The inset illustrates the schematic crystal structure (large white spheres, middle red spheres, small blue spheres are U, Ru, and Si atoms, respectively) with the spin arrangements in the antiferromagnetic phase. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

that has the zero points along the Γ and X lines in the Brillouin zone as well as the horizontal line nodes as shown in Fig. 2. This symmetry of superconducting order parameter $\Delta(\boldsymbol{k})$ has sign change at the points connecting with the antiferromagnetic vectors $\boldsymbol{Q}_c$ = (0, 0, 1) and (1, 0, 0), which are equivalent for the body-centered tetragonal structure. This sign-changing order parameter is consistent with the superconductivity mediated by the antiferromagnetic spin fluctuations. The commensulate antiferromagnetic fluctuations lead to the enhanced dynamical susceptibility at $\boldsymbol{Q}_c$ and hence the pairing interaction becomes large and repulsive at $\boldsymbol{Q}_c$, leading to the sign change of the superconducting order parameter $\Delta(\boldsymbol{k}) = -\Delta(\boldsymbol{k}+\boldsymbol{Q}_c)$ [28]. The disappearance of superconductivity in the pressure-induced antiferromagnetic phase implies that the $\boldsymbol{Q}_c$ spin fluctuations are suppressed by the antiferromagnetic ordering. Recent theoretical calculations also point to the chiral *d*-wave superconductivity produced by the antiferromagnetic fluctuations [29].

The ± signs in Eq. (1) stems from the degenerate nature of the two states under the tetragonal symmetry in the system. The two states are characterized by the phase winding directions of the order parameter in the *ab* plane as shown in Fig. 2. Recent lower critical field $H_{c1}(T)$ measurements detect some unusual anomaly below the superconducting transition [30]. From this observation, a possibility of the phase transition inside the superconducting state due to the degeneracy of the chiral *d*-wave has been pointed out. Namely, if the hidden order state breaks the fourfold rotational symmetry, the superconducting states with two different chiralities (the states with + and − sign in the Eq. (1) form) may have different transition temperatures. Although the origin of the $H_{c1}(T)$ anomaly should be scrutinized, this has triggered the search for the in-plane fourfold symmetry breaking in the hidden order phase.

2.2. Magnetic torque as a sensitive probe for the rotational symmetry breaking

Among experimental probes, the magnetic torque $\boldsymbol{\tau} = \mu_0\boldsymbol{M}V \times \boldsymbol{H}$ is a particularly sensitive probe for detecting the magnetic anisotropy, where V is a sample volume and $\boldsymbol{M}$ is the induced magnetization. The torque is also a thermodynamic quantity which is the angle derivative of free energy. The torque in tiny crystals can be measured by using the micro-tip cantilever [31,32], which has a sensitivity orders of magnitude larger than the commercial SQUID magnetometer. This technique has been widely used for the de Haas-van Alphen quantum oscillation measurements at high magnetic fields or for determination of the anisotropy parameter in layered superconductors. We use this technique to measure in-plane anisotropy of magnetic susceptibility in the hidden-order phase [33]. The torque measurements in magnetic fields $\boldsymbol{H} = (H\cos\phi, H\sin\phi, 0)$ rotating within the tetragonal *ab* plane in URu_2Si_2 (where ϕ is the angle from the [100] direction) provide a stringent test whether the hidden order parameter breaks the crystal fourfold symmetry. In such a geometry $\boldsymbol{\tau}(\phi,T,H)$ has a twofold oscillation with respect to ϕ-rotation:

$$\tau_{2\phi} = \frac{1}{2}\mu_0 H^2 V\left[(\chi_{aa}-\chi_{bb})\sin 2\phi - 2\chi_{ab}\cos 2\phi\right], \quad (2)$$

where the susceptibility tensor χ_{ij} is given by $M_i = \sum_j \chi_{ij} H_j$ [see Fig. 3a].

It should be stressed that in a system holding the tetragonal C_4 symmetry, $\tau_{2\phi}$ should be zero because $\chi_{aa} = \chi_{bb}$ and $\chi_{ab} = 0$. Even when we consider non-linear (higher order) susceptibility, the C_4 symmetry allows only fourfold oscillation with respect to ϕ-rotation, and cannot lead to the twofold component. Finite values of $\tau_{2\phi}$ may appear if a new electronic or magnetic state emerges that breaks the tetragonal symmetry. In such a case, rotational symmetry breaking is revealed by $\chi_{aa} \neq \chi_{bb}$ or $\chi_{ab} \neq 0$ depending on the direction of the orthorhombic anisotropy.

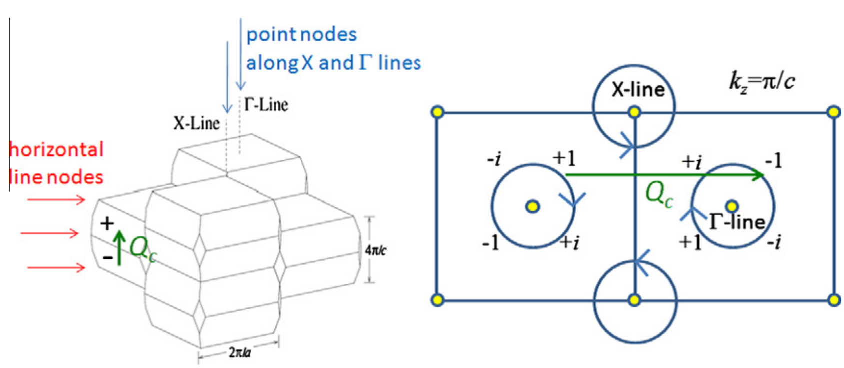

Fig. 2. Schematic arrangements of the Brillouin zone for the body-centered tetragonal structure of URu_2Si_2 (left). The superconducting order parameter with chiral *d*-wave symmetry described by Eq. (1) has horizontal nodes in the planes shown by the red arrows as well as point nodes along the X and Γ lines indicated by the blue arrows. A cross sectional cut at $k_z = \pi/c$ including two neighboring Brillouin zones is also depicted (right) with the phase winding directions (circular arrows) for one of the degenerate chiral *d*-wave order parameters. The other order parameter has the opposite phase winding directions in each position. At the two points connected by the antiferromagnetic wave vector $\boldsymbol{Q}_c$ (green arrows), which is (0,0,1) (left) or (1,0,0) (right), the superconducting order parameter changes sign. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)