Quantitatively determining the degree of spin fluctuations in actinide metals and compounds

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Abstract
Actinide materials are well-documented for nuclear properties, but their 5f electrons that produce exotic phenomena are not, due to the complexity of a dual localized and itinerant nature that remains a mystery. Particular interest is given to the electronic correlations at the localized and itinerant boundary where strong spin fluctuations are present. We report the identification of an intensity defined by integrating the normalized resistivity that approximately provides a quantitative measure of spin fluctuations. The intensity is very sensitive to the tuning of non-thermal parameters such as pressure and chemical doping, probing the anomalies in the evolution of spin fluctuations close to a valence or magnetic instability. In this way, our results are not only connected with the long-standing controversy on the anomalous low-temperature resistivity behavior of actinide metals, but also highlight an unconventional type of superconducting pairing, mediated by valence and/or spin fluctuations, for a wealth of 4f and 5f-electron systems. In an unified picture proposed, it is helpful to determine the degree of spin fluctuations for understanding the origin of the emergent superconductivity in systems with correlated electrons.

1. Introduction

Actinide materials are known for the instability of the 5f electron shell due to the near degeneracy of multiple electronic configurations. In the early series stretching from Th to Np, 5f electrons are itinerant and strongly participate in bonding similar to the 5d series. The size of the atoms decreases as the atomic number Z increases, reflecting the progressive filling of the bonding states. Then, in contrast, a large jump of the size occurs in the vicinity of Pu, and for heavier actinides (Am and beyond), 5f electrons are well localized. They do not contribute to bonding between neighboring atoms, resulting in little change in the size similar to the rare-earth series. The 5f electrons of Pu exactly settle at the brink between itinerancy and localization, making Pu the most complex element in the periodic table [1]. Notably, α-Pu shows an astoundingly large resistivity in elemental metals and exhibits a negative temperature coefficient where the resistivity increases as the temperature decreases [2]. On account of the electronic correlations that emerge from the competition of two opposing tendencies, δ-Pu has a Sommerfeld coefficient of the specific heat which is an order of magnitude larger than any other elemental metals [3]. As recently demonstrated by core-hole photoemission spectroscopy [4], resonant x-ray emission spectroscopy [5], and neutron spectroscopy [6], the ground state of Pu is governed by valence fluctuations connected with a quantum mechanical superposition of multiple localized and itinerant electronic configurations.

The conundrum of spin fluctuations in actinide materials was first proposed in Np and Pu to explain the anomalous behavior of the resistivity where the primary mechanism is the spin–flip scattering in narrow 5f bands [7]. Formally, the spin fluctuation spectrum is defined via the fluctuation dissipation theorem in terms of the imaginary part of the wave vector and frequency-dependent spin susceptibility, Im χ(q, ω). A measure of the wave vector average of Im χ(q, ω) can be provided by the NMR probe at ω equal to the NMR frequency, while the
linear coefficient of the specific heat, $\gamma$, depends on a complicated weighting of $\text{Im} \chi(q, \omega)$ over all $q$ and $\omega$ spaces, due to virtual emissions and reabsorptions of spin fluctuations. Something like this also applies for the quadratic coefficient of the resistivity, $\alpha$, which is often found to be proportional to the square of $\gamma (\alpha/\gamma^2$ being the Kadawaki-Woods ratio). Meanwhile, the physics of spin fluctuations driven via valence fluctuations was captured by the Kondo impurity problem [8], which is a special case of the Anderson impurity model [9]. In this model, the system is described by a magnetic impurity that interacts with the sea of conduction electrons. At high temperatures, 5f electrons are fairly localized, typically predicting a paramagnetic or magnetically ordered state. However, below the Kondo temperature, conduction electrons tend to align their spins antiparallel with localized magnetic moments that hence become partly or completely compensated [10]. Accordingly, the ground state appears as the formation with a reduced or cancelled magnetic moment. Through the hybridization with conduction electrons, 5f electrons effectively lead to the delocalization into the Fermi surface where heavy itinerant quasiparticles (so called the heavy fermions) form. In virtual valence fluctuations, 5f electrons are continuously hopping into and out of the Fermi sea via the quasiparticle resonance, identifying an electronic mechanism responsible for strong fluctuations of the configuration among different high and low spin states [11].

In this work, we report the discovery of a flavor in resistivity measurements which manifests as a measure of spin fluctuations for actinide materials. The intensity by integrating the normalized resistivity can accurately assess the enhancement of the resistivity at low temperatures, which is reminiscent of the Stoner enhancement factor, quantitatively determining the degree of spin fluctuations [12]. We show that by pressure or chemical doping, the intensity tends to diverge on approaching a critical point, no matter it refers to a valence or magnetic instability. In addition, our results provide an unified picture relevant to the emergence of superconductivity (SC) in typical heavy-fermion systems, highlighting an unconventional type of SC, mediated by valence and/or spin fluctuations [13]. In the following, the integrated intensity of actinide metals and compounds will be shown to illustrate the previously neglected yet substantial fundamentals for spin fluctuations.

2. Experiments

The resistivity was measured using a Quantum Design Physical Property Measurement System (PPMS). A standard four wire AC technique was used, and the data were recorded on warming, in the temperature range from 2 K to 300 K. The resistivity was normalized, relative to the value at 300 K, and then the data were integrated to obtain an accumulated intensity. To be a dimensionless number, similar to 1, the intensity was divided by 298 K, the temperature window. For simplification, the intensity ($I$) is expressed as follows,

$$I = \frac{1}{298 \text{ K}} \int_{2 \text{ K}}^{300 \text{ K}} \frac{\rho(T)}{\rho(300 \text{ K})} dT.$$

For common metals, $I$ is near 0.5. Nevertheless, a higher $I$ is expected for materials with strong spin fluctuations, due to the increase of the low-temperature resistivity. For high-precision resistivity data, the relative error of $I$ is smaller than the data point in the figures. In resistivity measurements at high pressures, the pressure was applied using a BeCu cell with Daphne7474 as the pressure-transmitting medium, and was then determined by the superconducting transition temperature ($T_{SC}$) of Pb. Owing to the actual difficulties in handling of actinide materials, the data for neptunium and Plutonium compounds were reproduced from the published literatures.

3. Results and discussions

An example of our resistivity measurements is presented in figure 1. The resistivity was measured on a high-quality UGe$_2$ single crystal (RRR $\approx 430$) with the configuration that $l/a$. We know that the ferromagnet UGe$_2$ crystallizes in the ZrGa$_2$-type orthorhombic structure [14], and is a prototype to exhibit an unconventional SC in a limited pressure range on the border of ferromagnetism (FM) [15]. At ambient pressure, UGe$_2$ has a rather high Curie temperature, $T_C \approx 52$ K, with a large magnetic moment, $\sim 1.5 \mu_B$. The FM order collapses at a critical point, $P_C \approx 1.5$ GPa [16], while the SC occurs within the FM region, in a pressure range from 1.0 GPa to 1.5 GPa [15]. In the FM phase, an anomalous crossover ($T_X \approx 30$ K at ambient pressure) deserves attention, which denotes the boundary between two different states, namely weakly polarized FM1 and strongly polarized FM2 [17]. Hitherto, despite the consensus that the SC is closely related to the vanishment of $T_X$ at a critical point, $P_X \approx 1.2$ GPa, where $T_{SC}$ just shows a maximum, $\sim 0.8$ K, the understanding of the coexistence between FM and SC remains a matter of intense debate [18–25].

Here we show the pressure evolution of the intensity by integrating the normalized resistivity of UGe$_2$, mostly in the vicinity of $P_X$. The subtle change of the intensity is mainly ascribed to the modification of spin fluctuations. It is noted that a sharp discontinuity occurs at $P_X$, in consistency with the anomaly observed in the
pressure dependence of the spin fluctuation spectrum [26]. In this manner, our results clearly bridge the intensity with the degree of spin fluctuations, and further display an intrinsic connection between SC and pressure-enhanced fluctuations developed at PX. Analogously, an anomaly at around PC is seen, but is rather weak.

Another science challenge on 5f electrons is the quest to understand the mysterious hidden-order (HO) phase of URu2Si2 [26]. The HO phase occurs below 17.5 K and coexists with an unconventional SC below ~1.5 K. In the HO phase, neutron scattering experiments reveal a small antiferromagnetic moment, about 0.03 μB/U, parallel to the tetragonal c axis [27], which is too small to account for the entropy of ~0.2Rln(2) in the specific heat jump [28]. The small moment is thought to be extrinsic due to the presence of antiferromagnetic regions induced by inhomogeneous strain [29]. Recently, since the HO phase exists in proximity to a pressure-induced large-moment antiferromagnetism (LMAFM), it is believed that a comprehensive understanding of both phases is crucial in unraveling the nature of the HO phase [30]. The boundary between two phases is a first-order phase transition at a critical pressure that lies in the range 0.5–1.5 GPa [31–34]. Although presumable order parameters must be different, transport and thermodynamic properties for these two phases are nearly indistinguishable.

In figure 2, the HO phase has been modified by chemical doping, providing an opportunity to probe HO and LMAFM phases simultaneously at ambient pressure. The substitution of smaller Fe ions for Ru ions acts as a chemical pressure, well reproducing general features of both phases [35–41]. As the doping level increases, the hump of the normalized resistivity drops and shifts to higher temperatures, suggesting a gradual weakening of spin fluctuations. Consequently, the integrated intensity shows a monotonous decrease. We note that the
tendency of the intensity is reminiscent of $T_{SC}$ in the phase diagram. Although the nature of the HO phase remains hidden, it is clear that the SC in the HO phase is ultimately mediated by spin fluctuations close to a critical point at hypothetical negative pressures.

Having established the route to quantitatively determine the degree of spin fluctuations, it is necessary to take into account the feasibility for other actinide metals and compounds. The prior condition is an excellent metallic behavior of the resistivity, including superconductors, at least at extremely low temperatures. Secondly, because of the anisotropy in resistivity measurements, the direction along the first nearest neighbor of the actinide atoms is preferred. In some senses, it is an alternative to perform measurements on polycrystalline samples. In superconducting ferromagnets $\text{UGe}_2$ [15], $\text{URhGe}$ [42], and $\text{UCoGe}$ [43], uranium atoms form zigzag chains along the a axis of the orthorhombic structure for special circumstances [44]. Thirdly, we comment on resistivity measurements in a magnetic field, generally termed as magnetoresistance. Contrary to pressure or chemical doping, a magnetic field can be varied continuously, and thus the data of magnetoresistance are usually measured as a function of magnetic field, instead of temperature. In high magnetic fields above 16 T, treating the temperature dependence of magnetoresistance is seldom used. On the other hand, the analysis of magnetoresistance is difficult when the materials are undergoing a field-induced quantum criticality [45] or metamagnetic transitions [46]. For example, a $T_{SC}$ of 1.6 K is reported in the recently discovered spin-triplet superconductor $\text{UTE}_2$, which has a large and anisotropic upper critical field exceeding 40 T [47]. For $H/\lambda$ a peculiarity is the hump-like maximum of the magnetic susceptibility at around 35 K [48], corresponding to a metamagnetic transition at $H_{m} \approx 35$ T, as evidenced by a sharp increase of the resistivity at 1.4 K in the normal state [49]. At extremely low temperatures, the SC in $\text{UTE}_2$ survives at least up to the metamagnetic transition, showing a reinforcement of the SC on approaching $H_{m}$ [50]. Similar behavior has been observed in many heavy-fermion materials, where $H_{m}$ is linked with the temperature at the maximum of the magnetic susceptibility by a correspondence 1 T $\leftrightarrow$ 1 K. Although the origin of the energy scale is unclear, these phenomena are likely to point to the contribution of spin fluctuations near $H_{m}$ which are markedly enhanced by the magnetic field.

Finally, the integrated intensity (I) of the normalized resistivity for actinide materials is summarized, as shown in figure 3. The data used for the calculations of U, Np, $\alpha$-Pu, and Am are reproduced from the reference [2]. Their intensities are 0.53, 0.64, 0.91, and 0.57, respectively. For $\alpha$-Pu, a high value of $I \approx 0.91$ indicates that strong spin fluctuations are present. Interestingly, the intensities for many heavy-fermion superconductors are comparable with or even higher than that of $\alpha$-Pu. For clarity of an electronic origin, the intensity of typical heavy-fermion superconductors is plotted versus the characteristic temperature of spin fluctuations, $T_{0}$, which is the width of the spin fluctuation spectrum in the energy space [51]. Despite a few exceptions, the observation of a systematic study stimulates us to propose an unified picture to understand the role of spin fluctuations for the appearance of SC in heavy-fermion systems, including cerium-based compounds likewise. It is probable that a certain degree of spin fluctuations, similar to $\alpha$-Pu, should be essential for the SC. And as $T_{0}$ increases, the intensity favoring the SC shows a moderate decrease, implying an easier occurrence of the SC. It is known that a high $T_{0}$ generally results in a high $T_{SC}$ in unconventional superconductors. In cuprates for instance, $T_{0}$ is much

![Figure 3. Comparison of the integrated intensity with the characteristic temperature, $T_0$. The latter is essentially a bandwidth, i.e., the energy spread of the spin fluctuation spectrum, which are reproduced from published data. The solid line schematically shows the role of spin fluctuations for the appearance of superconductivity in typical heavy-fermion systems. The dashed line shows the intensity of $\alpha$-Pu for comparison.](image)
higher than 100 K, and as a consequence, the intensity is relatively small, with almost no hump detected in resistivity measurements.

We now turn to the interest revived in Plutonium-based heavy-fermion systems [52], in which PuCoGa5 shows an unconventional SC below $T_{SC} \approx 18.5$ K [53], the highest in 4f and 5f-electron systems so far. Surprisingly, the intensity for PuCoGa5 is 0.63, much smaller than expected. It is thus interesting to compare PuMGa5 and PuMIn5 ($M = \text{Co, Rh}$) where the remarkable difference suggests different origins for these two series of superconductors [54]. While the antiferromagnetism (AFM) has been found close to PuRhIn5 with cadmium doping, various chemical substitutions in PuCoGa5 did not induce any magnetic ordering [55]. In this scenario, the PuMIn5 superconductors are mediated by AFM fluctuations, comprising a small dome of SC, whereas the PuMGa5 superconductors should reside on a large dome of SC mediated by valence fluctuations. For the SC mediated by valence fluctuations, a high degree of spin fluctuations may be not necessary. Here, referring to a valence (or charge density) transition, a famous case is CeCu2Si2, which has two pressure-induced domes of SC. They are a lower $T_{SC}$ dome attributed to spin fluctuations and a higher $T_{SC}$ dome attributed to valence fluctuations [56]. Although the physics remains elusive, valence fluctuations seem to carry spin fluctuations that have a comparably large characteristic energy. Ongoing inelastic neutron scattering on $^{242}\text{PuCoGa}_5$ single crystals would be helpful for explicitly determining the fluctuations with their momentum dependencies and energy scales. Moreover, we note that the linear coefficient of the specific heat (a measure of the hybridization) in $\alpha$-Pu is rather low (16–25 mJ mol$^{-1}$ K$^2$). This indicates that a high strength of the hybridization is not necessary for materials with strong spin fluctuations.

4. Conclusion

The discovery of our resistivity measurements provides a new route to quantitatively determine the degree of spin fluctuations in actinide metals and compounds. The intensity by integrating the normalized resistivity is straightforward in comparison with the analysis of the resistivity, $\rho = \rho_0 + AT^n$, where $\rho_0$ is the residual resistivity and $AT^n$ is the scattering term. For the electron-electron scattering, $n = 2$. Apparently, the analysis of $AT^n$ is complicated, but is simplified by the intensity. Since $A$ depends on spin fluctuations over wide ranges of $q$ and $\omega$, the resistivity at any given temperature is governed by the thermally activated spin fluctuations in the frequency range below a scale of the temperature. Thus, our investigations are relevant to the anomalous low-temperature resistivity behavior, opening the door to advance the research area of correlated matter with strong spin fluctuations. As intrinsically connected with an unconventional type of superconducting pairing, the integrated intensity is very helpful in classifying novel superconductors, especially near a valence or magnetic instability. The puzzle on valence and spin fluctuations is an open question, which deserves special attention in future studies, for both lanthanide and actinide materials.

5. Additional proof

The intensity for UPt3 is 0.81, indicating that the role of spin fluctuations can not be the dominating one for the SC. Although UPt3 is unambiguously an unconventional superconductor with a multicomponent superconducting order parameter, the state above $T_{SC} \approx 0.5$ K is a Fermi liquid with strong mass renormalization where the usual BCS theory should hold. As stated in the reviewed literature, the coupling between SC and AFM ($T_N \approx 5$ K) in UPt3 may be the weakest in the current picture [57].

In a recent report, the coexistence of SC and FM in UGe2 is described in terms of the two-fluid model, i.e., heavy itinerant quasiparticles coexisting with residual unhybridized localized moments [58]. The pressure evolution of the two fluids shows a sudden delocalization of part of 5f electrons at $P_N$, indicating that strong valence fluctuations should play a central role for the SC [59]. FM fluctuations near $P_C$ also exist, but are relatively weak. The intensity of UGe2 is 0.83, which shows no much change at high pressures, favoring the picture as mentioned above.

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