Electronic states of URu_2Si_2 studied using high-quality single crystal growth and high-pressure techniques

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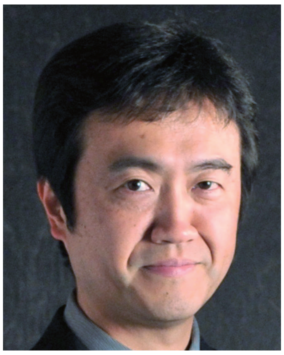

- **Characteristics of the hidden-order state and superconductivity of URu_2Si_2 have been investigated on a high-quality single crystal using high-pressure techniques.**
- **We have found that the resistivity in the hidden order state has an anomalous temperature exponent, and it is closely related to the superconducting transition temperature.**

Abstract

A uranium compound URu_2Si_2 which is known as a heavy fermion system shows a phase transition at 17.5 K. However the origin of the order parameter of this phase transition has not been clarified since its discovery in 1985, despite the extensive study. It is therefore called "hidden order" state. In the present study, we have found that the resistivity of high quality single crystal shows an anomalous behavior in the hidden order state of URu_2Si_2. It is also clarified that anomalous resistivity is closely related to the superconducting transition temperature.

In this study, Drs. T. D. Matsuda and N. Tateiwa have played major roles in the characterization of quality dependence of physical properties and high-pressure study, respectively. The results have been published in J. Phys. Soc. Jpn [1]. and Phys. Rev. B [2].

1. Introduction

Actinides, including uranium and plutonium, are the important elements in atomic energy science. They are also known as elements with quite different behavior compared to other elements. One of the prominent examples is the multiple structural transformation of plutonium metal. Plutonium has the usual highly symmetric cubic crystal structure just below the melting temperature, in a similar way to that often observed in conventional metals. However, the structure changes with decreasing temperature, showing 6 different types of structures. Finally at room temperature it transforms in the anisotropic monoclinic structure. This behavior is mainly dominated by the electrons in the 5f shell with a large degree of freedom. A structural transition to a low symmetry generally lifts degeneracy of 5f states resulting in a stable ground state with the lowest energy. However, when atoms are confined in a crystal lattice, electrons would try by themselves to realize a stable ordered state. Typical examples are unconventional superconductivity and heavy fermion formation often observed in actinide compounds, where a strong electron correlation – one of the major subjects in solid state physics – plays an important role. A uranium compound URu_2Si_2 uses two phase transitions occurring at 1.4 K and 17.5 K to reduce the energy of its electrons.

The first one is known as the superconducting transition. However, the origin of the latter one has not been clarified since its discovery in 1985, despite the extensive investigations by scientists in the world. We approached to this problem by using a single crystal with an extremely high quality.

2. Experimental

URu_2Si_2 has the tetragonal $ThCr_2Si_2$-type crystal structure. Two phase transitions were found at 17.5 K and around 1 K as demonstrated in anomalies in specific heat as shown in Fig. 1 [3]. It was clarified that the phase transition at 1 K corresponds to the

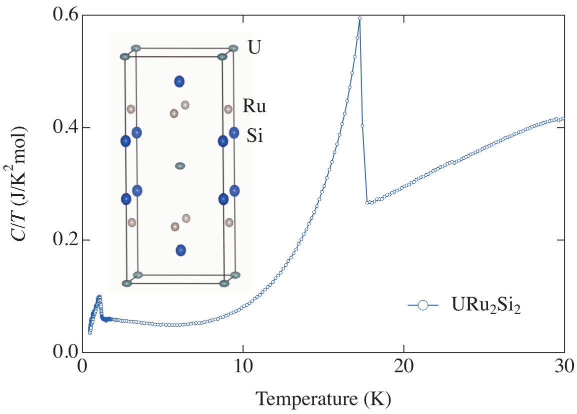

Fig.1. Crystal structure and temperature dependence of specific heat of URu_2Si_2

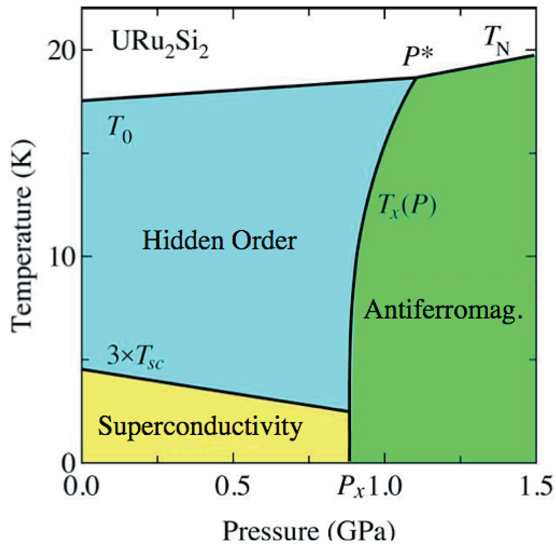

Fig. 3. Pressure phase diagram of URu_2Si_2

superconducting transition. However the order parameter which characterizes this transition at 17.5 K has not been detected.

It is also known that the resistivity behavior in the ordered state generally reflects the nature of order parameter. For example, lattice vibration gives a contribution proportional to T^5 (T: temperature), while the strong electron-electron scattering gives T^2. Although electrical resistivity can be easily measured at any laboratories, the impurities in the sample cause large residual resistivity and often make detailed investigation impossible. It is therefore very important to prepare a high quality sample with low residual resistivity.

Our idea to approach the hidden-order problem is to investigate electrical resistivity in detail. For this purpose we performed high quality single crystal growth and resistivity measurement at low temperatures and at high pressures.

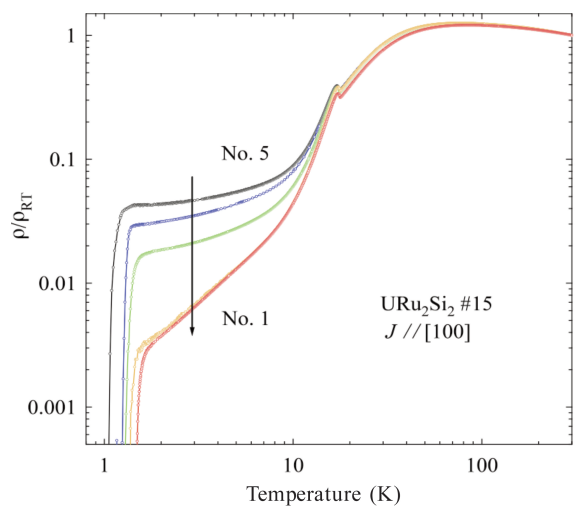

Fig. 2. Temperature dependence of electrical resistivity of URu_2Si_2 single crystals with different residual resistivities. The low temperature behavior as well as the superconducting transition temperature strongly depend on the crystal quality.

3. Results

Single crystals of URu_2Si_2 have been prepared using a purification of uranium metal using Solid-State Electrotransport (SSE), Czochralski pulling method and subsequent annealing of the crystal under ultrahigh vacuum [1, 4]. As a result we have succeeded in preparing a crystal with the lowest residual resistivity. The electrical resistivities measured on this sample as well as those with different quality are shown in Fig. 2. Resistivity behavior of the best sample (No. 1) differs from the sample with larger residual resistivity. Here, residual resistivity is defined as the resistivity just above the superconducting transition at low temperature.

The sample with poor quality (No.5) behaves as T^2 as observed in other actinide compounds in general. However, the resistivity of No. 1 sample significantly differs from it. As mentioned in Introduction, resistivity behavior reflects the electron scattering in the ordered state. The present observation is therefore expected to provide important information on the nature of

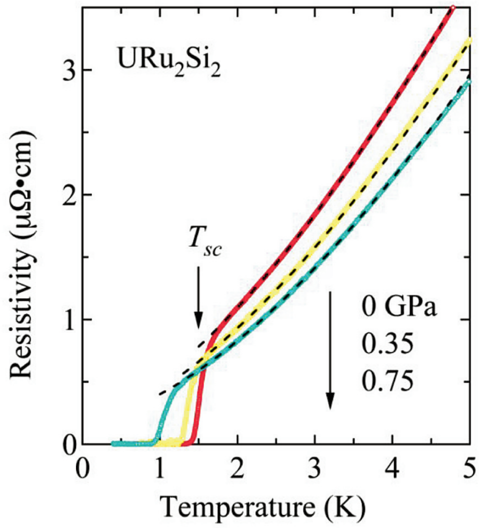

Fig. 4. Electrical resistivity of URu_2Si_2 under high-pressure

hidden-order.

We further extend this investigation under high pressure. Figure 3 shows the pressure dependence of the transition temperatures of both superconductivity and hidden-order. With increasing pressure, the transition temperature of hidden-order (T_0) and that of superconductivity (T_{sc}) change gradually. However, they are replaced by an antiferromagnetic ordering above a critical pressure P_x. Resistivity measurement under pressure would give a change in the electron scattering in different states.

Figure 4 shows the resistivity behavior under constant hydrostatic pressures. With increasing pressure, resistivity behavior above T_{sc} changes. We found that the temperature dependence of resistivity can be decomposed in two parts: one corresponds to a 'normal' scattering proportional to T^2 due to electron-electron scattering and another one corresponds to 'anomalous' component proportional to T. Note also that at low temperatures other contributions to resistivity such as lattice vibrations can be neglected. The total resistivity can be therefore expressed as follows:

$$\rho = \rho_0 + \alpha_1 T + \alpha_2 T^2 \qquad (1)$$

The pressure dependence of the contribution of each term is plotted in Fig. 5. 'Anomalous' component α_1 decreases with increasing pressure and discontinuously decreases at the critical pressure. α_1 in the high-pressure antiferromagnetic phase is still finite. This is most likely due to the remaining hidden-order component existing above the first-order transition line, as suggested in other measurements. High pressure data at 1.51 GPa shows $\alpha_1 = 0$, suggesting that there is no remaining hidden-order state. From this we concluded that the anomalous scattering α_1 only exists in the hidden-order state. On the other hand, the 'normal' contribution α_2 is almost temperature independent. It means that electronic state in both hidden-order and antiferromagnetic states are similar. In fact, Fermi surfaces obtained in both phases agree fairly well [5,6], in consistent with our observation.

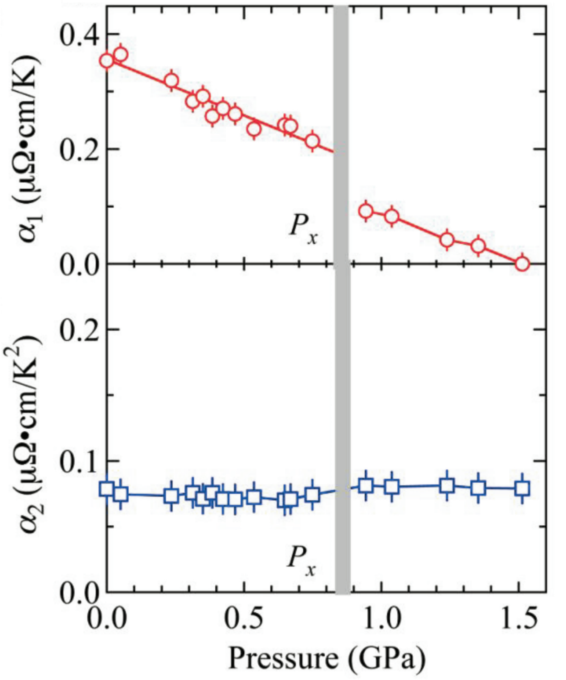

Fig. 5. Pressure dependence of the coefficient α_1 and α_2

We also note that the magnitude of the anomalous resistivity (α_1) changes as a function of pressure. Furthermore it was found that α_1/α_2 is closely related to the superconducting transition temperature, as shown in Fig. 6. As mentioned above the normal component α_2 is almost pressure independent. The present result therefore demonstates that α_1 is proportional to T_{sc}. This observation strongly suggests that the anomalous resistivity only existing in the hidden-order phase drives heavy fermion superconductivity at lower temperature.

4. Concluding remarks

We studied the long-standing problem in URu_2Si_2 using a extremely high quality single crystal growth and high-pressure techniques. As

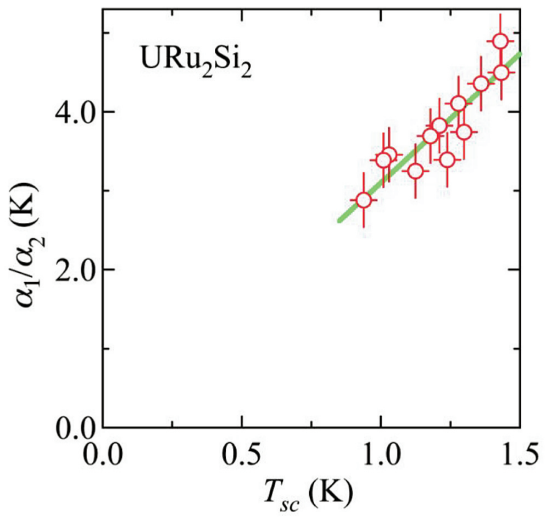

Fig. 6. Relationship between the ratio of anomalous to normal resistivity and superconducting transition temperature

a result, hidden-order state causes an anomalous scattering on conduction electrons and furthermore it is intimately related to the heavy fermion superconductivity in this compound.

Although the nature of the hidden-order is still unclear, the present observation indicates that the anomalous scattering is one of the very important properties characterizing the 'hidden-order' state.

The present anomalous resistivity coefficient α_1 can be found in other correlated electrons systems such as high-temperature superconducting cuprates and heavy fermion systems. On the other hand, an ordered phase which does no accompany an explicit phase transitions often observed in those compounds. One of the prominent examples is so-called pseudo-gap phase of high-temperature superconductors. 'Hidden-order' state and other mysterious ordering might be a general characteristics of correlated electron systems.

Our high-quality single crystal stimulates scientists developing new novel measurement techniques[5-7]. We expect a new findings concerning the hidden-order using these techniques.

5. Future prospects

Understanding unusual electronics states realized in strongly correlated electron systems is one of the important subjects in solid state science. Actinides' 5f electron systems placed under unexplored various conditions such as chemical environments and extreme physical conditions would provide a new novel electronic state. Finding and understanding such new state of matter give us new descriptions of electronic states and it can be applied to other fields of science. Our aim is therefore to explore actinide materials using various techniques.

References

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