Research Group for Actinide Materials Science

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Various phase transitions observed in actinide compounds originate from their 5*f* electrons. Because of the spatial extent of the wave function, 5*f* electron behaviour is extremely sensitive to the physical/chemical environment and can be manipulated with varying experimental parameters such as pressure, magnetic field or low temperature to find new phenomena. Some of the interesting properties are particularly sensitive to small amounts of impurities and defects. In this context, sample preparation with high perfection provides new insight particularly for phenomena occurring at low temperatures, where impurity disorder can deeply disturb and hence hide the intrinsic behavior. We use various experimental techniques to purify the actinide compounds. Here we report recent results on the electronic structure investigations on a prototypical actinide compound.

High-quality single crystal growth of URu_2Si_2 and $ThRu_2Si_2$

URu_2Si_2 is well known as a heavy fermion superconductor coexisting with another phase transition called 'hidden-order'. Recent developments in high-quality single crystal growth enabled various new investigations related to its novel properties [1]. One of the prominent examples is growing evidence on orthorhombic distortion occurring in the hidden-order phase. In addition to the magnetic anisotropy reported previously [2], a structural deformation was detected in a high-resolution synchrotron X-ray diffraction experiment [3]. Using the microscopic probe NMR, a precise line-width measurement of a ^{29}Si-NMR line strongly indicated orthorhombic anisotropy [4]. Further, new insight into this compound's unconventional superconductivity was discovered by the observation in huge Nernst signal in the normal state which is extremely sensitive to the sample quality [5].

We have extended our study to isostructural $ThRu_2Si_2$ where Th does not have a 5f electron and it is therefore expected to provide underlying information on electronic and lattice structures. We have succeeded in growing a high-quality single crystal of $ThRu_2Si_2$ using in-house purified Th metal and subsequent electrotransport annealing under ultra-high-vacuum conditions [6]. This sample enabled detailed investigations of its electronic structure using quantum de Haas-van Alphen (dHvA) oscillations. Figure Fig. 1 shows a prototypic experimental signal. The dHvA frequency is proportional to the Fermi surface cross sectional area perpendicular to the applied magnetic field. The topology of the Fermi surfaces can therefore be obtained from dHvA frequencies as a function of field angle. The obtained Fermi surfaces of $ThRu_2Si_2$ differed from those of URu_2Si_2, indicating that 5f electrons in URu_2Si_2 participate in conduction bands to modify the size of Fermi surfaces. On the other hand, the Fermi surface of $ThRu_2Si_2$ can be well explained by electronic structure calculations based on a Th^{4+} configuration. The present observation suggests then that the conventional assumption of a localized U^{4+} ($5f^2$) configuration is not valid in URu_2Si_2. Interestingly, the Fermi surface characteristics of $ThRu_2Si_2$ are very similar to those of the isostructural prototypical heavy fermion $CeRu_2Si_2$. This implies that the total conduction electron count for both compounds is the same. This result demonstrates that the highly correlated 4f electron of $CeRu_2Si_2$ participates in the Fermi surface at low temperatures and contributes to Fermi surfaces as do electrons in ordinary metals such as $ThRu_2Si_2$ and reinforces the previous observations [7].

Further experiments are planned to clarify electronic evolutions from $ThRu_2Si_2$ to URu_2Si_2 by gradually replacing Th by U to introduce 5f electrons in the system.

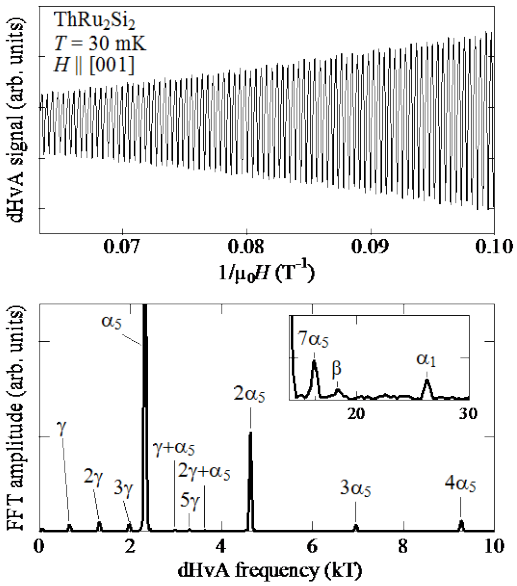

Fig. 1 Typical dHvA oscillation and its Fourier transform recorded for a $ThRu_2Si_2$ single crystal. dHvA frequency is proportional to the Fermi surface cross section perpendicular to the applied magnetic field. Inset of the bottom panel magnifies high frequency region. In addition to fundamental branches α_i, β and γ, their harmonics are observed, which usually occur in high quality samples.

References

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