Effect of uranium defect in unconventional superconductor UTe_2

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Superconductivity is a fairly common phenomenon occurring in metals at low temperatures. However, in some class of metals, the superconducting properties look different from ordinary superconductors. Examples of such superconductors include the so-called high-temperature cupurates where superconductivity takes place above 100 K. On the other hand, actinide and lanthanide compounds often exhibit unconventional superconductivity. Unlike ordinary metals, conduction electrons in actinide or lanthanide compounds often acquire large effective mass originated from strong electron correlation. As a result, superconductivity occurring in these compounds is called "heavy fermion" superconductivity.

UTe_2, discovered in 2019, is the most recent member of heavy fermion superconductors [1]. Its superconductivity arises from enhanced electron mass due to many-body interaction among conduction carriers. Crystal structure of UTe_2 is shown in Fig. 1(a). Uranium ladders decorated with tellurium are elongated along the *a*-axis. Reflecting the orthorhombic symmetry, superconducting upper critical field is highly anisotropic. In particular, unusual reinforcement of superconductivity occurs under magnetic field along the *b*-axis, until a field-induced magnetic phase transition takes place at higher field. Theoretical suggestions for possible use of the peculiar superconductivity for quantum computing make this compound important even in the context of practical applications.

It is noticed that the superconductivity in UTe_2 is extremely sensitive to the sample quality. The first report on the superconducting sample showed the superconducting transition temperature T_c of 1.5 K. However, some of the subsequent reports showed higher T_c. In general, residual resistivity at low temperature is a measure of 'quality' of metals because resistivity appears through electron scattering with impurities which break crystal periodicity. For elucidating the superconducting properties, it is crucial to prepare the high-quality samples. To do this we first characterized the origin of impurities [2].

We employed two conventional methods. The first one is the single crystal x-ray diffraction where crystallographic parameters including the occupancy for each crystallographic site can be determined. In addition, chemical composition as well as the possible spatial distribution of is determined semi-quantitatively using electron-probe microanalyzer.

Figure 1(b) shows the comparison of the x-ray diffraction intensity (I_{obs}) and calculated values (I_{calc}) from the crystal structure model. When the model is correct, all points should fall on the solid line corresponding to $I_{obs} = I_{calc}$. As seen in Fig. 1(b) left panel, agreement between I_{obs} and I_{calc} is poorer for NS sample. In Fig. 1(b), the points significantly deviating from the calculation are highlighted. It turned out that those reflections consist of large contributions from uranium and tellurium sites but canceling each other. Such reflections are sensitive to the occupancy, in particular, of the uranium site in the present case. Figure 1(b) right panel shows the result of optimization of the uranium site occupancy x. I_{obs} can well be reproduced with uranium deficiency model $x = 0.962(2)$. In the superconducting samples, on the other hand, the similar analysis results in full occupancy on both uranium and tellurium sites within an

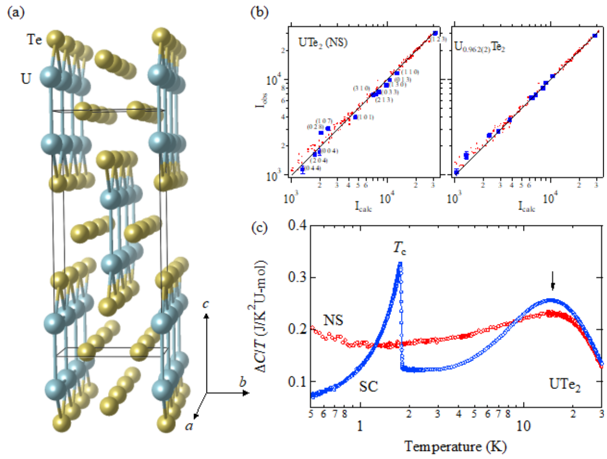

Fig. 1 (a) Crystal structure of UTe_2. (b) Results of x-ray diffraction on non-superconducting sample of UTe_2. Uranium deficiency model well describes experimental data. (c) Temperature dependence of electronic specific heat of superconducting (SC) and non-superconducting (NS) samples of UTe_2.

experimental accuracy. Examining crystallographic parameters as well as chemical composition, the uranium deficiency is not substituted by tellurium but rather left vacant.

Figure 1(c) shows the comparison of the electronic contribution to specific heat for both SC and NS samples. While a clear anomaly is observed at the superconducting transition temperature (T_c = 1.8 K) in the SC sample, superconductivity is not seen in the NS sample down to the lowest temperature investigated. At higher temperatures, different behavior persists even in the normal state. While the SC shows a marked enhancement of specific heat at 15 K shown by an arrow, the NS sample only shows a broad feature at the corresponding temperature. The peak at 15 K is generally interpreted as an energy gap in electronic states. The corresponding anomaly is also seen in the nuclear relaxation rate [3]. The present comparison suggests that this energy gap is not completely formed in the NS sample, leaving a large electronic specific heat at low temperatures, which might hinder the superconducting transition. The sensitivity of electronic state on the uranium defects is consistent with the one-dimensional magnetic correlation within the uranium ladder reported previously; the 4 % vacancy breaks every 13 ladder rungs.

Having established the origin of the defect, improvements on the crystal growth condition have immediately been made. Investigations using crystals with higher quality, namely with low residual resistivity, are underway.

References

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