Thermal expansion of UCu_2Sn in the basal plane

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Abstract

We reported from elastic moduli experiments that the hexagonal uranium compound UCu_2Sn undergoes a ferroquadrupolar ordering of non-Kramers doublet Γ_5 at $T_Q = 16$ K. However, a macroscopic strain has not been measured by an X-ray diffraction experiment because the strain is expected to be smaller than the experimental resolution due to the weak coupling between the strain and order parameters. To detect the spontaneous strain, we have carried out a thermal expansion measurement by a capacitance method with higher resolution from 4.2 to 40 K. The thermal expansion in the basal plane shows a remarkable change below T_Q, suggesting the emergence of the ε_{Γ_5} strain which couples to the ground state Γ_5.

Key words: UCu_2Sn; quadrupolar ordering; thermal expansion

1. Introduction

Ternary uranium compound UCu_2Sn has a hexagonal $ZrPt_2Al$-type structure (space group : $P6_3/mmc$) with a single U site . All constituent atoms are stacked in layers perpendicular to the hexagonal c-axis with a periodic sequence of Sn, Cu, U and Cu [1]. Lattice parameters of UCu_2Sn are $a = 4.457$ Å and $c = 8.713$ Å at room temperature. Recently, we reported that elastic modulus C_{66} shows a large softening with more than 57 % reduction of the stiffness at T_Q ($= 16$ K) [2]. The stiffness C_{66} is the liner response to ε_{Γ_5} ($= \varepsilon_{xy}$ and $= \varepsilon_{xx} - \varepsilon_{yy}$) strain in the hexagonal symmetry, where Γ_5 is the irreducible representation for the $6/mmm$ point group. The elastic anomaly originates from the ferroquadrupolar ordering of non-Kramers doublet Γ_5 which is the ground state of $5f^2$ in the crystal electric field of UCu_2Sn. In fact, the quadrupole-quadrupole (q-q) coupling constant of Γ_5, g'_{Γ_5}, was positive by our analysis of elastic moduli considering the strain-quadrupole coupling and the q-q coupling. For the ferroquadrupolar ordering, the spontaneous strain should emerge macroscopically below T_Q. To observe the spontaneous strain, we previously carried out the X-ray diffraction experiment. However, no indication of the macroscopic strain was observed within our experimental resolution. We numericaly calculated the magnitude of strain taking account of the strain-quadrupole coupling constant of Γ_5, g_{Γ_5} ($\simeq 8.6$ K), in the analysis of elastic modulus [3]. The evaluated value ($\simeq 5.6 \times 10^{-4}$) was smaller than the experimental resolution ($\simeq 1 \times 10^{-3}$). To detect such a small spontaneous strain, we have carried out the thermal expansion measurement using the sensitive three-terminal capacitance method with higher resolution ($\simeq 1 \times 10^{-8}$) in the present work.

2. Experimental

A single crystal of UCu_2Sn was grown by a Bridgman method [4]. The detail of sample growth was reported in Ref.4. Electron Probe Micro Analysis for the UCu_2Sn single crystal detected an impurity phase of

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UCuSn at ~ 4 % of the total volume. A size of sample, which was used on our measurements, was $2.824 \times 2.908 \times 3.288$ mm^3. Thermal expansion $\Delta l/l$ was measured as a function of temperature T from 4.2 to 40 K with 0.1 K temerature interval along a- and b-axes. Measurements were carried out by means of a parallel-plate capacitance method with 0.1 mm space between the fixed plate and the movable plate. The fixed plate and the movable plate have an area of $\simeq 1.55 \times 10^2$ mm^2. The b-axis is defined as perpendicular to the a-axis in the hexagonal c-plane.

3. Results & Discussion

Fig.1 and Fig.2 show the temperature dependence of thermal expansion $\Delta l/l$ along a- and b-axes, respectively. We have normalized the value of $\Delta l/l$ at 40 K. Above $T_{\rm Q}$, $\Delta l/l$ decreases monotonically with decreasing temperature along a- and b-axes. Along the a-axis, $\Delta l/l$ increases with decreasing temperature below $T_{\rm Q}$. Meanwhile, $\Delta l/l$ along the b-axis decreases with decreasing temperature below $T_{\rm Q}$. In a hexagonal symmetry, $\Delta l/l$ along a- and b-axes are expected to show the same behavior but the results of $\Delta l/l$ along a- and b-axes show the opposite behavior each other below $T_{\rm Q}$, demonstrateing the emergence of the spontaneous $\varepsilon_{xx} - \varepsilon_{yy}$ strain which is one of the ε_{Γ_5} strains. We have found the macroscopic spontaneous strain due to the ferroquadrupolar ordering of Γ_5 in UCu_2Sn for the first time. The thermal expansion $\Delta l/l$ along b-axis separates gradually from $\Delta l/l$ along a-axis below ~ 20 K. The elastic modulus C_{66} starts to soften from the same temperature region.

4. Conclusion

We have measured the thermal expansion $\Delta l/l$ along a- and b-axes by the capacitance method from 4.2 to 40 K. The result shows the emergence of the ε_{Γ_5} strain which originates from the ferroquadrupolar ordering of the ground state non-Kramers doublet Γ_5.

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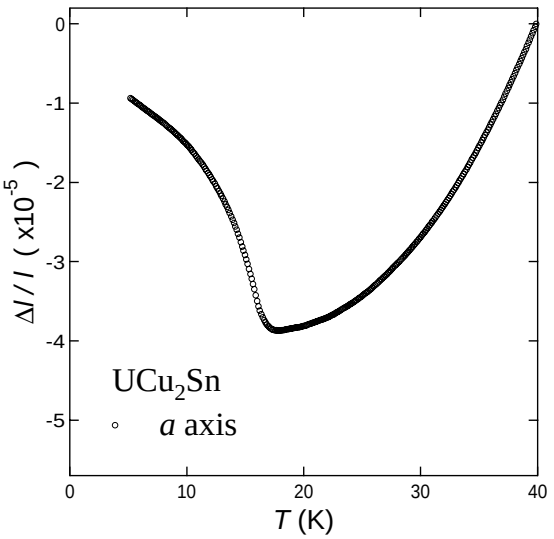

Fig. 1. Temperature dependence of thermal expansion $\Delta l/l$ along a-axis.

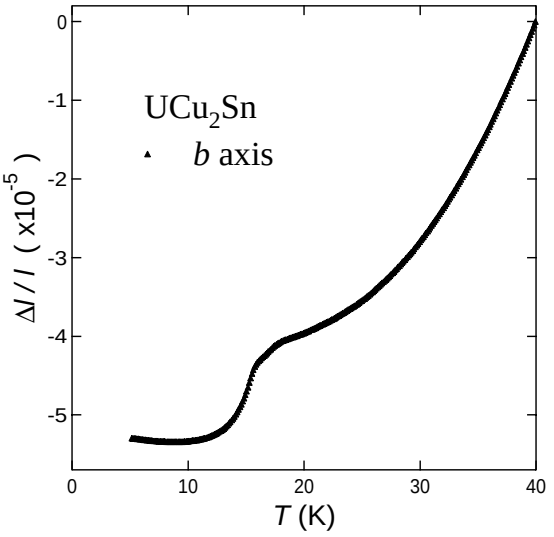

Fig. 2. Temperature dependence of thermal expansion $\Delta l/l$ along b-axis.

References

[1] T.Takabatake, H.Iwasaki, H.Fujii, S.Ikeda, S.Nishigori, Y.Aoki, T.Suzuki, T Fujita, J. Phys. Soc. Jpn. **61** (1992) 778.

[2] T.Suzuki, I.Ishii, N.Okuda, K.Katoh, T.Takabatake, T.Fujita, Phys. Rev. B **62** (2000) 49.

[3] B. Lüthi, in *Dynamical Properties of Solids* edited G. K. Horton and A. A. Maradudin (NorthHolland, Amsterdam 1980).

[4] T.Takabatake, M.Shirase, K.Katoh, Y.Echizen, K.Sugiyama, T.Osakabe, J. Magn. Magn. Mater. **177-181** (1998) 53.