

XAFS analysis of the As-filled skutterudite compoundsYukako YAMAGUCHI¹, Kiyofumi NITTA², Takafumi MIYANAGA¹
Katsuhiko TAKEGAHARA¹, Hitoshi SUGAWARA³, Hideyuki SATO⁴¹Department of Advanced Physics, Hirosaki University, Hirosaki, Aomori 036-8561, Japan²Spring-8, Sayo, Hyogo 679-5198, Japan³Faculty of Science, Kobe University, Kobe 657-8501, Japan⁴Graduate School of Science, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan**Introduction**

The filled skutterudite compounds as RT_4X_{12} (R=Rare earth elements, T=Fe, Ru, Os, X=P, As, Sb) have many interesting physical properties with the combination of constituent elements. 12 X atoms forms the basket of the icosahedron which is a characteristic of this structure surround R ion, and the R ion is known to cause the anharmonic vibration called "rattling". It is reported that this anharmonic vibration contributes to disturb the lattice heat conduction, so the filled skutterudite compounds have attracted considerable attention as thermoelectric material. In recent years, the study on this rattling is conducted flourishingly. So far the behavior of the rattling is observed by the technique such as Raman scattering [1], XAFS [2], the supersonic wave [3]. However, the unified mechanism is not still provided. In this study, we performed a systematic investigation by XAFS to reveal the rattling mechanism in the As-filled skutterudite compounds.

Experimental and data analysis

RF_4As_{12} (R=La, Ce, Nd), RRu_4As_{12} (R=La, Ce, Nd) are studied in this project, but we show the preliminary result for $CeRu_4As_{12}$.

The thermal vibrations and the geometric disorder of the atomic position contribute to a Debye-Waller factor equivalent to distribution of the atomic distance from Ru to As. If a certain rare-earth element R causes a rattling in a basket of As, it seems that the behavior reflects to a Debye-Waller factor. It is a purpose to clarify the essence of the rattling by analyzing a Debye-Waller factor of XAFS. X-ray absorption measurements were made at beam line of NW10A at Photon Factory, KEK. Data analyses was performed using XANADU code [4] and FEFF8.01 code [5].

Results and discussion

Figure 1 shows $\chi(k)$ spectra of Ru K-edge EXAFS for $CeRu_4As_{12}$ measured at 25K and 300K as an example. Data quality is fairly good. Figure 2 shows the Fourier transforms (|FT|) of Ru K-edge EXAFS for $CeRu_4As_{12}$ measured from 25K to 300K. First peak is Ru-As and we analyze this peak by non-linear least square method. Figure 3 shows temperature dependence of the Debye-Waller factor, σ^2 , for $CeRu_4As_{12}$. Solid line is the results of Einstein fitting. We analyzed all EXAFS Debye-Waller factors obtained from the experiments in the same manner.

Einstein temperature, Θ_E , for $CeRu_4As_{12}$ is 362K and that for $NdRu_4As_{12}$ is 350K. They are higher than that for Sb-systems. We perform similar analysis for other As systems.

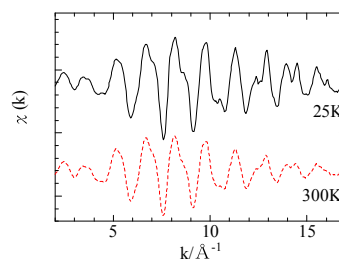


Fig. 1. $\chi(k)$ spectrum of the Ru K-edge of 25K and 300K for $CeRu_4As_{12}$

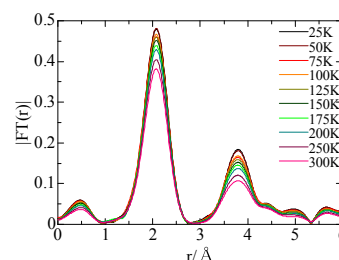


Fig. 2. FT of the Ru K-edge of 25 - 300K for $CeRu_4As_{12}$

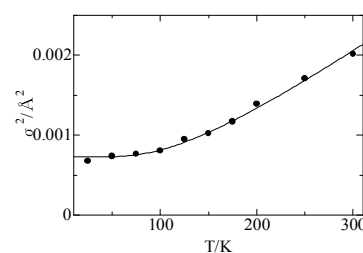


Fig. 3. Temperature dependence of the Debye Waller factor for $CeRu_4As_{12}$. Solid line is the result of Einstein fitting.

References

- [1] N. Ogita, *et al.*, Physica B., 383, (2006) 128.
- [2] K. Nitta, *et al.*, J. Phys. Soc. Jpn., 77, (2008) 063601.
- [3] T. Goto, *et al.*, Phys. Rev. B., 69, (2004) 180511.
- [4] H. Sakane, *et al.*, Jpn. J. Appl. Phys., 32, (1993) 4641
- [5] S.I. Zabinsky, *et al.*, Phys. Rev. B 52, (1995) 2995