Materials Science

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EXAFS thermal factor analysis on filled skutterudites RT_4Sb_{12} (R=La, Ce, Pr, Nd, Sm, T=Fe, Ru, Os)

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Introduction

Ternary compounds of filled skutterudites RT_4X_{12} (R=rare-earth, T=transition metal, X=pnictogen) shows various physical properties by changing the combination of atoms. Especially, anharmonic oscillation of rare-earth ions called “rattling” is key feature to understand such variational physical properties.

On the other hand, Extended X-ray Absorption Fine Structure (EXAFS) method is strong tool studying the dynamics of atomic fluctuation as well as the electronic state and the local structure of materials. Temperature dependent study of EXAFS reveal the contribution to atomic fluctuation separated from thermal vibrations or static distortions.

Experiment and Data analyses

Single crystal of the filled skutterudites RT_4Sb_{12} were grown by the Sb-flux method [1]. R L_{III} and Fe K-edge X-ray absorption spectra for powder samples prepared from the single crystals were obtained at BL9A, NW10A with a Si(111), Si(311) double crystal monochrometer at Photon Factory(KEK). The measurements were carried out in transmission mode with the detector of the ionization chambers in the temperature range from 25K to 300K.The EXAFS analyses were performed using the REX2000 software[2], and we could obtain inter atomic distance, the second order(or Debye-Waller factor) and the third order cumulant.

Result and Discussion

Figure1 shows of static distortion(σ_{static}) of Debye-Waller factor obtained from the equation,

$$C_2=h^2\coth(\Theta_E/2T)/4\pi\mu k_B\Theta_E+\sigma_{static}^2.$$

where C_2 is Debye-Waller factor, Θ_E is the Einstein temperature, μ is the reduced mass of Einstein vibrator and σ_{static} is the static distortion of each atomic pair. The abscissa axis shows the one-dimensional cage space defined by the differences between interatomic distance and each atomic (or ionic) radius.

In Fig.1 (a), Θ_E for Os(Fe, Ru)-Sb shows high values and does not depend on the cage space. On the other hand, Θ_E for R-Sb slightly decreases as the increasing Θ_E.

In Fig.2, static distortion for Os (Ru, Fe)-Sb is almost constant as the cage space. Only for the Fe-Sb atomic pair is most large value. The static disorder for R-Sb in RRu_4Sb_{12} and ROs_4Sb_{12} strongly depends on the cage space. It is interesting that the static distortion for Fe-Sb shows larger values and almost constant. The static distortion of R-Sb for $RFeS_4b_{12}$ does not so depend on the cage space.

From this thermal factor analysis of R-Sb and T-Sb atomic pairs, we achieved to know the relations between the cage space and thermal oscillation and the static distortion. From this systematic study, we could investigate the dynamics of rare-earth ions inside Sb icosahedrons cages .as the combinations of thermal oscillation and static distortion.

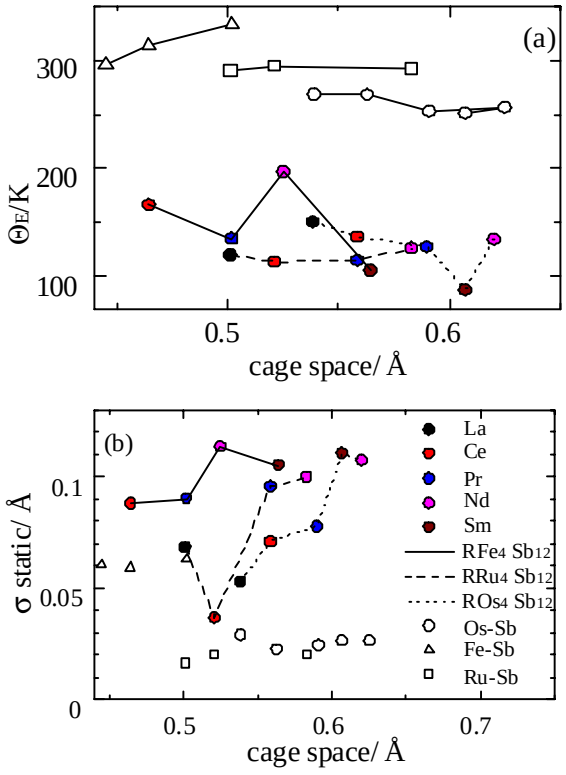

Figure 1 Cage space dependence of (a) Einstein temperature and (b) static disorder.

Reference

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[2]T. Taguchi et. al., Physica Scripta. Vol. **T115**, 205, (2005)