Visualization of rattling in filled skutterudite compounds

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Filled skutterudite compounds LnT_4X_{12} where Ln is rare earth, T is transition metal and X is pnictogen, exhibit wide variety of physical properties by combination of Ln, T and X. Recently, large thermal vibration of a rare earth ion, so-called ratting, becomes one of the key issue as a possible origin of interesting physical properties including unconventional heavy-fermion superconductivity in $PrOs_4Sb_{12}$, Nd-based heavy-fermion behavior in $NdOs_4Sb_{12}$ and so on. In order to reveal detailed pictures of the rattling and its relationship with f-electron, single crystal neutron diffraction experiments on $PrOs_4Sb_{12}$ and $NdOs_4Sb_{12}$ were carried out on 4-circle diffractometer FONDER. The maximum entropy method is employed in the structural analysis to avoid arbitrariness, which was achieved using software PRIMA and VESTA.

Figure 1 shows obtained nuclear scattering length density (hereafter, nuclear density) distributions of $PrOs_4Sb_{12}$ and $NdOs_4Sb_{12}$ at room temperature. These results correspond to the space- and time averaged nuclear density reflecting thermal vibration. Taking into account the difference in the coherent scattering length between Pr and Nd, b_{Pr}=4.58 fm and b_{Nd}=7.69 fm, isosurface levels in Fig. 1 are set at 4.58 fm/Å^3 and 7.69 fm/Å^3 for(a) $PrOs_4Sb_{12}$ and (b) $NdOs_4Sb_{12}$, respectively.

Widely spread nuclear density distributions could be seen at the center of the cage, corresponding to Pr and Nd. Note that the shapes for both Pr and Nd are not spherical, namely, both ions exhibit anisotropic thermal motion. A contrast between Pr and Nd is found in anisotropy and magnitude of spatial extent. In $PrOs_4Sb_{12}$, the distribution of Pr has an almost cubic shape with the edge parallel to the ⟨100⟩ direction and the width is broadest along the ⟨111⟩ direction. The spatial extent of Nd distribution in $NdOs_4Sb_{12}$ is wider than that of Pr, and becomes rather isotropic. The width along the ⟨100⟩ direction is slightly larger than that along ⟨111⟩ in $NdOs_4Sb_{12}$, which is contrary to $PrOs_4Sb_{12}$. It should be pointed out that both ⟨100⟩ and ⟨111⟩ directions for Ln ion correspond to the voids of the Sb icosahedron cage. In contrast, the nuclear distribution of Os and Sb in $PrOs_4Sb_{12}$ quite resembles to those in $NdOs_4Sb_{12}$. Further diffraction studies on detailed temperature dependence and extension to $LaOs_4Sb_{12}$ could provide keen insights on anisotropy with regard to a role of 4f electron and its relationship to various physical properties.

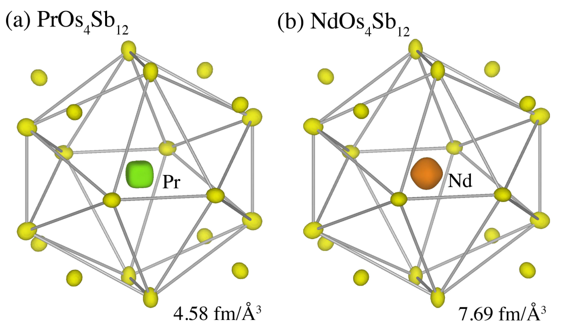

Fig. 1. Nuclear density distributions of $PrOs_4Sb_{12}$ and $NdOs_4Sb_{12}$ at room temperature.