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First-principles calculation of phonon dispersion of skutterudite compounds

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In some compounds with cage structures, including skutterudite compounds, it is suggested that a vibration of guest atoms in a cage relates to their anomalous behaviours. One charasteristic property is that guest atoms in the large cages show large atomic displacement, where anharmonic interactions between the guset atoms and the cages will become important. In fact, the experimental results of Raman, neutron, and X-ray inelastic scatterings have shown that a fourth-order anharmonic interaction dominates the temperature dependence of vibrational energy of the guest atoms. Since anharmonic interactions influence not only energy but also life-time and amplitude of vibrations, the study of anharmonicity is important. Therefore, we have investigated as a first step inter-atomic force constants and phonon dispersions using first-principles calculation in the harmonic approximation.

The calculations were performed by ABINIT code (http://www.abinit.org), which is based on the density functional formalism and planewave basis with norm-conserving pseudopotential. We used the local density approximation (LDA) for the exchange-correlation energy. At first we optimized structural parameters to minimize the total energy. Then, in the optimized structure, inter-atomic force constants were calculated using the density functional perturbation theory. The calculations were made for nine La-filled skutterudite compounds.

The optimized structural parameters are shown in Table, which are the lattice parameter a and the atomic position of pnictogen (0 y z). The table also lists the distance d between La and pnictogen atoms and the energy of the T_u mode which is a vibration of La atoms with the zero wavelength. The calculated a and d are smaller than experimental ones by ~ 0.05 Å. This is a typical trend found in calculations using LDA.

One of characteristic inter-atomic interactions is the 'self-interaction' coefficient k of the La atom. It is obtained from $dE = ku^2/2$, where dE is the energy increment when an La atom moves by a small distance u. If only La atoms move in the lowest-energy T_u mode, the energy of T_u mode is equal to $\omega = \sqrt{k/M}$, where M is the mass of an La atom. We list k and ω in Table.

In the poster, we will present phonon dispersion, specific heat, and atomic displacement parameters $< u^2 >$ of $LaRu_4P_{12}$ and $LaOs_4Sb_{12}$.

	a(Å)	y	z	d(Å)	T_u (cm^{-1})	k(mdyn/Å)	ω(cm^{-1})
$LaFe_4P_{12}$	7.743	0.3535	0.1516	2.978	118.7	1.221	122.2
$LaRu_4P_{12}$	8.019	0.3583	0.1437	3.096	92.0	0.746	95.5
$LaOs_4P_{12}$	8.036	0.3573	0.1435	3.094	90.1	0.779	97.6
$LaFe_4As_{12}$	8.205	0.3446	0.1566	3.106	88.0	0.901	104.9
$LaRu_4As_{12}$	8.450	0.3500	0.1499	3.217	70.3	0.542	81.4
$LaOs_4As_{12}$	8.473	0.3492	0.1495	3.219	68.3*	0.555*	82.3*
$LaFe_4Sb_{12}$	8.963	0.3358	0.1629	3.345	63.4*	0.615*	86.7*
$LaRu_4Sb_{12}$	9.173	0.3412	0.1580	3.449	47.1	0.346	65.0
$LaOs_4Sb_{12}$	9.202	0.3408	0.1569	3.452	42.0	0.332	63.3

Table : Calculated structural parameters, distance d between La and pnictogen atoms, the energy of La atom vibration, T_u mode, self-interaction coefficient k, and the energy ω obtained from k. The sign * means the preliminary result.