Ultrasonic investigation of off-center rattling in $Pr_{0.55}Nd_{0.45}Os_4Sb_{12}$

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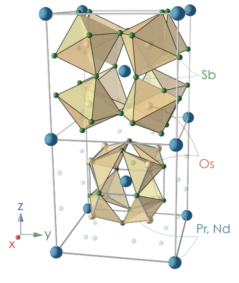
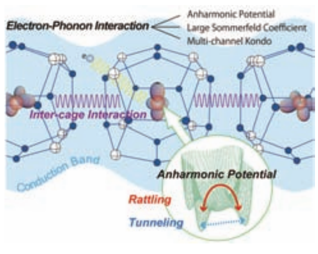


Crystal structure of the filled skutterudite ROs_4Sb_{12} (R = Pr, Nd).

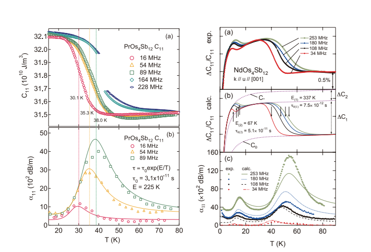


Physical schema of exotic phenomena regarding strong electron-phonon interaction.

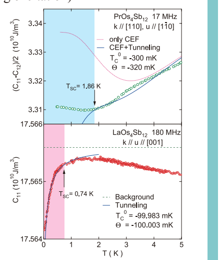
Introduction

Filled skutterudite $PrOs_4Sb_{12}$ is of particular interest because this compound exhibits unconventional superconductivity and magnetic field-induced quadrupolar-order. $NdOs_4Sb_{12}$ exhibits mean-field-type ferromagnetism. Both systems possess large Sommerfeld coefficient (shown on the right). These two compounds also show ultrasonic dispersion(s): single dispersion ~35 K for $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$; double dispersions ~15 K and ~40 K for Nd, and also low temperature softening in Γ_{23}-related elastic constants in $PrOs_4Sb_{12}$ and $LaOs_4Sb_{12}$ due to a thermally activated off-center motion of the rare-earth ion in an oversized atomic cage, known as ‘Rattling’ and ‘Tunneling’.

End compounds:
$PrOs_4Sb_{12}$ (HFSC): γ ~ 750 mJ mol^{-1} K^{-2} (originated from HF behavior)
$NdOs_4Sb_{12}$ (FM): γ ~ 520 mJ mol^{-1} K^{-2} (possibly due to low-lying excitation)

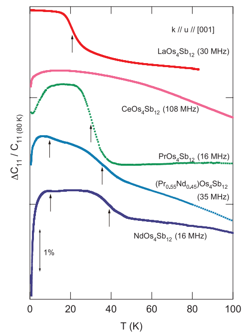


Ultrasonic dispersion due to rattling in $PrOs_4Sb_{12}$ (Left) and $NdOs_4Sb_{12}$ (Right). [1, 2]

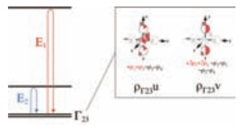


Elastic softening due to tunneling. [3]

Comparison of the Ultrasonic Dispersions



← Comparison of the relative change of elastic constants C_{11} and C_{44} in ROs_4Sb_{12} (R = La, Ce, Pr, $Pr_{0.45}Nd_{0.45}$, Nd) as a function of temperature.



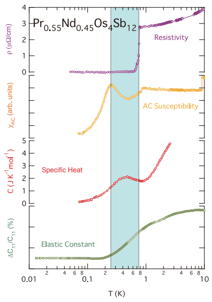
The double UD in the C_{11} of $NdOs_4Sb_{12}$ ($4f^3$) may be caused by variations in the anharmonicity of the potential due to differences in the electron–phonon interaction from those of $PrOs_4Sb_{12}$ ($4f^2$) and $LaOs_4Sb_{12}$ ($4f^0$), where only single UD has been observed thus far, and that of $CeOs_4Sb_{12}$ ($4f^0$), where no rattling was observed at a zero magnetic field. These differences in rattling characteristics and the number of $4f$ electrons imply that the interaction between $4f$ and conduction electrons also plays an important role in this issue.

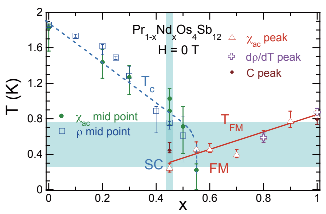
Motivation

Present research is motivated to verify a systematic change of rattling feature in the pseudo ternary systems $Pr_{1-x}Nd_xOs_4Sb_{12}$ and succesive transitions at low temperature, which is probably due to SC and FM coexisting at x = 0.45;

T_{SC} ~ 0.76 K
T_{FM} ~ 0.25 K.

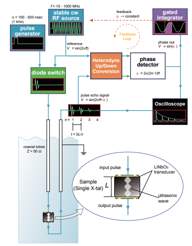
→A comparison of low-temp physical properties. [5]





Superconducting transition temp. T_c and ferromagnetic transition temp. T_{FM} vs Nd concentration x in the $Pr_{1-x}Nd_xOs_4Sb_{12}$ system. [4]

Experimental Details

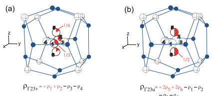


Elastic constant was measured by ultrasonic pulse-method with a phase comparator and heterodyne wave-detection circuit.

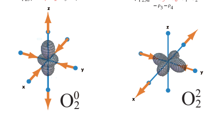
Γ_{23} off-center mode

The six numbered open spheres illustrated in the cage represent the positions of off-center potential minima along [100]. The Γ_{23} off-center mode is described by two charge distributions (a) $\rho_{\Gamma 23v}$ and (b) $\rho_{\Gamma 23u}$.

Γ_{23} off-center charge distributions

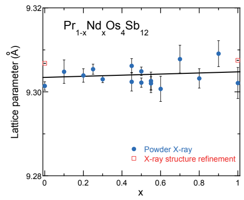


Coupled quadrupole moments and strains

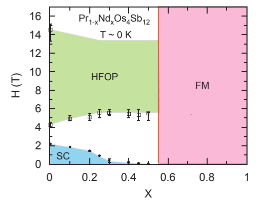


$\varepsilon_u = \frac{2\varepsilon_{zz} - \varepsilon_{xx} - \varepsilon_{yy}}{\sqrt{3}}$ $\varepsilon_v = \varepsilon_{xx} - \varepsilon_{yy}$

$Pr_{1-x}Nd_xOs_4Sb_{12}$

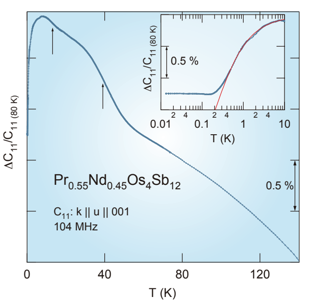


Nd substitution barely change the lattice parameter. Interestingly, the lattice constant of ROs_4Sb_{12} (R = rate earth) series seems like not following the lanthanide contraction even with a change of the guest R-ion, which indicates the rigidity of the $(OsSb_3)_4$ atomic cages, compared with phosphide and arsenide filled skutterudites.

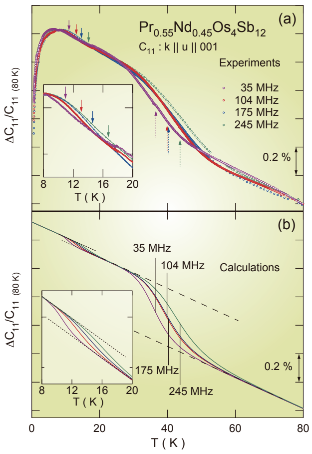


H-x diagram for $Pr_{1-x}Nd_xOs_4Sb_{12}$ at T ~ 0K extracted from ρ(T) measurements. The HFOP persists in $Pr_{1-x}Nd_xOs_4Sb_{12}$ up to x ~ 0.5, and peculiarly the kink in the lower phase boundary of the HFOP seems to correlate to the curvature breaking in the SC phase.

Results



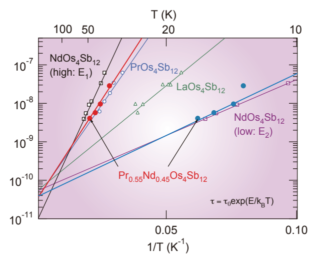
↑ Fig.1 Relative change in the elastic constant C_{11} as a function of temperature. The inset shows detailed behavior below 10 K displayed by log$T$ scale.



← Fig.2 (a) Frequency dependence of elastic constant C_{11} as a function of temperature. Upper and lower arrows display temperature that satisfy resonant condition $\omega\tau$ ~ 1. (b) Calculated elastic constant C_{11}. Dotted lines indicate high- and low-frequency limit $C_{11}(\infty)$ and $C_{11}(0)$, respectively. The inset is detailed behavior between 8 K to 20 K.

$$C_{11}(\omega) = C_{11}(\infty) - \sum_{i=1,2} \frac{\Delta C_i}{1+\omega^2\tau_i^2}$$

→ Fig.3 Arrhenius plots of the rattling parameters (τ_0, E). Solid lines are theoretical fit assuming relaxation time $\tau = \tau_0 \exp(E/k_BT)$ for filled skutterudite antimonides ROs_4Sb_{12} (R = La, Pr, Nd, and $Pr_{0.55}Nd_{0.45}$). Lower table shows comparison of rattling parameters of the filled skutterudites.



	E_1 (K)	$\tau_{0,1}$ (ps)	E_2 (K)	$\tau_{0,2}$ (ps)
$LaOs_4Sb_{12}$	127	50	-	-
$PrOs_4Sb_{12}$	225	31	-	-
$Pr_{0.55}Nd_{0.45}Os_4Sb_{12}$	240	29	74	36
$NdOs_4Sb_{12}$	337	7.5	67	51
$Pr(Os_{0.5}Ru_{0.5})_4Sb_{12}$	255	5.3	-	-
$LaFe_4Sb_{12}$	300	31	-	-

Summary

(1) Elastic constant C_{11} exhibits double ultrasonic dispersions at ~ 15 K and ~ 40 K, which are similar to the features found in $NdOs_4Sb_{12}$.
(2) Estimated rattling parameters possesses intermediate value between $PrOs_4Sb_{12}$ (E = 115 K) and $NdOs_4Sb_{12}$ (E_1 = 337 K).
(3) The levels off feature appears in C_{11} at around 200 mK imply that the Nd^{3+} ion’s degenerate CEF ground state will split due to the FM order even in the Pr^{3+} ion contributes to superconducting below T_{SC} ~ 0.76 K.

Reference

[1] T. Goto *et al.*: Phys. Rev. B **69** (2004) 185111.
[2] T. Yanagisawa *et al.*: J. Phys. Soc. Jpn. **77** (2008) 074607.
[3] Y. Nemoto *et al.*: J. Phys. Soc. Jpn. **77** (2008) Suppl. A 153.
[4] P.-C. Ho *et al.*: Physica B **403** (2008) 1038.
[5] P.-C. Ho *et al.*: to be published.

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