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LETTER

Magnetic-Field-Independent Ultrasonic Dispersions in the Magnetically Robust Heavy Fermion System $SmOs_4Sb_{12}$

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Elastic properties of the filled skutterudite compound $SmOs_4Sb_{12}$ have been investigated by ultrasonic measurements. The elastic constant $C_{11}(\omega)$ shows two ultrasonic dispersions at ~15 and ~53 K for frequencies ω between 33 and 316 MHz, which follow a Debye-type formula with Arrhenius-type temperature-dependent relaxation times and remain unchanged even with applied magnetic fields up to 10 T. The corresponding activation energies were estimated to be E_2 = 105 K and E_1 = 409 K, respectively. The latter, E_1, is the highest value reported thus far in the Sb-based filled skutterudites. The presence of magnetically robust ultrasonic dispersions in $SmOs_4Sb_{12}$ implies a possibility that an emergence of a magnetically insensitive heavy fermion state in this system is associated with a novel local charge degree of freedom, which causes the ultrasonic dispersion.

KEYWORDS: filled skutterudite, heavy fermion, rattling, elastic constant, ultrasonic dispersion

Intermetallic compounds with cagelike structures have been studied both experimentally and theoretically for several decades.[1,2] Recently, this area of research has received intense interest, since various exotic physical properties, *e.g.*, heavy-electron states of heavier rare-earth (Pr, Nd, and Sm) and La-based systems, multipole ordering, unconventional superconductivity, high thermoelectric figures of merit, and anharmonic vibrations of the guest ions, are observed in these types of compounds.[3–5] It has been suggested that these exotic phenomena originate from the characteristic cage structure in these systems. A well-known intermetallic system, with a cagelike structure, that exhibits a variety of correlated electron phenomena, are the filled skutterudite.[3–6] For instance, an unconventional heavy fermion (HF) state is observed for $SmOs_4Sb_{12}$,[7] where the electronic specific heat coefficient γ = 820 mJ mol^{-1} K^{-2} is insensitive to an applied magnetic field $H \parallel [100]$ up to 8 T.[8] This result is in contrast to the expected suppression of γ with magnetic field for the conventional (magnetic) Kondo effect, where the heavy fermion ground state is derived from spin degrees of freedom, as is often observed for Ce- and Yb-based compounds. In contrast, the exotic HF state in $SmOs_4Sb_{12}$ might be explained by a novel many-body effect, such as the multichannel Kondo effect, which is derived from multipole degrees of freedom and/or local charge fluctuations originating from off-center degrees of freedom of guest ions.[9,10]

By ultrasonic measurements, a frequency-dependent upturn of the elastic constant with an ultrasonic attenuation peak, called an ultrasonic dispersion (UD), has been observed in the cage compounds, such as $R_3Pd_{20}Ge_6$ (R = La, Ce, Pr) and ROs_4Sb_{12} (R = La, Pr, Nd).[11–13] Moreover, an unusual decrease in elastic constant has also been observed at very low temperatures in the La-based compounds. Owing to the copresence of UD and low-temperature elastic softening in non-4f La-based compounds, the origins of these phenomena are expected to share a common root cause, originating from a novel local charge degree of freedom. As a candidate for the origin, a model based on an off-center ionic configurations in a multiwell potential has been proposed. Here, the UD can be understood as a thermally activated off-center motion of guest ions, which was defined as '(off-center) rattling' in our previous papers.[13,14] On the contrary, a recent neutron scattering experiment on $PrOs_4Sb_{12}$ has revealed that the Pr-nuclei density distribution with an accuracy of 0.1Å is on-center-like without anisotropy at 8 K, while it is off-center-like with a strong anharmonicity at room temperature.[15] Therefore, it is still controversial whether the off-center configuration of guest ion is appropriate for understanding the above elastic anomalies.

Here, we note that the word 'rattling' has originally been used for the localized thermal vibration of the guest atom in an over-sized host cage with a large amplitude.[6] In the rattling systems, a low-lying optical phonon excitation of $\hbar\omega \sim$ meV has been commonly observed in several physical quantities, *e.g.*, Raman scattering, inelastic X-ray or neutron scattering and Einstein temperatures in the specific heat.[16–19] On the other hand, since the ultrasonic measurement with frequencies of ~ MHz can monitor relatively lower energy excitations of $\hbar\omega \sim 10^{-5}$ meV, a causal linkage between the optical phonon and the UD has not been confirmed thus far and still an open question. In order to shed light on the nature of the rattling phenomenon and UD, systematic changes in the characteristic parameters obtained from the UD and other physical properties among the Sb-based filled skutterudites will be reviewed in the present paper. In addition, although ultrasonic measurements of $SmOs_4Sb_{12}$ have already been reported,[20] there is no mention of the ultrasonic frequency dependence. The present study is focused on investigating the UD in $SmOs_4Sb_{12}$ in order to examine the magnetically robust novel local charge degree of freedom, which might be

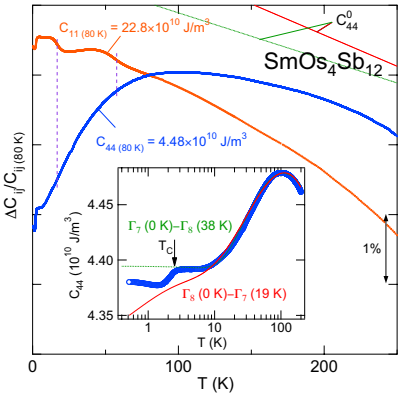

Fig. 1. (Color online) Elastic constants C_{11} (176 MHz) and C_{44} (165 MHz) of $SmOs_4Sb_{12}$ as a function of temperature T, displayed as a relative change for comparison. Inset: the green and red curves, displayed on a log$T$ scale, represent the fits of the quadrupole susceptibility by using the Γ_7 and Γ_8 ground states, respectively. The backgrounds of the elastic constant C^0_{44} curves for the fits are displayed as the same colored-dotted lines in the main panel.

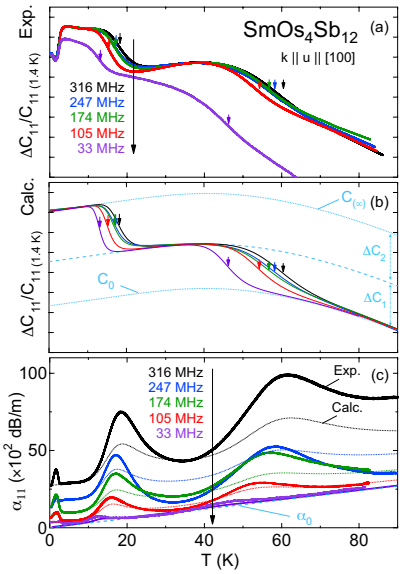

Fig. 2. (Color online) (a) Relative change in the elastic constant $\Delta C_{11}/C_{11}$ as a function of temperature T with frequencies varying from 33 to 316 MHz. (b) Theoretical fits to the frequency-dependent elastic constant. Arrows indicate the temperatures satisfying the resonant condition $\omega\tau \sim 1$, (c) Ultrasonic attenuation coefficient α_{11} as a function of temperature. Solid lines represent phenomenological calculations using the same parameters as used in the calculations in (b).

a possible candidate for the origin of the exotic HF state in $SmOs_4Sb_{12}$.

Single crystals of $SmOs_4Sb_{12}$ were grown using a molten flux growth method. The high quality of the single crystals used in the present study was confirmed by energy-dispersive X-ray spectroscopy, powder X-ray diffraction (for single crystals picked out of the same sample batch), the large residual resistivity ratio (RRR (300 K/ 2 K)) of 16.5, and an observation of acoustic de Haas-van Alphen signals at the elastic constant C_{11} (not shown). A cubic specimen, with a length of 0.775 mm, was used for the ultrasonic measurements. A change in sound velocity was detected by the conventional phase comparator method with pulsed ultrasound generated by $LiNbO_3$ transducers. Ultrasonic attenuation measurements were performed by recording the in-phase-(sine) and quadrature-(cosine) signals at fixed frequencies and the phase shift.[21] An Oxford Instruments Heliox-TL ^{3}He refrigerator with a superconducting magnet was used for measurements at $T \geq 300$ mK and $H \leq 10$ T.

A comparison of the elastic constants C_{11} and C_{44}, as a function of temperature, is displayed as the relative change in $\Delta C_{ij}/C_{ij}$ in Fig. 1. Note that $C_{11} = C_B + \frac{4}{3}\frac{C_{11}-C_{12}}{2}$ consists of the Γ_1-symmetry bulk modulus $C_B = \frac{C_{11}+2C_{12}}{3}$ and Γ_{23}-symmetry $\frac{C_{11}-C_{12}}{2}$ mode, while the C_{44} mode is associated with Γ_4 symmetry. C_{11} shows two upturns that are frequency-dependent and magnetic-field-independent (details will be described later in Figs. 2 and 3). The temperatures for the appearance of these two UDs are marked by vertical dotted lines in Fig. 1. In contrast, C_{44} shows neither an upturn nor a frequency dependence at these temperatures, except for the elastic softening due to the crystalline electric field (CEF) effect from $\sim$ 100 to 10 K and a steplike decrease at $T_C = 2.5$ K, below which a weak ferromagnetic moment develops for $SmOs_4Sb_{12}$.[7]

A similar contrasting behavior between C_{11} and C_{44} was also observed in the ROs_4Sb_{12} (R = La-Nd) systems. The ultrasonic mode dependence suggests that the UD is derived from a symmetrical off-center charge fluctuation of the guest ion, *e.g.*, Γ_{23}-symmetry off-center mode for ROs_4Sb_{12} and Γ_5 symmetry for $R_3Pd_{20}Ge_6$.[12] We note that our results, the softening of C_{44} in particular, are completely different from those of the previous work by Nakanishi *et al.*[20] The major difference could simply come from the fact that higher frequency and shorter pulses, *i.e.*, more focused ultrasonic waves, were used in the present measurements, which help to minimize the irregular reflection of ultrasonic waves from the crack or cavity on the surface or inside the crystal. A good example of this effect can be seen in a deviation of the lowest frequency (33 MHz) data of C_{11} in Fig. 2(a).

The softening of C_{44} can be analyzed in terms of quadrupole susceptibility based on the CEF effect.[22] Thus far, two different CEF energy level schemes have been proposed for the Sm^{3+} ion ($J = 5/2$) in $SmOs_4Sb_{12}$ in the O_h symmetry: *i.e.*, Γ_7(0 K) - Γ_8(38 K) and Γ_8(0 K) - Γ_7(19 K).[7,23] The inset of Fig. 1 shows theoretical fits to the experimental data below 200 K for both CEF schemes, both of which reproduce the softening of C_{44} down to $\sim$ 10 K. The leveling off of C_{44} below $\sim$ 10 K is described by the Van Vleck-type behavior, derived from a Γ_7-doublet ground state using fitting parameters including a coupling constant for a quadrupole-strain interaction $|g| = 258$ K and an intersite quadrupole interaction $g' = -0.79$ K. In contrast, the scheme with a Γ_8-quartet ground state gives a fit, which deviates from the C_{44} data due to a Curie-type contribution from the Γ_8-quartet ground state, that persists down to T_C. We should, however, note that the large value of the low-temperature specific heat indicates that complex many-body physics dominates the low-temperature state in $SmOs_4Sb_{12}$.[8] Therefore, it is likely that the deviation of the C_{44} data from the fit, *i.e.*, the shoulder for $T \lesssim$ 10 K, should probably be explained by a modified quadrupole susceptibility picture that includes a crossover from a Kondo singlet state (*e.g.*, as described for $CePd_2Al_3$)[24,25] or some other exotic Kondo states (*e.g.*, the multichannel Kondo ef-

fect).[10,26] Thus, the possibility of the Γ_8-quartet ground state still remains. If we skip the adjustment of the many-body effect, the fitting parameters for the Γ_8 model assume relatively larger values: $|g| = 369$ K and $g' = -1.76$ K. In the present paper, we will focus on the ultrasonic dispersions and leave the discussion of the correlated electron physics on the CEF effects for a later publication.

Figures 2(a) and 2(c) show the relative change in the elastic constant $\Delta C_{11}/C_{11}$ and the ultrasonic attenuation coefficient α_{11} as a function of temperature with fixed frequencies varying from 33 to 316 MHz. The two upturns in C_{11}, indicated by the arrows in Fig. 2(a), are found to shift to higher temperatures with increasing frequency. Maxima in α_{11} are also observed at ~ 53 and ~ 15 K, which show the same frequency dependence as the upturns in C_{11}. The frequency-dependent elastic constant $C(\omega)$, with ω being the ultrasonic frequency, and the attenuation coefficient $\alpha(\omega)$ are well described phenomenologically, as shown in Fig. 2(b), by a Debye-type dispersion given by $C(\omega) = C_{(\infty)} - \sum_{i=1,2} \frac{\Delta C_i}{1+\omega^2\tau_i^2}$ and $\alpha(\omega) = \alpha_0 - \sum_{i=1,2} \frac{\Delta C_i}{2\rho v_{i,\infty}^3} \frac{\omega^2\tau_i}{1+\omega^2\tau_i^2}$, where an Arrhenius-type temperature dependence of the relaxation time, $\tau_i = \tau_{0,(i)} \exp(E_i/k_B T)$, is assumed.[14] Here, $i = 1$ and 2 are the indices for analyzing two separated sets of UDs at ~ 53 and ~ 15 K, respectively. Here, ΔC_i in the above equations is the total change for each upturn between the high-frequency limit C_∞ and low-frequency limit C_0 of the elastic constant, which are estimated by the extrapolation of the temperature dependence of C_{11} over 20 K, without the UDs and possible CEF effect (as seen below 10 K in Fig. 2(a)), as displayed as dotted curves in Fig. 2(b). Two parameter sets of activation energy E_i and attempt time $\tau_{0,(i)}$ were obtained from the phenomenological fits for the relaxation $\omega\tau \sim 1$ in Figs. 2(b) and 2(c), even though the ultrasonic attenuation peaks are often experimentally observed to be sharper than the phenomenological fits.[14] The parameters are summarized in Table I with the related compound parameters of UD and also other physical properties, including the lattice constant a, the Debye and Einstein temperatures Θ_D and Θ_E, and the Sommerfeld coefficient γ.

Figure 3 shows C_{11} vs T at a frequency of 105 MHz and for various magnetic fields between 0 and 10 T. The UDs are unaffected by magnetic fields up to 10 T except for the field dependent CEF effect that appears in C_{11} below 15 K, as can be seen in the enlarged scale in the inset of Fig. 3. Such insensitivity in the UD to magnetic fields is commonly observed in the ROs$_4$Sb$_{12}$ (R = La-Nd) system, providing strong evidence that the UD is not magnetic but rather electric, *i.e.,* the novel local charge degree of freedom, in origin.

Figures 4(a) and 4(b) show systematic changes in the UDs and an Arrhenius plot of the relaxation time τ_i vs $1/T$ for ROs$_4$Sb$_{12}$ (R = La-Sm), respectively. The double UDs thus far only have been observed in NdOs$_4$Sb$_{12}$ ($4f^3$) and SmOs$_4$Sb$_{12}$ ($4f^4$), while LaOs$_4$Sb$_{12}$ ($4f^0$) and PrOs$_4$Sb$_{12}$ ($4f^2$) show single UD. It has been pointed out that the observed double UDs are comparable to the ^{123}Sb-NQR measurements on ROs$_4$Sb$_{12}$ (R = La-Sm), which show double or triple peaks in the temperature dependence of $1/T_2$ for R = Pr-Sm due to a certain local charge fluctuation with a correlation time of ~ 10^{-6} s, while LaOs$_4$Sb$_{12}$ shows only a single peak.[27]

When the rare-earth ion changes from R = La to Sm, not only the guest ion mass but also the amount of free space for

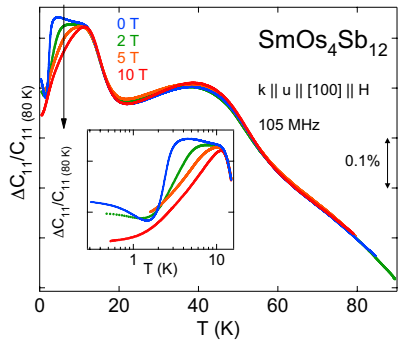

Fig. 3. (Color online) Temperature dependence of the relative change of C_{11} for various magnetic fields H along the [001] direction. The inset displays an enlarged view below 20 K on a log$T$ scale.

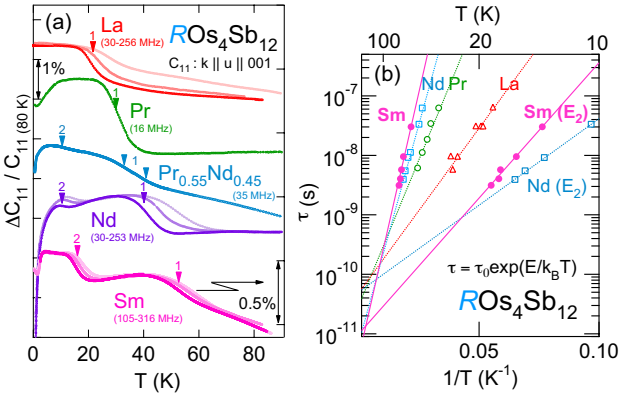

Fig. 4. (Color online) (a) Comparison of the UDs that appear at elastic constant C_{11} of ROs$_4$Sb$_{12}$ (R = La-Sm) at several frequencies. Lower arrowheads with numbers 1 and 2 indicate the relaxation point $\omega\tau_i \sim 1$ for $i = 1$ and 2, respectively. The displayed data have been shifted to eliminate overlapping with each other and the SmOs$_4$Sb$_{12}$ data are magnified three times for the $\Delta C_{11}/C_{11}$-axis. (b) Arrhenius plots of the characteristic parameters of the UDs ($\tau_{0(i)}$, E_i) for ROs$_4$Sb$_{12}$ (R = La-Sm).

the Einstein oscillator in the atomic cage increases, owing to the lanthanide contraction, *i.e.*, the ionic radius of the guest ion decreases, while the cage radius remains nearly constant for ROs$_4$Sb$_{12}$.[6] We speculate that such changes in the guest ion conditions give rise to an additional anharmonicity of the potential, *e.g.*, flat base or double well, for the local ionic configuration, and result in the emergence of additional UD and NQR peaks in R = Nd and Sm.

In Fig. 4(b), the slope of the line is equivalent to the activation energy and the markers indicate frequencies and temperatures, where the relaxation of UD meets a resonant condition $\omega\tau_i \sim 1$. We find that the activation energy E_1 and Sommerfeld coefficient γ increase and $\tau_{0,(1)}$ decreases from R = La to Sm in the ROs$_4$Sb$_{12}$ series, as summarized in Table I. The increase in γ generally means that a characteristic temperature of the electronic correlation, such as Kondo temperature, exhibits an opposite (decrease) tendency. Therefore, the similar increase tendencies of E_1, E_2, and γ found in the present study imply that the activation energies are not *directly* connected to the characteristic temperature of the electron mass-enhancement mechanism in SmOs$_4$Sb$_{12}$.

Next, we compare the Einstein temperature with activation energies. If we pick out the lower activation energy of each

Table I. Comparison of characteristic parameters of UDs and rattling, and some physical properties of the related filled skutterudites.[6,12,19,28]

	Lattice Constant a (Å)	Debye Temp. Θ_D (K)	Einstein Temp. Θ_E (K)	Sommerfeld Coef. γ (mJ mol^{-1}K^{-2})	Activation Energy E_1 (K)	Attempt Time $\tau_{0,(1)}$ (ps)	E_2 (K)	$\tau_{0,(2)}$ (ps)
$LaOs_4Sb_{12}$	9.3081	270	60.5	36-56	127	50	-	-
$CeOs_4Sb_{12}$	9.3011	304	-	92-180	-	-	-	-
$PrOs_4Sb_{12}$	9.3031	165-320	-	310-750	225	31	-	-
$NdOs_4Sb_{12}$	9.2989	255	39	520	337	7.5	67	51
$SmOs_4Sb_{12}$	9.3009	294	40.1	820	409	4.4	105	10
$LaRu_4Sb_{12}$	9.2700	275	72.8	47	-	-	-	-
$LaFe_4Sb_{12}$	9.1395	314	87.6	122	300	31	-	-
$PrFe_4Sb_{12}$	9.1351	-	-	~1000	360	3	-	-

compound for comparison, *i.e.,* E_1 = 127 K for La, E_2 = 67 K for Nd, and E_2 = 105 K for Sm, they seem to be qualitatively comparable to the systematic change in Einstein temperature Θ_E; 60.5 K for La, 39 K for Nd, and 40.1 K for Sm in Table I. This finding implies that the optical phonon and UD could be linked to the unknown prefactor of ~ 2. This linkage might be due to a novel energy dissipation mechanism via conduction electron-phonon interaction, as previously proposed by Hattori *et al.*[10] If there exists such a conduction electron-phonon interaction, it cannot be ruled out that the origin of activation energies is *indirectly* involved in the effective mass-enhancement mechanism, which might be mediated by the interaction between conduction electrons and phonons in the present system.

Finally, we discuss the possible copresence of Curie-type softening and UD in $SmOs_4Sb_{12}$. The low-temperature softening as observed in $LaOs_4Sb_{12}$ could, however, not be found in the present study of $SmOs_4Sb_{12}$ because the marked changes in elastic constants due to ferromagnetic ordering at 2.5 K and the CEF effect under magnetic fields hide such features. In order to elucidate the possible ground states of the local charge degrees of freedom and extract the CEF level scheme in the present compound, further measurements, such as elastic constant $(C_{11}$-$C_{12})/2$ and C_{44} measurements in magnetic fields, are required. In the present discussions, the Sm-ion valence fluctuations[29] are not taken into account. We can, however, conclude that the magnetically robust HF compound $SmOs_4Sb_{12}$ at least has the magnetic field-independent relaxation of local charge degree of freedom, which cause ultrasonic dispersions and will be a key to understanding the novel many-body effect in the present system.

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