Elastic properties of the non-Fermi-liquid metal CeRu$_4$Sb$_{12}$ and dense Kondo semiconductor CeOs$_4$Sb$_{12}$

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We have investigated elastic properties of the Ce-based filled skutterudite antimonides CeRu$_4$Sb$_{12}$ and CeOs$_4$Sb$_{12}$ by means of ultrasonic measurements. CeRu$_4$Sb$_{12}$ shows a slight increase around 130 K in the temperature dependence of the elastic constants C_{11}, $(C_{11}-C_{12})/2$, and C_{44}. No apparent lattice softening toward low temperature expected by a quadrupolar response of the $4f$-electronic ground state of the Ce ion was observed at low temperatures. On the other hand, CeOs$_4$Sb$_{12}$ shows a pronounced lattice softening toward low temperature in the longitudinal C_{11} as a function of temperature below about 15 K, while a slight lattice softening was observed in the transverse C_{44} below about 1.5 K, in contrast to no softening in CeRu$_4$Sb$_{12}$ at low temperatures. Furthermore, CeOs$_4$Sb$_{12}$ shows a steep drop around a phase-transition temperature of T_s =0.9 K in both C_{11} and C_{44}. The lattice softening observed in C_{11} below about 15 K cannot be explained reasonably only by the crystalline electric-field (CEF) effect. Instead, it is most likely to be responsible for a coupling between elastic strain and a relevant renormalized quasiparticle band with a small energy gap in the vicinity of the Fermi level. The elastic properties and the $4f$ ground state of Ce ions in CeRu$_4$Sb$_{12}$ and CeOs$_4$Sb$_{12}$ are discussed in terms of the CEF effect and the band structure in the vicinity of Fermi level.

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I. INTRODUCTION

Filled skutterudite compounds, RETr$_4$X$_{12}$ (RE: rare earth; Tr=Fe, Ru, Os; X: pnictogen) exhibit a wide range of electrical and magnetic properties, mainly due to the $4f$-electronic state of RE.[1] In this family, semiconducting behavior is seen mostly in Ce and U compounds.[2–4] The energy gap formation in dense Kondo systems is one of the most interesting subjects in strongly correlated electron systems. The strong electric correlation effect of f electrons plays a crucial role in the systems. In the Ce compounds of CeFe$_4$P$_{12}$, CeFe$_4$As$_{12}$, CeRu$_4$P$_{12}$, and CeOs$_4$P$_{12}$ the energy gap has been estimated to be of 110, 10, 86, and 34 meV, respectively.[3,5,6] The energy gap is approximately proportional to the lattice constant in Ce-based filled skutterudites.[7] More interestingly, the non-Fermi-liquid (NFL) behavior appears by the disappearance of the energy gap in the vicinity of Fermi level at low temperatures, which is realized in CeRu$_4$Sb$_{12}$.[7–10] Furthermore, CeRu$_4$Sb$_{12}$ exhibits intermediate valence behavior in the magnetic susceptibility and electrical resistivity in which an anomalous temperature dependence appears below about 100 K. The electronic contributions to the specific heat is rather large with 380 mJ/mol K^2.[8] Recent optical and ultrahigh resolution photoemission studies found that CeRu$_4$Sb$_{12}$ exhibits a charge gap feature below 70 K with the energy gap of Δ =47.1 meV.[11] A further interesting behavior: NFL appears below about 1 K in the specific heat with the unusual ln T dependence and below 5 K in the resistivity with a $T^{1.65}$ dependence.[9]

Recently, Bauer *et al.* have found that the resistivity of CeOs$_4$Sb$_{12}$ exhibits a semiconductorlike behavior at low temperatures with energy gap of 5–15 K by the use of an activated conduction law in the temperature range $25 < T$ < 50 K.[12] They also estimated the Kondo temperature T_K to be about 90 K. The relatively large specific-heat coefficient $\gamma = C/T$ of 96 mJ/mol K^2 indicates the formation of heavy Fermion system at low temperatures. These experimental results indicate that the dense Kondo system with the energy gap at the Fermi level is realized in CeOs$_4$Sb$_{12}$. The magnetic susceptibility was explained by the crystalline electric-field (CEF) effect with the energy splitting of Δ=327 K between the Γ_7 doublet ground state and the Γ_8 quartet excited state.[12] It also suggested that the Ce ions are almost trivalent in CeOs$_4$Sb$_{12}$. The lattice parameters of REOs$_4$Sb$_{12}$ follow the lanthanide contraction at room temperature, at least.[13] The well-localized nature of Ce ions expected by the CEF effect is not consistent with the character of a dense Kondo system with an energy gap. Furthermore, CeOs$_4$Sb$_{12}$ shows a clear anomaly at T_s=0.9 K in the specific heat, indicating an intrinsic phase transition.[7,14] However, the estimated entropy is extremely small amount of 2% of R ln 2 which is expected by the Γ_7 doublet ground state. Thus we suppose that this phase transition is not ascribable to the well-localized $4f$ electronic state derived from the Ce ion. Interestingly, this transition shifts to higher temperatures with increasing magnetic field, reported by Namiki *et al.*[14] They proposed that this transition may be responsible for the instability of the Fermi surface, e.g., charge-density wave (CDW) or spin-density wave (SDW) transition. In this way a dual character

of $4f$ electronic state of the Ce ions, localized and/or itinerant nature, has been reported in $CeOs_4Sb_{12}$ to date.

Ultrasonic measurements are particularly suited to study via the quadrupolar response of the $4f$ electronic ground state split by the CEF effect if the $4f$ electrons are localized well. The elastic constants, as the quadrupolar susceptibility, measure the diagonal (Curie terms) and off-diagonal (Van Vleck terms) quadrupolar matrix elements. The quadrupolar response of the $4f$ electronic ground state split by CEF effect causes a characteristic anomaly in the temperature dependence of the elastic constants due to the terms.[15–17] On the other hand, narrow quasiparticle bands formed by heavily renormalized quasiparticles cause elastic anomalies as well if the $4f$ electrons are delocalized.[15,18,19]

In this paper we report on the elastic properties of single-crystal $CeRu_4Sb_{12}$ and $CeOs_4Sb_{12}$ by means of ultrasonic measurements. The present results indicate that the low-temperature properties are less governed by the CEF effect in both systems. Instead, the $4f$ electronic states of Ce ions is likely explained reasonably by the delocalized picture with an energy gap at the Fermi level especially for the case of $CeOs_4Sb_{12}$. The preliminary reports have been published in Ref. 20.

II. EXPERIMENT

Single crystals of $CeRu_4Sb_{12}$ and $CeOs_4Sb_{12}$ were prepared using a molten-metal-flux growth method with Sb flux. The specimen used in our study has a size of $2.27 \times 4.14 \times 1.28$ mm^3 for $CeRu_4Sb_{12}$, and $1.0 \times 0.9 \times 0.5$ mm^3, respectively, but only with the crystallographic $\langle 100 \rangle$ axis for $CeOs_4Sb_{12}$. In consequence, the measurements of the elastic constants C_{11} and C_{44} were possible for $CeOs_4Sb_{12}$ in this study. The sound velocity (v) was measured by an ultrasonic apparatus based on a phase comparison method in a magnetic field up to 12 T generated by a superconducting magnet. The plates of $LiNbO_3$ transducers were used to generate and detect the sound waves with the frequencies from 5 to 30 MHz. The transducers were glued on the parallel planes of the sample by the elastic polymer Thiokol. The absolute value of the elastic constant $C = \rho v^2$ by using the density ρ of the crystal was estimated explicitly for $CeRu_4Sb_{12}$ with a lattice parameter $a = 9.2721$ Å, but the estimation was impossible for $CeOs_4Sb_{12}$ because the thickness of the present sample was not large enough.

III. EXPERIMENTAL RESULTS

A. Elastic property of $CeRu_4Sb_{12}$

In Fig. 1 we give an overview of the temperature dependence of the three elastic constants C_{11}, $(C_{11} - C_{12})/2$, and C_{44} for the cubic $CeRu_4Sb_{12}$. C_{11} was measured by the longitudinal sound wave with frequencies of $10-30$ MHz propagated along the $\langle 100 \rangle$ axis. $(C_{11} - C_{12})/2$ and C_{44} were measured by the transverse sound wave with frequencies of $5-15$ MHz propagated along the $\langle 110 \rangle$ axis with the polarization parallel to the $\langle 1-10 \rangle$ axis and propagated along the $\langle 100 \rangle$ axis with the polarization parallel to the $\langle 010 \rangle$ axis,
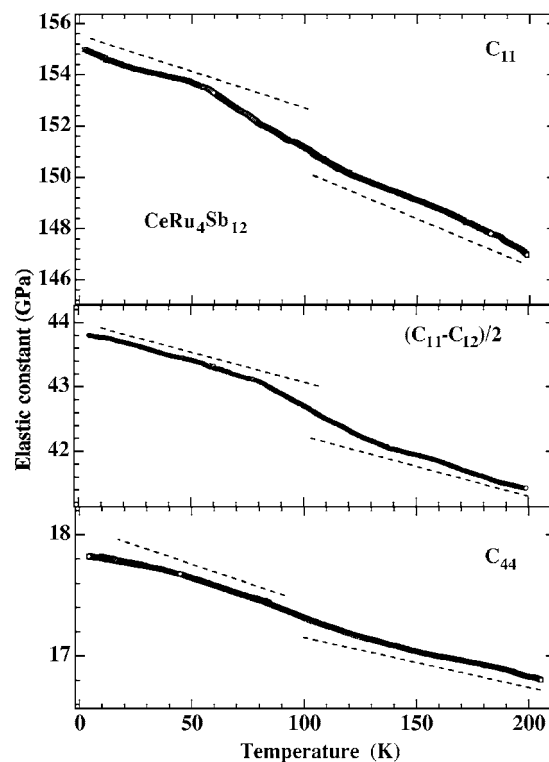

FIG. 1. Temperature dependence of the elastic constants C_{11}, $(C_{11} - C_{12})/2$, and C_{44} for $CeRu_4Sb_{12}$.

respectively. The $CeRu_4Sb_{12}$ data exhibit roughly normal behavior: a stiffening with decreasing temperature and an almost temperature dependence at the lowest temperatures. However, it is seen that the elastic constants exhibit a slight increase around 130 K. It should be noted that the magnetic susceptibility and electrical resistivity exhibit an anomalous temperature dependence around this temperature,[8] as will be discussed in detail later. Figure 2 shows the temperature dependence of C_{44} under selected fields along the $\langle 100 \rangle$ axis. The monotonic increase shows little change even in magnetic fields up to 12 T at low temperatures within the experimental error. It should be also noted that the NFL behavior appears in the temperature dependence of the electrical resistivity and specific heat in this temperature range below 5 K.[8] It seems that the elastic constant may be insensitive to the NFL property. The absolute values of the elastic constants and calculated bulk modulus $C_B = (C_{11} + 2C_{12})/3$, and Poisson ratio $\gamma = C_{12}/(C_{11} + C_{12})$ from C_{11} and $(C_{11} - C_{12})/2$ at both 77 and 4.2 K are listed in Table I.

B. Elastic property of $CeOs_4Sb_{12}$

Next, we give an overview of the elastic constants C_{11} and C_{44} for the cubic $CeOs_4Sb_{12}$. Figure 3 shows the relative change of elastic constants C_{11} and C_{44} as a function of temperature. Both elastic constants exhibit a stiffening monotonically with decreasing temperature. However, a pronounced lattice softening toward low temperature was observed in C_{11} below around 15 K as highlighted by the circle in Fig. 3, which is markedly different from that of $CeRu_4Sb_{12}$. In addition, a steep drop was observed at T_s in
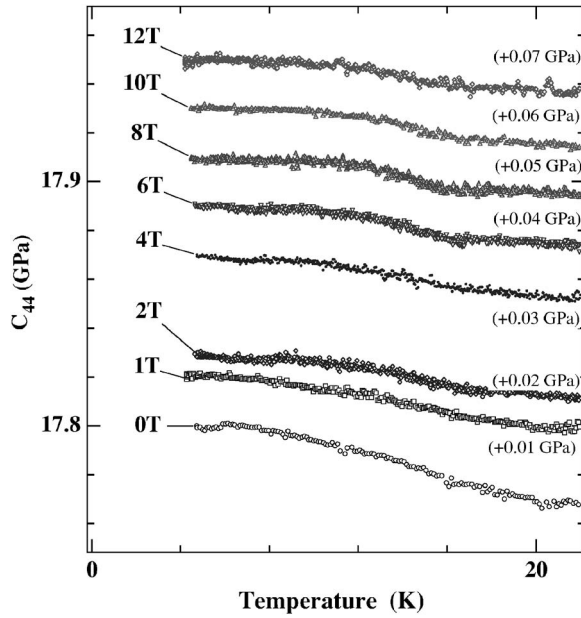
FIG. 2. Temperature dependence of the elastic constants C_{44} for CeRu$_4$Sb$_{12}$ under selected fields along the $\langle 100 \rangle$ axis.

the elastic constants C_{11} and C_{44}. The lattice softening in C_{44} toward the transition temperature T_s is less pronounced as also highlighted by circles in Fig. 3. The degree of the softening in C_{11} is of about 1% down to the lowest temperature of 0.5 K in this study. It should be noted that a slight increase was observed around 35 K in the elastic constants C_{11} and C_{44}. This prominent feature will be discussed in detail later in terms of anharmonic oscillation of Ce ions in a cage of Sb icosahedron. The inset of Fig. 3 shows the temperature dependence of C_{11} under selected fields, and the low-temperature region is shown separately in Fig. 4, combined with the derivative of C_{11} with respect to temperature. Their offsets are shifted arbitrarily to avoid the overlap and for clarity. The softening was gradually suppressed with increasing field. The middle point of the drop shifts to higher temperatures with increasing field as indicated by the arrows in the inset of Fig. 3. Furthermore, the steep drop, probably due to the phase transition, was gradually suppressed with increasing field. The characteristic softening and the steep drop were almost undetectable above 5 T. Figure 5 shows the field dependence of C_{11} at selected temperatures. The C_{11}

TABLE I. The absolute values of each elastic constant, the calculated bulk modulus $C_B = (C_{11} + 2C_{12})/3$ and Poisson ratio $\gamma = C_{12}/(C_{11} + C_{12})$ for CeRu$_4$Sb$_{12}$ at both 77 and 4.2 K.

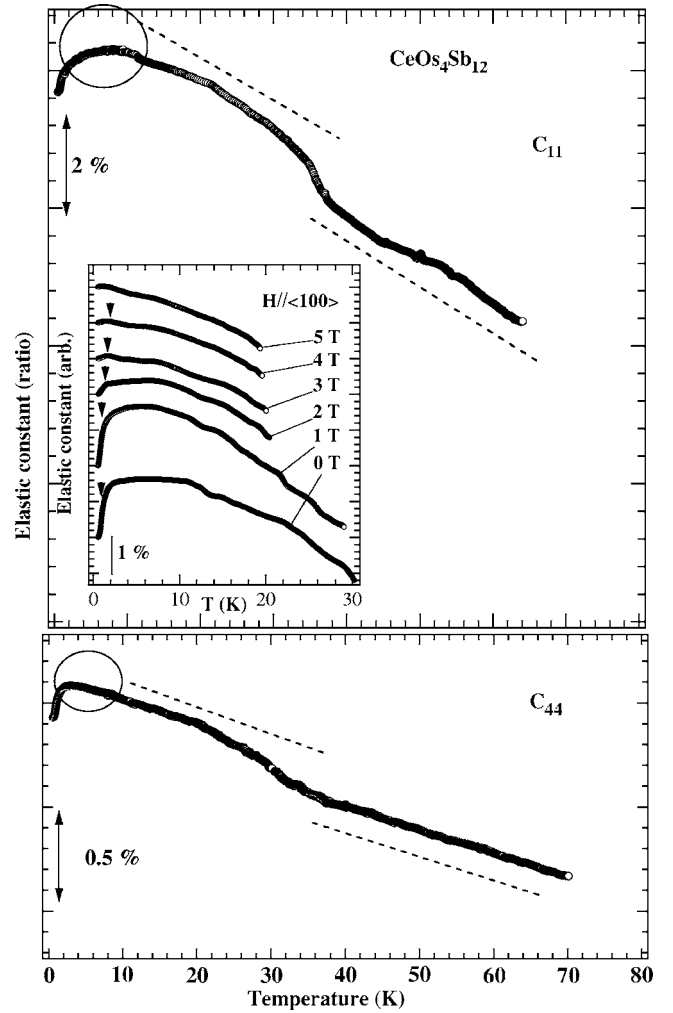|	Elastic constants	
Mode	at 4.2 K	at 77 K
C_{11}	155 GPa	152 GPa
$(C_{11} - C_{12})/2$	43.8 GPa	43.1 GPa
C_{44}	17.8 GPa	17.5 GPa
$C_B = (C_{11} + 2C_{12})/3$	96.6 GPa	94.5 GPa
$\gamma = C_{12}/(C_{11} + C_{12})$	0.303	0.302

FIG. 3. Temperature dependence of the elastic constants C_{11} and C_{44} for CeOs$_4$Sb$_{12}$. Inset shows the temperature dependence of the C_{11} for CeOs$_4$Sb$_{12}$ under selected fields along the $\langle 100 \rangle$ axis. Their offsets are shifted arbitrarily. The circles are marked for contrast.

exhibits a monotonic increase with increasing field. However, a distinct anomaly was observed at around 1 T at the temperature of 1.2 K. This transition corresponds to the phase boundary of the aforementioned ordered state.[7,14] Figure 6 shows the temperature dependence of C_{44} under selected fields, combined with the derivative of C_{44} with respect to temperature. Again, in contrast to C_{11}, a softening toward the transition temperature of 0.9 K is less pronounced as highlighted by the circles in Fig. 3. A steep drop, probably due to the phase transition, is observed in C_{44} as well. Similar to C_{11}, the drop was gradually suppressed with increasing field. Figure 7 shows a comparison between the temperature dependence of the attenuation and the corresponding elastic constant in zero field. A remarkable upturn was observed below T_s. Note that the temperature where C_{11} shows the remarkable softening is close to the temperature where the attenuation shows the upturn, as readily recognized in Fig. 7. In general, such an increase of the attenuation indicates an increase in scattering for the propagating sound wave below T_s. This is one of the prominent features in the ordered phase. The anomaly shifts to lower temperatures with in-
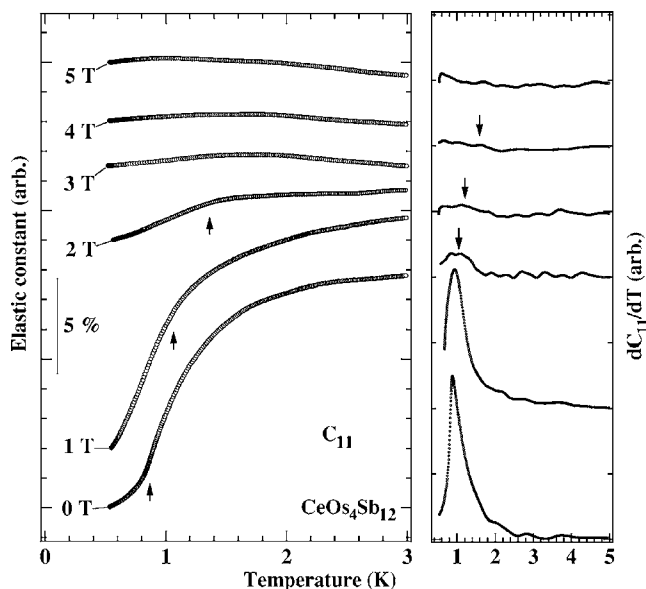
FIG. 4. (Left) Low-temperature part of the temperature dependence of the C_{11} under selected fields along the $\langle 100 \rangle$ axis. (Right) The corresponding dC_{11}/dT curves in the low-temperature region. Their offsets are shifted arbitrarily.

creasing field along the $\langle 100 \rangle$ axis as shown in the inset of Fig. 7. From analysis of these results, one can depict the magnetic phase diagram for CeOs$_4$Sb$_{12}$ as shown in Fig. 8. The boundary goes up to the higher field side and becomes gradually obscure in the high-temperature region, consistent with the previous specific-heat and electric resistivity measurements.[7,14] This nature is reminiscent of a ferromagnetic or antiferroquadrupolar ordering.

IV. DISCUSSION

First, we would like to examine the level scheme of Ce^{3+} ion split by the cubic crystalline electric-field (CEF) effect

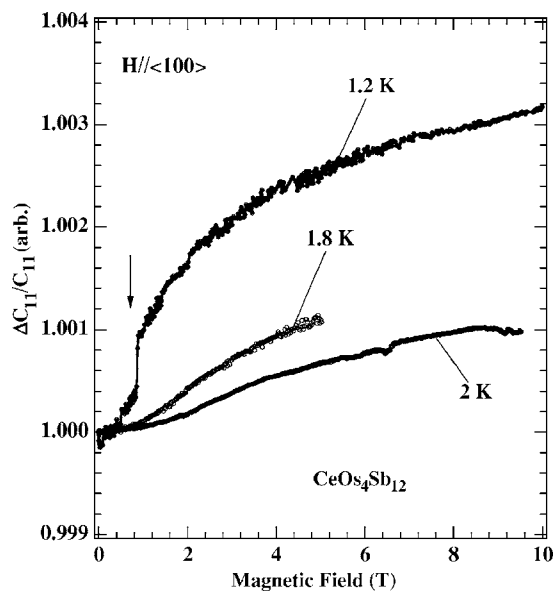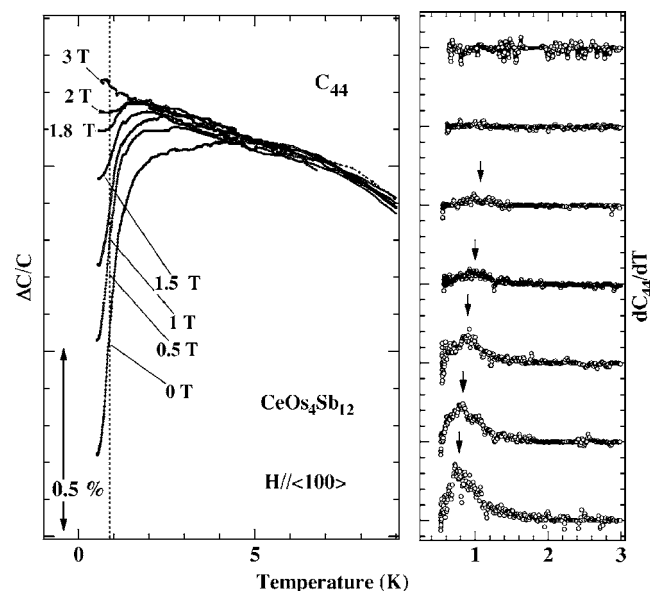FIG. 6. (Left) Low-temperature part of the temperature dependence of the C_{44} under selected fields along the $\langle 100 \rangle$ axis. (Right) The corresponding dC_{44}/dT curves in the low-temperature region. Their offsets are shifted arbitrarily.

into a Γ_7 doublet and Γ_8 quartet. As mentioned in the Introduction, the following CEF level scheme was proposed: $\Gamma_7(0)-\Gamma_8(327\ \text{K})$ with the effective magnetic moment μ_{eff} $=2.0\mu_B/\text{Ce}$, assuming that the Ce ions are trivalent in

FIG. 5. Magnetic-field dependence of the elastic constant C_{11} at selected temperatures around the transition temperature.
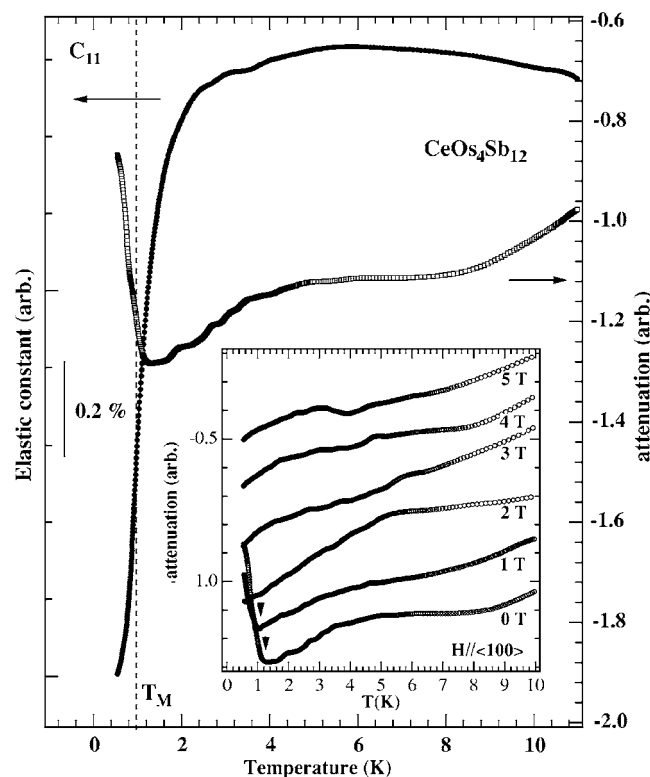
FIG. 7. Temperature dependence of the attenuation and the corresponding elastic constant C_{11} in zero field. Insets shows the temperature dependence of the attenuation under selected fields along the $\langle 100 \rangle$ axis.
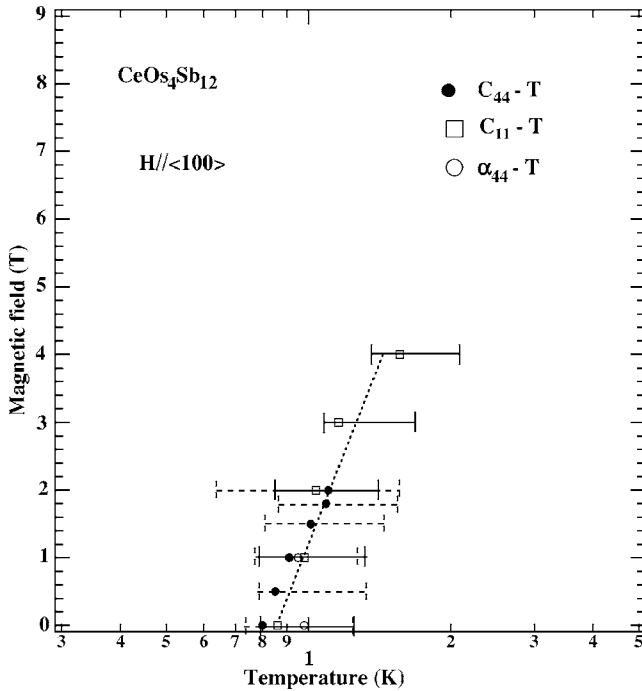
FIG. 8. Magnetic phase diagram for $CeOs_4Sb_{12}$ for the field along the $\langle 100 \rangle$ axis deduced from the present results. The broken line is guide to eyes. The horizontal solid and broken lines represents the error bars for C_{11} and C_{44}, respectively. They were determine by the full width at half maximum of the anomaly observed around T_s in the derivative of the elastic constants with respect to temperature.

$CeOs_4Sb_{12}$.[12] In general, a characteristic anomaly as a function of temperatures is expected in relevant elastic constants in rare-earth compounds with well-localized $4f$ electrons reflecting on the ground state of the $4f$ multiplet split by the CEF effect. In particular, a characteristic minimum is expected at the temperature of around $\Delta/2$ K if one sets a following level scheme: $\Gamma_7(0) - \Gamma_8(\Delta K)$ with $J = 5/2$ in the temperature dependence of $(C_{11} - C_{12})/2$, whereas it is not expected in that of C_{44}, based on the linear quadrupolar response of the system to an external strain in CEF state. The details are referred to in Refs. 15–17. Figure 9 shows the calculated quadrupolar susceptibility $-\chi_{\Gamma3}^{(s)}(T)$ and $-\chi_{\Gamma5}^{(s)}(T)$ taking account of the proposed CEF level scheme, which corresponds to the elastic constants $(C_{11} - C_{12})/2$ and C_{44}, respectively. It is apparent that the low-temperature properties can be far from described qualitatively by a well-localized Ce-ion picture in both systems of $CeOs_4Sb_{12}$ and $CeRu_4Sb_{12}$, comparing the experimental results (Figs. 1 and 3) and theoretical ones (Fig. 9). With this as background we turn to an account of the observed elastic anomalies in both systems.

Here, we discuss the slight increase structure appearing at around 130 K in the temperature dependence of the elastic constants in $CeRu_4Sb_{12}$. As mentioned in the Introduction, anomalous behavior was observed in the temperature dependence of the resistivity and magnetic susceptibility. The magnetic susceptibility begins to deviate significantly from the Curie-Weiss (CW) law expected in a trivalent Ce ion state
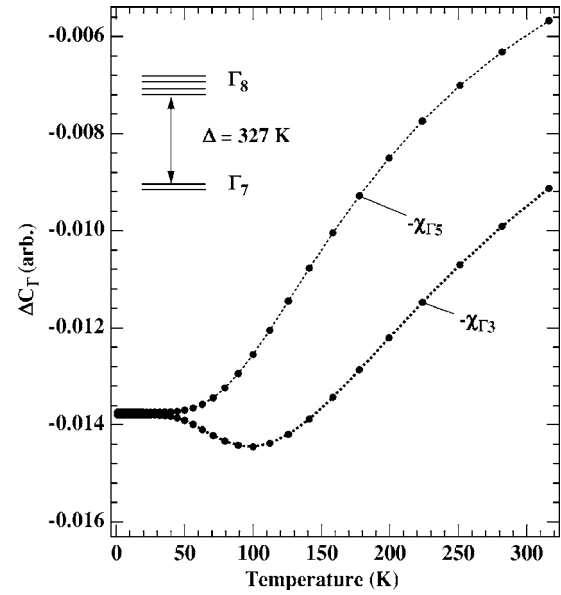

FIG. 9. Theoretical results of the temperature dependence of the quadrupolar susceptibilities $-\chi_{\Gamma3}^{(s)}(T)$ and $-\chi_{\Gamma5}^{(s)}(T)$ for $CeOs_4Sb_{12}$, belonging to the $(C_{11} - C_{12})/2$ and C_{44}, respectively.

below 150 K.[8,9] The resistivity shows a change of the slope below around 150 K, followed by a steep decrease below 80 K. The observed slight increase seems to be concerned closely with the anomaly in the magnetic susceptibility and resistivity. Specifically, the slight increase of elastic constants is most likely related to the onset of the deviation of the magnetic susceptibility from the CW law. The anomalous behavior lets us conjecture an intermediate-valence (IV) compound. The slight increase of the elastic constants may suggest that volume instability due to an unstable ionic state of the Ce ions takes place around 150 K in $CeRu_4Sb_{12}$. Hence it is reasonable to suggest that a lack of the elastic anomaly expected by the CEF effect may be attributed to the intermediate-valence $4f$ system at low temperature in $CeRu_4Sb_{12}$.

Next, we discuss the elastic anomalies observed in $CeOs_4Sb_{12}$. We need to postulate a different way except for the CEF effect to explain the lattice softening toward low temperature observed below 15 K in $C_{11} - T$. As mentioned in the Introduction, the dense Kondo system with the pseudoenergy gap is realized, probably due to the strong hybridization between Ce $4f$ states and conduction electron states in $CeOs_4Sb_{12}$.[7,12,14] In such a system, the deformation potential coupling to the conduction bands with the pseudogap formed around the Fermi level E_F can be regarded as significant.[15,18,19] Here, we adopt this model as applied to the typical valence fluctuation systems such as SmB_6, $CeNiSn$, and $CeRhIn$.[19,21] This essential coupling may be described by

$$E_k = E_k^0 + g_{k\Gamma}\varepsilon_\Gamma + (h_{k\Gamma})^2 \varepsilon_\Gamma^2 / \Delta_k, \qquad (1)$$

where E_k, g_k, and h_k denote the energy of the upper or lower quasiparticle band, the first and the second deformation potential coupling constants, respectively. Δ_k denotes the band gap at the Fermi level. Then, the free energy of the conduction electrons is described as follows:
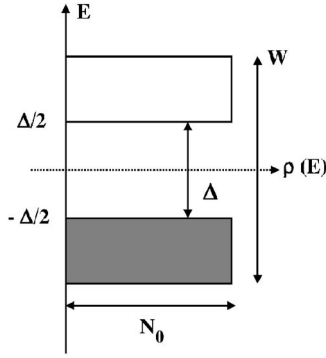
FIG. 10. Schematic representation of the densities of the $4f$-electronic state for $CeOs_4Sb_{12}$ symmetrically lying above and below E_F.

$$F_{el} = n\zeta - k_B T \sum_k \ln\left[1 + \exp\left(\frac{\zeta - E_k}{k_B T}\right)\right]. \qquad (2)$$

Here, n denotes the number of the conduction electrons in the quasiparticle band and ζ is the chemical potential. From the second derivative of the free energy with respect to the elastic strain ε_Γ, one obtains an analogous expression for the elastic constants as in the case of magnetoelastic interaction with Fermi-Dirac distribution function $f_k = 1 + \exp[(\zeta - E_k)/k_B T]^{-1}$ as follows:

$$C_\Gamma(T) = \frac{\partial^2 F}{\partial \varepsilon_\Gamma^2} = C_\Gamma^0 + \sum_k \frac{\partial^2 E_k}{\partial \varepsilon_\Gamma^2} f_k - \frac{1}{k_B T} \sum_k \left(\frac{\partial E_k}{\partial \varepsilon_\Gamma}\right)^2 f_k (1 - f_k)$$

$$+ \frac{1}{k_B T} \frac{\left[\sum_k \frac{\partial E_k}{\partial \varepsilon_\Gamma} f_k (1 - f_k)\right]^2}{\sum_k f_k (1 - f_k)}. \qquad (3)$$

The second, third, and fourth terms of Eq. (3) represent the Van Vleck and the Curie terms, respectively. C_Γ^0 in Eq. (3) is the background elastic constant without the contribution of the f electrons. Variation in C_Γ^0 originates mainly from anharmonic effects of the crystal. This formalism is very similar to that of the strain susceptibility of the localized $4f$-electron system without the Fermi distribution function f_k. Here, the conservation law of the total number of electrons is employed in the quasiparticle band. For simplicity, the simple quasiparticle band model is assumed, in which the dispersion of band energy is neglected and the Fermi level ε_F is located at the middle of energy gap Δ. In addition, g_k and Δ_k are independent of the wave vector k. Furthermore, the rectangular density of states N_0 for the upper and lower bands with the same bandwidth W were introduced. The present model is illustrated in Fig. 10. It is noted that a shift of the chemical potential is not expected with changing the temperature for this symmetric two-band model.

We have analyzed the lattice softening with Eq. (3) under the above-mentioned conditions. The fitting results are shown by the dotted lines in Fig. 11. Here, the background elastic constant C_Γ^0 was assumed to have the linear temperature dependence. The obtained parameter was summarized in Table II. The estimated value of the band gap Δ is in good agreement with that determined by the electrical resistivity
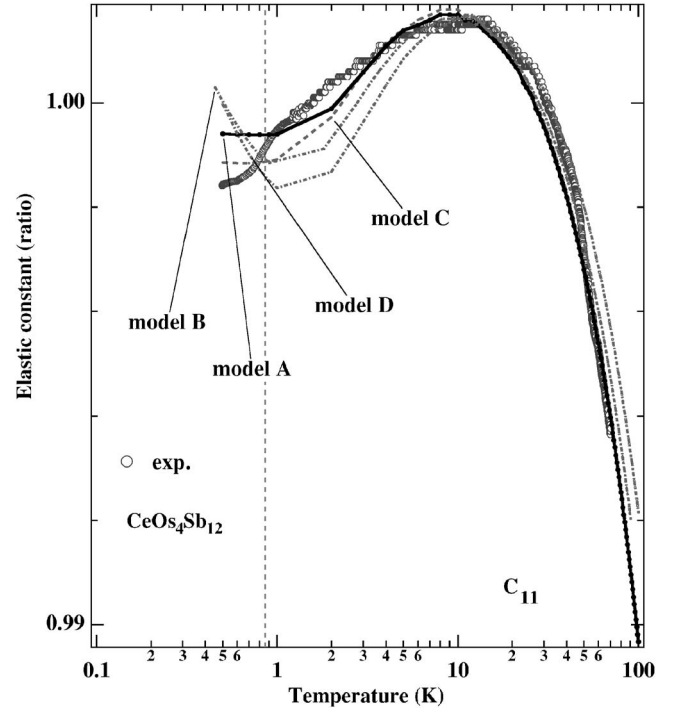

FIG. 11. Theoretical results of the temperature dependence of the elastic constant C_{11} based on the deformation potential using Eq. (3).

measurement.[12] This value is, however, quite small compared with that determined by the optical conductivity measurement.[22] In the latter case, it is almost of ten times in magnitude. This inconsistency may be ascribed to the complicated band structure in the vicinity of Fermi level E_F, and that the band gap has the strong temperature dependence as generally expected in conventional dense Kondo semiconductors and semimetals. The multiple formation of the energy gap may be realized in $CeOs_4Sb_{12}$, which is developing with decreasing the temperature.

The absence of a characteristic softening in C_{44}, in contrast to that in C_{11}, can be accounted for qualitatively in terms of the selective connection between the elastic softening and the Fermi surface from the view of their symmetry in $CeOs_4Sb_{12}$, deduced by that of the reference system $LaOs_4Sb_{12}$. According to de Haas–van Alphen experiments on $LaOs_4Sb_{12}$ and the corresponding band theory, the Fermi surface consists of a closed hole surface derived from the 47th band centered at the Γ point, and closed and multiply connected hole surfaces from the 48th band.[23,24] In particular, the multiply connected hole surface is relevant to the

TABLE II. Obtained parameters deduced from the present results as shown in Fig. 11.

Model	W	Δ
A	30 K	5 K
B	30 K	6 K
C	30 K	6 K
D	29 K	5.5 K

heavy electrons through the correlation effect, whose main parts are centered at the N point. Furthermore, the Fermi surface is missing in the direction of $\langle 100 \rangle$ and $\langle 111 \rangle$, and their equivalent. Hence it can be presumed that these multiply connected hole surfaces have the strong deformation coupling with the relevant elastic strain induced by a sound wave. If we set a charge density located at the N point, the character for them is decomposed into a direct sum of the irreducible representations as $\Gamma_1 \oplus \Gamma_3 \oplus \Gamma_4$. There can be a charge fluctuation mode with Γ_3 in consequence, which possibly couples to the elastic strains $\varepsilon_u = (2\varepsilon_{zz} - \varepsilon_{xx} - \varepsilon_{yy})$ or $\varepsilon_v = (\varepsilon_{xx} - \varepsilon_{yy})$ with Γ_3 symmetry, resulting in a lattice softening in $(C_{11} - C_{12})/2$. It should be noted that the lack of Γ_5 representation in the decomposition, to the contrary, would be indicative for the absence of the coupling to the elastic strains ε_{yz}, ε_{zx}, and ε_{xy}, resulting in the absence of the softening in C_{44}. These theoretical predictions can capture reasonably the experimental observations based on the Fermi surface topology. Again, this fact points to the significant importance of the deformation potential coupling in $CeOs_4Sb_{12}$. Unlike the results, no characteristic lattice softening was observed in $CeRu_4Sb_{12}$ which shows metallic behavior at low temperatures. The different manner in the temperature dependence of the elastic constants between $CeOs_4Sb_{12}$ and $CeRu_4Sb_{12}$ may provide evidence of the gap formation in $CeOs_4Sb_{12}$. The different band structure in the vicinity of the Fermi level would govern the observed elastic anomalies, as consistent with our aforementioned conjecture.

Next, we would like to make a comment on the phase transition at T_s in $CeOs_4Sb_{12}$. The most prominent feature is the absence of a clear cusp at T_s in the temperature dependence of the elastic constants. $CeOs_4Sb_{12}$ shows a steep drop, followed by a smooth bending at T_s in $C_{11} - T$ and $C_{44} - T$. This feature contrasts strikingly with a clear cusp as observed frequently in a well-localized $4f$ electron system such as the isostructural system $NdFe_4P_{12}$ which shows a ferromagnetic ordering at $T_c = 1.9$ K.[25] Interestingly, the very similar behavior was observed in some other filled skutterudite compounds such as $Pr_{1-x}La_xFe_4P_{12}$ being for $x = 0.05$ and 0.15, and $SmFe_4P_{12}$.[26,27] They all commonly exhibit a ferromagnetic ordering accompanied by releasing an extremely tiny entropy below the transition temperature in which the heavily renormalized quasiparticle bands are considered to be formed. Hence one can speculate that this transition might be attributed to instability of the Fermi surface such as spindensity wave, although the clear interpretation is still an open question. Nevertheless, it should be noted the markedly difference of the elastic behavior around T_s as opposed to that of a well-localized $4f$ electron system. To clarify the order parameter in the ordered phase, microscopic experiments are highly required.

Finally, we discuss the slight increase observed in the temperature dependence of C_{11} and C_{44} around 30 K in $CeOs_4Sb_{12}$. Since the reproducibility cannot be confirmed satisfactorily, it is difficult to conclude at present whether the observed anomaly is intrinsic or not. No notable anomaly, in fact, has been reported around this temperature in the other measurements thus far. We suppose that it may arise from instability of Ce ions in a highly anharmonic potential formed by a weak coupling of Ce ion in a cage of Sb icosahedron, commonly called "rattling." The characteristic increase lets us conjecture that observed in $PrOs_4Sb_{12}$ where a clear increase and the clear frequency dependence of the elastic constant were observed.[28] Such an elastic anomaly, probably due to the rattling motion, has been observed especially in the systems with a poly-anion Os_4Sb_{12}.[29] A relatively large size of a cage of Sb icosahedron might yield a condition for the rattling motion.[28] $CeOs_4Sb_{12}$ can be thus one of the excellent candidates. Whether this anomaly is intrinsic or not, and, if intrinsic, the origin is still open to question. Further experiments including the frequency dependence of ultrasonic measurements with a larger size of the single crystal are strongly requisite.

V. CONCLUDING REMARKS

We have studied the elastic properties of the non-Fermi-liquid metal $CeRu_4Sb_{12}$ and the dense Kondo semiconductor $CeOs_4Sb_{12}$ by ultrasonic measurements. A remarkable difference of the elastic behavior between both systems was found experimentally at low temperatures. The difference is likely to originate mainly from the renormalized band structure in the vicinity of the Fermi level. Failing to explain the present data by the CEF level scheme is indicative of the instability of the trivalent Ce state at low temperature in both systems, probably attributed to the predominant hybridization process to be between f^1 and f^0 configurations. The remarkable elastic softening in $CeOs_4Sb_{12}$ can be explained reasonably by the characteristic response of the deformation potential coupling to a pseudogapped band in the vicinity of the Fermi level. The steep drop of the elastic constants relevant to the phase transition in $CeOs_4Sb_{12}$ is greatly different from a cusp behavior as commonly observed at a magnetic transition due to the well-localized $4f$ electronic system. Although this phase is not understood clearly, the behavior lets us conjecture a weak magnetic ordering originating from the heavily renormalized quasiparticles. This is an open issue. Further experiments, in particular, microscopic experiments, are highly desirable to check our proposal and to address the temperature evolution of the ground state in $CeRu_4Sb_{12}$ and $CeOs_4Sb_{12}$.

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