Quadrupolar effect and rattling motion in heavy fermion superconductor PrOs$_4$Sb$_{12}$

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The elastic properties of a filled skutterudite PrOs$_4$Sb$_{12}$ with a heavy Fermion superconductivity at $T_C = 1.85$ K have been investigated. The elastic softening of $(C_{11} - C_{12})/2$ and $C_{44}$ with lowering temperature down to $T_C$ indicates that the quadrupolar fluctuation due to the CEF state plays a role for the Cooper paring in superconducting phase of PrOs$_4$Sb$_{12}$. A Debye-type dispersion in the elastic constants around 30 K revealed a thermally activated $\Gamma_{23}$ rattling due to the off-center Pr-atom motion obeying $\tau = \tau_0 \exp(E/k_B T)$ with an attempt time $\tau_0 = 8.8 \times 10^{-11}$ sec and an activation energy $E = 168$ K. It is remarkable that the charge fluctuation of the off-center motion with $\Gamma_{23}$ symmetry may mix with the quadrupolar fluctuation and enhance the elastic softening of $(C_{11} - C_{12})/2$ just above $T_C$.

**Keywords:** PrOs$_4$Sb$_{12}$, heavy fermion superconductor, quadrupolar effect, rattling motion

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The rare-earth cubic compounds based on Pr$^{3+}$ ions have received much attention because various unusual properties are expected at low temperatures. The system with a non-Kramers $\Gamma_3$ doublet possessing two quadrupoles $O^0_2 = (2J_x^2 - J_y^2 - J_z^2)/\sqrt{3}$ and $O^2_2 = J_y^2 - J_z^2$ favors a quadrupole ordering. We refer the $\Gamma_3$ ground state systems as a metallic compound PrPb$_3$ showing the antiferro-quadrupole ordering at $T_Q = 0.4$ K and a semiconductor PrPtBi undergoing ferro-quadrupole ordering at $T_Q = 1.2$ K. PrSb is known as a singlet ground state system. The elastic constant $(C_{11} - C_{12})/2$ is responsible for the quadrupolar susceptibility of $O^0_2$ and $O^2_2$ with $\Gamma_3$ symmetry, while $C_{44}$ is for the susceptibility of $O_{yx} = J_y J_z + J_z J_y$, $O_{xx} = J_x J_z + J_z J_x$, $O_{xy} = J_x J_y + J_y J_x$ with $\Gamma_5$ symmetry. The softening of $(C_{11} - C_{12})/2$ and $C_{44}$ is a useful prove to clarify the quadrupolar effects of Pr-based compounds.

Recently, Bauer et al. have found a new-type of the heavy Fermion superconductor in a filled skutterudite PrOs$_4$Sb$_{12}$ with space group $T_h^5$ (Im$\bar{3}$m). The heavy Fermion state with a large specific heat coefficient $\gamma = 750$ mJ/mol-K$^2$ of PrOs$_4$Sb$_{12}$ exhibits the superconducting transition at $T_C = 1.85$ K associated with a large jump $\Delta C/T_C \sim 500$ mJ/mol-K$^2$. A sign of the double transition in the specific heat has been found. The thermal transport measurement in fields suggests the two distinct superconducting phases in PrOs$_4$Sb$_{12}$. The nuclear spin relaxation rate $1/T_1$ of Sb indicates unconventional superconductivity possessing neither a coherence peak nor a $T^3$-power law. The muon spin relaxation in PrOs$_4$Sb$_{12}$ yields a penetration depth indicating a new-type of energy gap. The odd-parity Cooper pairing mediated by the quadrupole fluctuation is argued as unconventional heavy Fermion superconductivity in PrOs$_4$Sb$_{12}$. Because the magnetic susceptibility is rather silent to distinguish non-magnetic $\Gamma_{23}$ doublet from $\Gamma_1$ singlet, it has not been settled whether the CEF ground state of PrOs$_4$Sb$_{12}$ is $\Gamma_3$ doublet or $\Gamma_1$ singlet. The measurement of $(C_{11} - C_{12})/2$ and $C_{44}$ responsible for the quadrupolar susceptibility in PrOs$_4$Sb$_{12}$ is a central issue to clarify the CEF state and the interplay of the quadrupolar fluctuation to the superconductivity in PrOs$_4$Sb$_{12}$.

The reduction of the thermal conductivity in filled skutterudites RM$_4$Sb$_{12}$ (R: La or Ce. M: Fe or Co) is caused by a rattling motion due to a weakly bounded rare-earth ion in an oversized cage of Sb-icosahedron. The filled skutterudites with the cage are favorable for the thermoelectric device possessing a high coefficient of merit. The ultrasonic measurements are generally used to observe the rattling motion or off-center tunneling motion. We refer the rattling motion in clathrate materials Sr$_5$Ga$_4$Ge$_{13}$ and Ce$_3$Pd$_{20}$Ge$_{34}$, and an off-center tunneling of OH-ion doped in NaCo$_{20}$Ge$_{34}$, respectively. Our experiment on $C_{44}$ in La$_3$Pd$_{20}$Ge$_{34}$ revealed the ultrasonic dispersion around 20 K due to the rattling motion and elastic softening below 3 K due to off-center tunneling motion in cage. It became to be sure that the rattling and tunneling are common features of the clathrate compound with oversized cage. Quite recently a small off-center displacement of Pr-ion in PrOs$_4$Sb$_{12}$ has been observed by X-ray absorption measurements. The rattling motion in the clathrate compound PrOs$_4$Sb$_{12}$ with the heavy Fermion superconductivity has not been clarified yet.

The single crystal of PrOs$_4$Sb$_{12}$ with a length of 1.2 mm along the [110] direction for the present ultrasonic measurements was grown by a flux method. The ul-
below 20 K down to superconducting point. The Pr-ion off-center motion. The softening of (C11-C12)/2 of PrOs4Sb12 due to the quadrupolar fluctuation of the CEF states. The elastic constant C of PrOs4Sb12 was obtained by the transverse wave of 17 MHz propagating along k = [110] with polarization u = [110]. This (C11-C12)/2 mode is associated with the elastic strain εv = εxx - εyy. The increase of (C11-C12)/2 around 30 K of Fig. 1 originates from the Debye-type dispersion, where the ultrasonic wave frequency ω coincides with a relaxation time τ of the system as ωτ = 1. A relatively large lattice parameter a=0.930311 nm in PrOs4Sb12 may lead to the rattling motion of an off-center Pr-ion in an oversized cage of Sb-icosahedron. The resonant scattering of the ultrasonic wave by the rattling motion of Pr-ion over a potential hill brings about the Debye-type dispersion. The experimental determination of a relaxation time τ of the rattling is discussed latter.

A remarkable softening of (C11-C12)/2 below 20 K in Fig. 1 has been found with decreasing temperature. As shown in inset of Fig. 1, the softening of (C11-C12)/2 turns up around the superconducting transition TC = 1.85 K. The quadrupole-strain interaction, $H_{Q8} = \sum_i \sum_{\gamma} g_{\gamma}^{Q} \Omega_{\gamma}^{Q}(i) \varepsilon_{\gamma i}$, and the inter-site quadrupole interaction, $H_{QQ} = -\sum_i g_{\gamma}^{Q} \Omega_{\gamma}^{Q}(i) \varepsilon_{\gamma i}$, give rise to the elastic softening as $C_T = C_0^Q - N g_0^Q \chi_Q / (1 - g_0^Q \chi_Q)^2$. Here $\chi_Q$ is a quadrupolar susceptibility consisting of the Curie-term for diagonal parts and the Van Vleck-term for off-diagonal ones. The coupling of the quadrupole $O_2$ with $\Gamma_3$ symmetry to the elastic strain $\varepsilon_{\gamma i}$ is relevant for the softening in (C11-C12)/2 below 20 K of PrOs4Sb12.

The CEF Hamiltonian for the Pr3+ ion with the site symmetry $T_h$ is written as $H_{CEF} = B_2^O O_2 + B_4^O O_4 + B_6^O O_6$, where $O_4 = O_4^7 + 5O_4^8$, $O_6 = O_6^7 - 21O_6^8$, and $O_6 = O_6^{12} - O_6^{15}$. Two different types of the CEF models of $\Gamma_3 = \Gamma_4^{(2)}$ and $\Gamma_1 = \Gamma_4^{(2)}$ for PrOs4Sb12 have been proposed so far. The solid line in Fig. 1 is based on the doublet model of $\Gamma_3 = \Gamma_4^{(2)}$ and $\Gamma_1 = \Gamma_4^{(2)}$, and the off-center motion of Pr-ion over a potential hill brings about the Debye-type dispersion. The doublet model seems to be favorable for the softening of (C11-C12)/2.

Recently, Kohgi et al. have proposed a singlet ground state CEF model of $\Gamma_1 (0 K)$, $\Gamma_4^{(2)} (7.9 K)$, $\Gamma_4^{(2)} (135 K)$, and $\Gamma_2 (205 K)$ with $B_2^O = 2.37 \times 10^{-2} K$, $B_6^O = 1.32 \times 10^{-3} K$, $B_6^O = 1.08 \times 10^{-2} K$. The dashed line for $|\gamma_{\Gamma_4} = 79 K$ and $g_{\gamma} = 0.22 K$ based on the singlet model also reproduces the softening of (C11-C12)/2 except for a small deviation below a minimum around 3.5 K in the fitting. The off-center motion Pr ion with $\Gamma_3$-symmetry may enhance the elastic softening of (C11-C12)/2 just above $T_C$, that will considerably renormalize the one-ion susceptibility of the dashed line in Fig. 1. Even in the case of the $\Gamma_4^{(2)}$ model, the charge fluctuation due to the off-center motion may reproduce the softening of (C11-C12)/2 just above $T_C$ proportional to reciprocal temperature. In order to set the alternative CEF model of $\Gamma_3 = \Gamma_4^{(2)}$ or $\Gamma_1 = \Gamma_4^{(2)}$, further experiments are necessary. The Debye-type dispersion was employed to reproduce the anomaly around 20 K of solid and dashed lines in Fig. 1. We discuss this point again in Fig. 3.

In Fig. 2, we show temperature dependence of $C_{44}$ obtained by the transverse wave propagating along [110] with polarization along [001]. The softening of $C_{44}$ below 60 K is described in terms of the quadrupolar susceptibility for the $\Gamma_4^{(2)}$-type quadrupole. The solid line in Fig. 2 is responsible for the $\Gamma_3 = \Gamma_4^{(2)}$ model with parameters $|\gamma_{\Gamma_4}| = 34.4 K$ and $g_{\gamma} = -0.002 K$. The dashed line is a fit for the $\Gamma_1 = \Gamma_4^{(2)}$ model with $|\gamma_{\Gamma_4}| = 70 K$ and $g_{\gamma} = -0.07 K$. Because the quadrupolar susceptibility of $C_{44}$ for both models is dominated by the Van Vleck term responsible for off-diagonal processes, the determination of the CEF state by $C_{44}$ is rather indirect as similar as the magnetic susceptibility. It should be noted that no sign of the ultrasonic dispersion has been found in $C_{44}$.
Figure 3 represents $C_L = (C_{11} + C_{12} + 2C_{44})/2$ of PrOs$_4$Sb$_{12}$ obtained by the longitudinal wave along the [110] direction. A remarkable frequency dependence of 17.8 MHz, 52.0 MHz and 87.7 MHz in Fig. 3 is described in terms of the Debye-type dispersion as, $C_L(\omega) = C_L(\infty) - \left\{C_L(\infty) - C_L(0)\right\}/(1 + \omega^2\tau^2)$. Here $\omega$ is the angular frequencies of the ultrasonic wave. Arrows in Fig. 3 indicate the temperature being the resonant condition of $\omega\tau = 1$. The anomaly of $(C_{11} - C_{12})/2$ around 30 K is also well described by the Debye dispersion of the solid and dashed lines in Fig. 1.

In inset of Fig. 3 the temperature dependence of the relaxation time $\tau$ obtained by $C_L$ is presented together with the results of $(C_{11} - C_{12})/2$ of Fig. 1 and 11, that is not presented here. The relaxation time due to the rattling motion obeys the temperature dependence of $\tau = \tau_0 \exp(E/k_B T)$ with an attempt time $\tau_0 = 8.8 \times 10^{-11}$ sec and an activation energy $E = 168$ K. Utilizing a harmonic oscillation of $\zeta(z) = (1/\pi \tau_0)^{1/2} \exp(-z^2/2\tau_0^2)$, we estimated a mean square displacement $z_0 = (1/2\pi)(h\tau_0/M)^{1/2}$ = 0.079 nm for Pr-ion in the present potential of the cage. This result is comparable to $z_0 = 0.048$ nm of CePd$_{120}$Ge$_{34}$ and $z_0 = 0.012$ nm of La$_2$Pd$_{220}$Ge$_{34}$.

The twelve Sb-atoms have a distance 0.3542 nm from the center of the cage. Because the Sb-atom is absent along the [100] axis, the Pr-ion may favor an off-center motion over six minimum points of potential at $r_1 = (a, 0, 0)$, $r_2 = (-a, 0, 0)$, $r_3 = (0, a, 0)$, $r_4 = (0, -a, 0)$, $r_5 = (0, 0, a)$, $r_6 = (0, 0, -a)$. Here, the mean square displacement of Pr-atom extends over the potential minima as $a \sim z_0/2 = 0.04$ nm. On the other hand, the Os-atom locating at 0.4028 nm from the center of the cage along the three-fold [111] axis prevents the off-center motion along the [111] axis. The Sb-atoms closely locating to the two-fold [110] axis may hinder the off-center motion along the [110] axis.

The ultrasonic dispersion has been observed in the transverse wave of $(C_{11} - C_{12})/2$ of the strain $\varepsilon_z$ with $\Gamma_{23}$ symmetry. The longitudinal modes of $C_{11}$ and $C_L$ consisting of the strain $\varepsilon_u = (2\varepsilon_{zz} - \varepsilon_{xx} - \varepsilon_{yy})/\sqrt{3}$ with $\Gamma_{23}$ symmetry in part also show ultrasonic dispersion. On the other hand, the $C_{44}$ mode responsible for the elastic strain with $\Gamma_{4}^{(2)}$ symmetry does not show the dispersion effect. These results indicate that the thermally activated rattling motion being coupled to the elastic strains $\varepsilon_u$ and $\varepsilon_v$ has the $\Gamma_{23}$ symmetry. It is of particular importance to project out the off-center mode for the irreducible representations at a center of the cage with the point group symmetry $T_{h}^{23}$. Operating a symmetry element $R$ of $T_h$ on the atomic density $\rho_i = \rho(\rho_i)$ at the minimum point $\rho_i (i = 1, 2, \cdots, 6)$, one obtains six dimensional representation matrices $D_{ij}(R)$. The character $\chi(R)$ being a trace of the representation matrix reduces to direct sum $\Gamma_1 \oplus \Gamma_{23} \oplus \Gamma_{4}^{(2)}$. The projection operator is used to pick up the $\Gamma_1$, $\Gamma_{23}$ and $\Gamma_{4}^{(2)}$ representations consisting of the fractional atomic density of Pr-ion over the six minimum points. In the present case of PrOs$_4$Sb$_{12}$, the $\Gamma_{23}$ off-center mode of $\rho_{\Gamma_{23}u} = 2\rho_5 + 2\rho_6 - \rho_1 - \rho_2 - \rho_3 - \rho_4$ and $\rho_{\Gamma_{23}v} = \rho_1 + \rho_2 - \rho_3 - \rho_4$ with the fractional atomic distribution in Fig. 4 is the ground state of the system. $\rho_{\Gamma_{23}v}$ means the distribution of fraction 1/2 at $\rho_5$. 

FIG. 2: Temperature dependence of the elastic constant $C_{44}$ of PrOs$_4$Sb$_{12}$ measured by ultrasonic wave of 74 MHz. The softening of below 60 K down to superconducting point $T_C = 1.85$ K is described in terms of the quadrupolar susceptibility $\chi_Q$ of solid line for $\Gamma_{23}$-$\Gamma_{4}^{(2)}$ model and dashed line for $\Gamma_1$-$\Gamma_{4}^{(2)}$ model. Inset shows the detail around $T_C$.

FIG. 3: Temperature dependence of $C_L = (C_{11} + C_{12} + 2C_{44})/2$ in PrOs$_4$Sb$_{12}$ measured by ultrasonic waves of 17.8, 52.0 and 87.7 MHz. Arrows indicate the temperatures that the relaxation time $\tau$ of Pr-ion rattling coincides with the sound wave frequencies $\omega$ as $\omega\tau = 1$. Inset shows temperature dependence of relaxation time.
FIG. 4: The $\Gamma_{23}$ rattling mode of $\rho_{23a} = 2\rho_5 + 2\rho_6 - \rho_1 - \rho_2 - \rho_3 - \rho_4$ and $\rho_{23v} = \rho_1 + \rho_2 - \rho_3 - \rho_4$, being responsible for the ultrasonic dispersion in PrOs$_4$Sb$_{12}$.

and $\rho_6$ sites and null at $\rho_1, \rho_2, \rho_3, \rho_4$. And $\rho_{23v}$ is responsible for fraction 1/3 at $\rho_1$ and $\rho_2$, 1/6 at $\rho_5$ and $\rho_6$, and null at $\rho_3$ and $\rho_4$. The total symmetric mode $\rho_{14} = \rho_1 + \rho_2 + \rho_3 + \rho_4 + \rho_5 + \rho_6$ and the polar mode $\rho_{14} = \rho_1 - \rho_2, \rho_{4v} = \rho_3 - \rho_4, \rho_{4v} = \rho_5 - \rho_6$ may correspond to the excited states. $\rho_1$, represents the mean fraction 1/6 over the six sites. $\rho_{23v}$, for instance, has fraction 1/3 at $\rho_1$ and null at $\rho_2$, and fraction 1/6 at $\rho_3, \rho_4, \rho_5, \rho_6$.

Recent ultrasonic measurements on the clathrate compound La$_3$Pd$_{20}$Ge$_6$ by our group has successfully showed the rattling and tunneling motions of an off-center La ion in cage[7]. In the present clathrate compound of PrOs$_4$Sb$_{12}$ with a cage of Sb-icosahedron, the thermally activated rattling motion over the potential hill brings about the ultrasonic dispersion with the relaxation time $\tau$ in inset of Fig. 3. With lowering temperature, the thermally activated rattling dies out completely without showing the structural transition. Consequently, the tunneling motion of Pr-ion through the hill in keeping the site symmetry to be T$_h$ is relevant at low temperatures. The tunneling motion being accompanied by charge fluctuation interacts with conduction electrons through the channels of Pr-Sb bonding. It is remarkable that the off-center tunneling motion with $\Gamma_{23}$ symmetry may bring about the enhancement of the elastic softening in $(C_{11} - C_{12})/2$ just above $T_C$. The theoretical work for the interplay of the tunneling motion to the superconductivity by Cox and Zawadowski may be relevant for the present PrOs$_4$Sb$_{12}$.

In conclusion we have successfully observed the elastic softening in $(C_{11} - C_{12})/2$ and $C_{44}$ above $T_C$. It is, however, still difficult to determine a CEF state in the alternative model of $\Gamma_{23}$-$\Gamma_{44}^{(2)}$ or $\Gamma_1$-$\Gamma_4^{(2)}$. The field dependence of the elastic constant in particular is necessary to settle the CEF state of the system. Nevertheless, it is worthwhile to emphasize the fact that the softening of $(C_{11} - C_{12})/2$ and $C_{44}$ indicate a crucial role of the quadrupolar fluctuation to the heavy Fermion superconductivity in PrOs$_4$Sb$_{12}$. Furthermore, the ultrasonic dispersion due to the $\Gamma_{23}$ rattling motion with activation energy $E = 168$ K has been found. The $\Gamma_{23}$-type charge fluctuation associated with the off-center tunneling motion in particular may enhance the elastic softening of $(C_{11} - C_{12})/2$ just above $T_C$. The more accurate investigation is necessary to clarify the CEF state in PrOs$_4$Sb$_{12}$ and the interplay of the quadrupole fluctuation and the off-center motion of Pr-ion to the unconventional superconductivity.

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