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Quadrupolar effect and rattling motion in the heavy-fermion superconductor $PrOs_4Sb_{12}$

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The elastic properties of a filled skutterudite $PrOs_4Sb_{12}$ 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 $PrOs_4Sb_{12}$. A Debye-type dispersion in the elastic constants around 30 K revealed a thermally activated Γ_{23} rattling due to the off-center Pr-atom motion obeying $\tau = \tau_0 \exp(E/k_BT)$ with an attempt time $\tau_0 = 8.8\times10^{-11}$ s and an activation energy $E = 168$ K. It is remarkable that the charge fluctuation of the off-center motion with Γ_{23} symmetry may mix with the quadrupolar fluctuation and enhance the elastic softening of $(C_{11}-C_{12})/2$ just above T_C.

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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 Γ_3 doublet possessing two quadrupoles $O_2^0 = (2J_z^2 - J_x^2 - J_y^2)/\sqrt{3}$ and $O_2^2 = J_x^2 - J_y^2$ favors a quadrupole ordering. We refer the Γ_3 ground state systems as a metallic compound $PrPb_3$ showing the antiferro-quadrupole ordering at $T_Q = 0.4$ K[1] and a semiconductor PrPtBi undergoing ferro-quadrupole ordering at $T_Q = 1.2$ K.[2] PrSb is known as a singlet ground state system.[3] The elastic constant $(C_{11} - C_{12})/2$ is responsible for the quadrupolar susceptibility of O_2^0 and O_2^2 with Γ_3 symmetry, while C_{44} is for the susceptibility of $O_{yz} = J_yJ_z + J_zJ_y$, $O_{zx} = J_zJ_x + J_xJ_z$, $O_{xy} = J_xJ_y + J_yJ_x$ with Γ_5 symmetry. The softening of $(C_{11}-C_{12})/2$ and C_{44} is useful proof 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 $PrOs_4Sb_{12}$ with space group T_h^5 $(Im\overline{3})$.[4] The heavy fermion state with a large specific heat coefficient $\gamma = 750$ mJ/mol K^2 of $PrOs_4Sb_{12}$ 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.[5] The thermal transport measurement in fields suggests the two distinct superconducting phases in $PrOs_4Sb_{12}$.[6] The nuclear spin relaxation rate $1/T_1$ of Sb indicates unconventional superconductivity possessing neither a coherence peak nor a T^3-power law.[7] The muon spin relaxation in $PrOs_4Sb_{12}$ yields a penetration depth indicating a new type of energy gap.[8] The odd-parity Cooper pairing mediated by the quadrupole fluctuation is argued as unconventional heavy-fermion superconductivity in $PrOs_4Sb_{12}$.[9] Because the magnetic susceptibility is rather silent to distinguish non-magnetic Γ_{23} doublet from Γ_1 singlet, it has not been settled whether the CEF ground state of $PrOs_4Sb_{12}$ is Γ_{23} doublet or Γ_1 singlet.[4,10] The measurement of $(C_{11}-C_{12})/2$ and C_{44} responsible for the quadrupolar susceptibility in $PrOs_4Sb_{12}$ is a central issue to clarify the CEF state and the interplay of the quadrupolar fluctuation to the superconductivity in $PrOs_4Sb_{12}$.

The reduction of the thermal conductivity in filled skutterudites RM_4Sb_{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.[11] The filled skutterudites with the cage are favorable for the thermoelectric device possessing a high coefficient of merit.[12] The ultrasonic measurements are generally useful to observe the rattling motion or off-center tunneling motion. We refer the rattling motion in clathrate materials $Sr_8Ga_{16}Ge_{30}$ (Ref. 13) and $Ce_3Pd_{20}Ge_6$,[14] and an off-center tunneling of OH-ion doped in NaCl.[15,16] Recently, our experiment on C_{44} in $La_3Pd_{20}Ge_6$ revealed the ultrasonic dispersion around 20 K due to rattling motion and elastic softening below 3 K due to off-center tunneling motion in cage.[17] It came to be seen that rattling and tunneling are common features of the clathrate compound with oversized cage. Quite recently a small off-center displacement of Pr ion in $PrOs_4Sb_{12}$ has been observed by x-ray absorption measurements.[18] The rattling motion in the clathrate compound $PrOs_4Sb_{12}$ with the heavy-fermion superconductivity has not been clarified yet.

The single crystal of $PrOs_4Sb_{12}$ with a length of 1.2 mm along the [110] direction for the present ultrasonic measurements was grown by a flux method. The ultrasonic velocity v was detected by a phase comparator based on a mixer technology. The piezoelectric $LiNbO_3$ transducers of x-cut and 36° y-cut were used for the measurements of transverse and longitudinal ultrasonic waves, respectively. The elastic constant $C = \rho v^2$ of $PrOs_4Sb_{12}$ with a lattice constant $a = 0.930311$ nm was estimated by the density $\rho = 9.75$ g/cm^3. A ^{3}He-evaporation fridge down to 500 mK was employed.

Figure 1 shows temperature dependence of $(C_{11}-C_{12})/2$ of $PrOs_4Sb_{12}$, which was obtained by the transverse wave of 17 MHz propagating along $\boldsymbol{k} = [110]$ with polarization $\boldsymbol{u}$

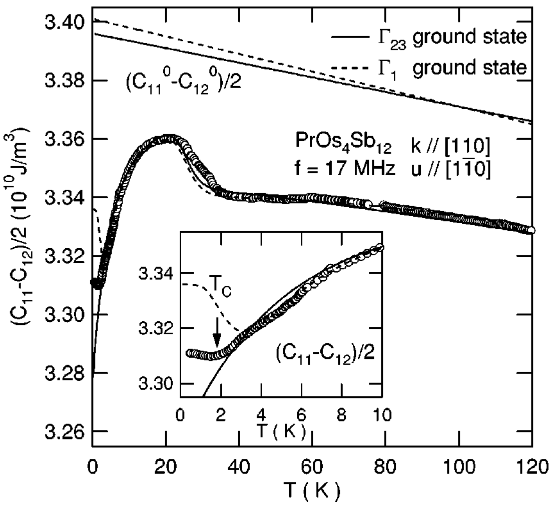

FIG. 1. Temperature dependence of $(C_{11}-C_{12})/2$ in $PrOs_4Sb_{12}$ measured by ultrasonic wave of 17 MHz. The anomaly around 30 K originates from the Γ_{23} rattling due to the Pr-ion off-center motion. The softening of $(C_{11}-C_{12})/2$ below 20 K down to superconducting point $T_C=1.85$ K is due to the quadrupolar fluctuation of the CEF states. The solid line and dashed line are fits by the quadrupolar susceptibility χ_Q for Γ_{23}-$\Gamma_4^{(2)}$ and Γ_1-$\Gamma_4^{(2)}$ models, respectively. Inset shows the detail around T_C.

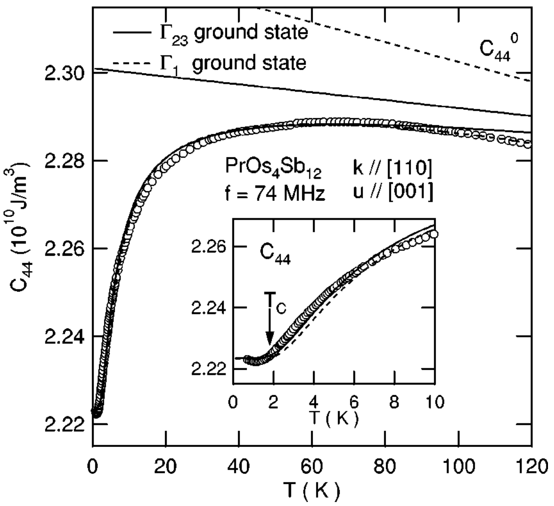

FIG. 2. Temperature dependence of the elastic constant C_{44} of $PrOs_4Sb_{12}$ measured by ultrasonic wave of 74 MHz. The softening of below 60 K down to superconducting point $T_C=1.85$ K is described in terms of the quadrupolar susceptibility χ_Q of solid line for Γ_{23}-$\Gamma_4^{(2)}$ model and dashed line for Γ_1-$\Gamma_4^{(2)}$ model. Inset shows the detail around T_C.

$=[1\bar{1}0]$. This $(C_{11}-C_{12})/2$ mode is associated with the elastic strain $\varepsilon_v=\varepsilon_{xx}-\varepsilon_{yy}$. The increase of $(C_{11}-C_{12})/2$ 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 $\omega\tau=1$. A relatively large lattice parameter $a=0.930311$ nm in $PrOs_4Sb_{12}$ 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 $(C_{11}-C_{12})/2$ below 20 K in Fig. 1 has been found with decreasing temperature. As shown in inset of Fig. 1, the softening of $(C_{11}-C_{12})/2$ turns up around the superconducting transition $T_C=1.85$ K. The quadrupole-strain interaction, $H_{QS}=-\Sigma_i\Sigma_{\Gamma\gamma}g_\Gamma O_{\Gamma\gamma}(i)\varepsilon_{\Gamma\gamma}$, and the inter-site quadrupole interaction, $H_{QQ}=-\Sigma_i g'_\Gamma\langle O_{\Gamma\gamma}\rangle O_{\Gamma\gamma}(i)$, give rise to the elastic softening as $C_\Gamma=C_\Gamma^0-Ng_\Gamma^2\chi_Q/(1-g'_\Gamma\chi_Q)$.[19] Here Σ_i means a sum over rare earth sites in unit volume. χ_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_2^2 with Γ_{23} symmetry to the elastic strain ε_v is relevant for the softening in $(C_{11}-C_{12})/2$ below 20 K of $PrOs_4Sb_{12}$.

The CEF Hamiltonian for the Pr^{3+} ion with the site symmetry T_h is written as $H_{CEF}=B_4O_4+B_6O_6+B_6^tO_6^t$, where $O_4=O_4^0+5O_4^4$, $O_6=O_6^0-21O_6^4$ and $O_6^t=O_6^2-O_6^6$.[20] Two different types of the CEF models of Γ_{23}-$\Gamma_4^{(2)}$ and Γ_1-$\Gamma_4^{(2)}$ for $PrOs_4Sb_{12}$ have been proposed so far.[4,21] The solid line in Fig. 1 based on the doublet model of Γ_{23}(0 K), $\Gamma_4^{(2)}$(8.2 K), $\Gamma_4^{(1)}$(133 K), and Γ_1(320 K) for $B_4^0=6.75\times10^{-2}$ K, $B_6^0=-1.23\times10^{-3}$ K, and $B_6^t=-0.12\times10^{-2}$ K with $|g_{\Gamma_{23}}|=97$ K and an inter-site coupling $g'_{\Gamma_{23}}=-0.27$ K reproduces the softening of $(C_{11}-C_{12})/2$ mostly proportional to reciprocal temperature. The doublet model seems to be favorable for the softening of $(C_{11}-C_{12})/2$.

Recently, Kohgi *et al.* have proposed a singlet ground state CEF model of Γ_1(0 K), $\Gamma_4^{(2)}$(7.9 K), $\Gamma_4^{(1)}$(135 K), and Γ_{23}(205 K) with $B_4^0=2.37\times10^{-2}$ K, $B_6^0=1.32\times10^{-3}$ K, $B_6^t=1.08\times10^{-2}$ K.[21] The dashed line for $|g_{\Gamma_{23}}|=79$ K and $g'_{\Gamma_{23}}=0.22$ K based on the singlet model also reproduces the softening of $(C_{11}-C_{12})/2$ except for a small deviation below a minimum around 3.5 K in the fitting. The off-center motion Pr ion with Γ_{23}-symmetry may enhance the elastic softening of $(C_{11}-C_{12})/2$ just above T_C, which will considerably renormalize the one-ion susceptibility of the dashed line in Fig. 1. Even in the case of the Γ_1-$\Gamma_4^{(2)}$ model, the charge fluctuation due to the off-center motion may reproduce the softening of $(C_{11}-C_{12})/2$ just above T_C proportional to reciprocal temperature. In order to settle the alternative CEF model of Γ_{23}-$\Gamma_4^{(2)}$ or Γ_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 Γ_{23}-$\Gamma_4^{(2)}$ model with parameters $|g_{\Gamma_4}|=34.4$ K and $g'_{\Gamma_4}=-0.002$ K. The dashed line is a fit for the Γ_1-$\Gamma_4^{(2)}$ model with $|g_{\Gamma_4}|=70$ K and $g'_{\Gamma_4}=-0.07$ K. Because the quadrupolar susceptibility of C_{44} for both models is domi-

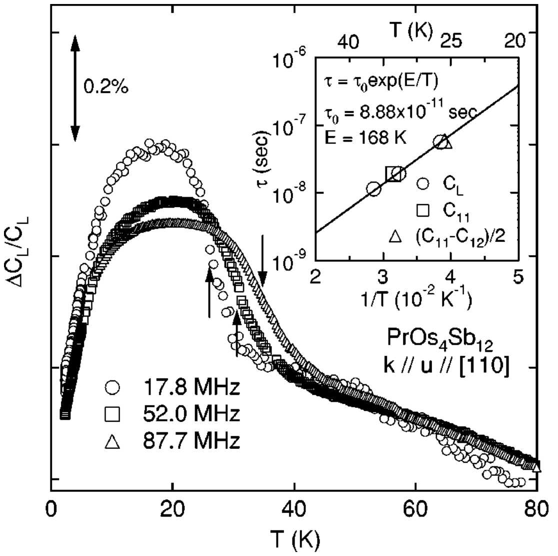

FIG. 3. Temperature dependence of $C_L=(C_{11}+C_{12}+2C_{44})/2$ in $PrOs_4Sb_{12}$ measured by ultrasonic waves of 17.8, 52.0 and 87.7 MHz. Arrows indicate the temperatures that the relaxation time τ of Pr-ion rattling coincides with the sound wave frequencies ω as $\omega\tau=1$. Inset shows temperature dependence of relaxation time.

nated 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} around 30 K.

Figure 3 represents $C_L=(C_{11}+C_{12}+2C_{44})/2$ of $PrOs_4Sb_{12}$ obtained by the longitudinal wave along the [110] direction. A remarkable frequency dependence of 17.8, 52.0, and 87.7 MHz in Fig. 3 is described in terms of the Debye-type dispersion as, $C_L(\omega)=C_L(\infty)-\{C_L(\infty)-C_L(0)\}/(1+\omega^2\tau^2)$. Here ω 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 the inset of Fig. 3 the temperature dependence of the relaxation time τ obtained by C_L is presented together with the results of $(C_{11}-C_{12})/2$ of Fig. 1 and C_{11}, which is not presented here. The relaxation time due to the rattling motion obeys the temperature dependence of $\tau=\tau_0\exp(E/k_BT)$ with an attempt time $\tau_0=8.8\times10^{-11}$ s and an activation energy $E=168$ K. Utilizing a harmonic oscillation of $\zeta(z)=(1/\pi z_0)^{1/2}\exp(-z^2/2z_0^2)$, 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.[22] This result is comparable to $z_0=0.048$ nm of $Ce_3Pd_{20}Ge_6$ (Ref. 14) and $z_0=0.012$ nm of $La_3Pd_{20}Ge_6$ (Ref. 17).

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 minimums as $a\sim z_0/2=0.04$ nm.

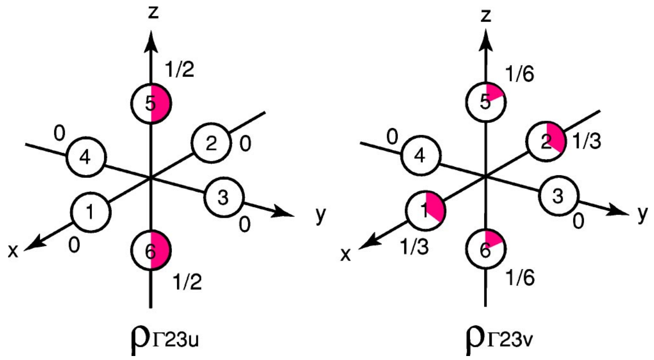

FIG. 4. The Γ_{23} rattling mode of $\rho_{\Gamma_{23u}}=2\rho_5+2\rho_6-\rho_1-\rho_2-\rho_3-\rho_4$ and $\rho_{\Gamma_{23v}}=\rho_1+\rho_2-\rho_3-\rho_4$ being responsible for the ultrasonic dispersion in $PrOs_4Sb_{12}$.

On the other hand, the Os atom locating at 0.4028 nm from the center of the cage along the threefold [111] axis prevents the off-center motion along the [111] axis. The Sb atoms closely locating to the twofold [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 ε_v with Γ_{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 Γ_{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 ε_u and ε_v has the Γ_{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.[23–25] Operating a symmetry element R of T_h on the atomic density $\rho_i=\rho(\rho_i)$ at the minimum point ρ_i $(i=1,2,\ldots,6)$, 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 Γ_1, Γ_{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 $PrOs_4Sb_{12}$, the Γ_{23} off-center mode of $\rho_{\Gamma_{23u}}=2\rho_5+2\rho_6-\rho_1-\rho_2-\rho_3-\rho_4$ and $\rho_{\Gamma_{23v}}=\rho_1+\rho_2-\rho_3-\rho_4$ with the fractional atomic distribution in Fig. 4 is the ground state of the system. $\rho_{\Gamma_{23u}}$ means the distribution of fraction 1/2 at ρ_5 and ρ_6 sites and null at ρ_1, ρ_2, ρ_3, ρ_4. And $\rho_{\Gamma_{23v}}$ is responsible for fraction 1/3 at ρ_1 and ρ_2, 1/6 at ρ_5 and ρ_6, and null at ρ_3 and ρ_4. The total symmetric mode $\rho_{\Gamma_1}=\rho_1+\rho_2+\rho_3+\rho_4+\rho_5+\rho_6$ and the polar mode $\rho_{\Gamma_{4x}}=\rho_1-\rho_2$, $\rho_{\Gamma_{4y}}=\rho_3-\rho_4$, $\rho_{\Gamma_{4z}}=\rho_5-\rho_6$ may correspond to the excited states. ρ_{Γ_1} represents the mean fraction 1/6 over the six sites. $\rho_{\Gamma_{4x}}$, for instance, has fraction 1/3 at ρ_1 and null at ρ_2, and fraction 1/6 at ρ_3, ρ_4, ρ_5, ρ_6.

Recent ultrasonic measurements on the clathrate compound $La_3Pd_{20}Ge_6$ by our group have successfully showed the rattling and tunneling motions of an off-center La ion in cage.[17] In the present clathrate compound of $PrOs_4Sb_{12}$ with a cage of Sb-icosahedron, the thermally activated rattling motion over the potential hill brings about the ultrasonic dispersion with the relaxation time τ in the 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 Γ_{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[22] may be relevant for the present $PrOs_4Sb_{12}$.

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 Γ_{23}-$\Gamma_4^{(2)}$ or Γ_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 $PrOs_4Sb_{12}$. Furthermore, the ultrasonic dispersion due to the Γ_{23} rattling motion with activation energy $E=168$ K has been found. The Γ_{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 $PrOs_4Sb_{12}$ and the interplay of the quadrupole fluctuation and the off-center motion of Pr-ion to the unconventional superconductivity.

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