

Simultaneous collapse of antiferroquadrupolar order and superconductivity in $\text{PrIr}_2\text{Zn}_{20}$ by nonhydrostatic pressure

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Superconductivity in $\text{PrIr}_2\text{Zn}_{20}$ appears at $T_c = 0.05$ K in the presence of an antiferroquadrupolar order below $T_Q = 0.11$ K. We have studied pressure dependences of T_c , T_Q , and non-Fermi-liquid behaviors in the resistivity $\rho(T)$ by using two pressure-transmitting media: argon, maintaining highly hydrostatic pressure, and glycerol, which solidifies above 5 GPa, producing nonhydrostatic pressure. Upon applying P with argon up to 10.6 GPa, T_c hardly changes, while T_Q monotonically increases from 0.11 to 0.23 K. With glycerol, however, T_Q and T_c simultaneously fall below 0.04 K at 6.3 GPa. The contrasting results indicate that on-site quadrupolar fluctuations induce superconductivity in this compound.

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I. INTRODUCTION

Unconventional superconductivity in correlated materials [1] such as high- T_c copper oxides [2], ruthenium oxides [3], iron-based pnictides [4], and heavy-fermion compounds [5] has attracted tremendous attention over three decades as one of the most intriguing aspects in condensed-matter physics. Fluctuations of magnetic dipole moments in them are thought to be the glue for superconducting pairing [1–5]. In recent years, pairing interaction due to quadrupole fluctuations has been proposed in praseodymium-based intermetallic compounds [6,7]. However, the role of the quadrupole freedom in superconductivity remains elusive in spite of detailed studies for several systems. $\text{PrOs}_4\text{Sb}_{12}$, for example, exhibits superconductivity below $T_c = 1.85$ K and a magnetic-field-induced antiferroquadrupole (AFQ) order above $B = 4.5$ T [7]. The field-induced AFQ order results from the crossing of the crystalline electric field (CEF) levels between the Γ_1 singlet ground state and the Zeeman-split Γ_4 level with quadrupolar freedom [8]. Because the superconducting phase is located in the vicinity of the quadrupole ordered phase, it was argued that the quadrupole fluctuations may play an important role in the superconductivity [8,9].

Recently, superconductivity was discovered in the series of compounds $\text{PrT}_2\text{X}_{20}$ ($T = \text{Ir, Rh}$, $X = \text{Zn}$; $T = \text{Ti, V}$, $X = \text{Al}$) [10–12]. The local point-group symmetry of the Pr site is T_d (or T) in the cubic $\text{CeCr}_2\text{Al}_{20}$ -type structure with the space group $Fd\bar{3}m$. The CEF ground state of $4f^2$ electrons in the Pr^{3+} ion was identified as a non-Kramers Γ_3 (or Γ_{23}) doublet with quadrupolar freedom [12]. Interestingly, the superconducting transition sets in below the ordering temper-

ature of either antiferroquadrupoles or ferroquadrupoles. In $\text{PrT}_2\text{Zn}_{20}$ ($T = \text{Ir}$ and Rh), AFQ transitions at $T_Q = 0.11$ and 0.06 K are followed by superconducting ones at $T_c = 0.05$ and 0.06 K, respectively [13,14]. In $\text{PrTi}_2\text{Al}_{20}$, on the other hand, a ferroquadrupole (FQ) transition at $T_Q = 2$ K is followed by a superconducting one at $T_c = 0.2$ K [15]. The coexistence of the AFQ or FQ order with the superconductivity leads to the proposition that the superconducting pairing interaction is mediated by quadrupole fluctuations [13–15].

In $\text{PrT}_2\text{Zn}_{20}$ ($T = \text{Ir, Rh}$), the electrical resistivity $\rho(T)$ decreases with a downward curvature on cooling from 2 K to T_Q , and the $4f$ contribution to the specific heat divided by temperature, C_{4f}/T , follows the form of $-\ln T$ for $0.2 < T < 0.8$ K [16]. These non-Fermi-liquid (NFL) behaviors in $\rho(T)$ and C_{4f}/T are consistent with the predictions from the two-channel Anderson lattice model [17]. Furthermore, the values of magnetic entropy at T_Q for $T = \text{Ir}$ and Rh are, respectively, 20% and 10% of $R\ln 2$ [13,14]. The reduced entropy was attributed to fluctuations of the quadrupole moments which persevere temperatures up to 20–30 times higher than T_Q .

Application of pressure can control the CEF level scheme as well as the strength of the hybridization between the $4f$ and conduction electrons (c - f hybridization), which is an important parameter for the quadrupole Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY)-type interaction among quadrupoles. Upon applying pressure to $\text{PrOs}_4\text{Sb}_{12}$ up to 8 GPa, T_c is suppressed monotonically, and the field-induced AFQ phase moves to lower fields [18]. This change in T_c was attributed to the decrease in the energy splitting between the Γ_1 singlet ground state and Γ_4 triplet excited state [18]. For $\text{PrTi}_2\text{Al}_{20}$, T_Q begins to be suppressed above 6 GPa, where T_c is strongly enhanced together with the effective mass of the quasiparticles [19]. The opposite changes in T_Q and T_c support the proposition that the enhanced

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fluctuations of quadrupoles in the vicinity of the quantum critical point strengthen the superconducting interaction.

We recall that the AFQ order is sensitive to the uniformity of the pressure applied to the crystal. In fact, an AFQ order at $T_Q = 0.4$ K in PrPb_3 suddenly disappears at 5 GPa [20], which coincides with the solidification pressure of the transmitting medium of glycerol [21]. It is likely that the solidification causes nonuniform pressure which lowers the local symmetry of Pr^{3+} and lifts the twofold degeneracy of the Γ_3 doublet. Therefore, comparing the effects of nonhydrostatic and highly hydrostatic pressures on T_Q and T_c in $\text{PrIr}_2\text{Zn}_{20}$ may give a clue for understanding the relation between the superconductivity and AFQ fluctuations. Bearing this in mind, we have measured the electrical resistivity $\rho(T)$ of $\text{PrIr}_2\text{Zn}_{20}$ under pressures applied using two types of pressure-transmitting media: glycerol, producing nonhydrostatic pressure above 5 GPa, and argon, keeping pressure hydrostatic up to 10 GPa [22,23]. We also compare these data with the pressure dependence of T_c for the BCS superconductor $\text{LaIr}_2\text{Zn}_{20}$ [10,24]. A previous study of the La-substituted system $\text{Pr}_{1-x}\text{La}_x\text{Ir}_2\text{Zn}_{20}$ showed that T_Q vanishes even at a small $x < 0.09$, while T_c hardly changes for a wide range, $0 \leq x \leq 0.47$ [24]. The independent behaviors between T_Q and T_c for the La-substituted system have not disclosed the role of AFQ fluctuations in the superconductivity. In this paper, we compare variations of T_Q and T_c under hydrostatic and nonhydrostatic pressures. The results indicate that on-site coupling between quadrupole fluctuations and conduction electrons induces superconductivity in $\text{PrIr}_2\text{Zn}_{20}$.

II. EXPERIMENTAL DETAILS

Single crystals of $\text{PrIr}_2\text{Zn}_{20}$ and $\text{LaIr}_2\text{Zn}_{20}$ were prepared by a melt-growth method from high-purity elements, Pr (4N), La (4N), Ir (3N), and Zn (6N), as reported previously [10]. The $\rho(T)$ under pressure was measured by an ac four-terminal method using a piston-cylinder pressure cell for $P \leq 2.1$ GPa and an opposed-anvil pressure cell for $P \geq 3.2$ GPa. The pressure was applied at room temperature. In the piston-cylinder cell, Daphne oils 7373 and 7474 were used as pressure-transmitting media which produce hydrostatic pressure until they solidify at 2.3 and 3.7 GPa at room temperature, respectively [25]. In the opposed-anvil cell, argon and glycerol were used as described above. The pressure was estimated from the pressure dependence of T_c of a piece of lead placed in the cell [26]. The magnitude of pressure gradients ΔP was estimated from the temperature width of the superconducting transition of the Pb manometer. A commercial Cambridge Magnetic Refrigerator mFridge mF-ADR50 was used to cool the pressure cell down to 0.04 K.

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) represent the low-temperature data of $\rho(T)$ under pressures applied using argon and glycerol as pressure-transmitting media, respectively. Upon applying pressure with argon, T_c in $\rho(T)$ remains unchanged, whereas the sharp drop at T_Q shifts to higher temperatures. The steady increase in T_Q suggests the AFQ order is stabilized by the increased RKKY-type interaction among the quadrupole

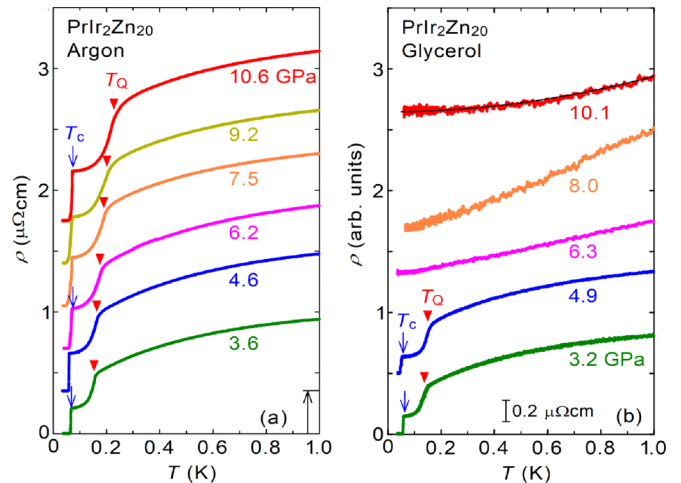


FIG. 1. Temperature dependence of the electrical resistivity $\rho(T)$ of $\text{PrIr}_2\text{Zn}_{20}$ under various pressures applied using (a) argon and (b) glycerol as pressure-transmitting media. The triangles indicate the AFQ ordering temperature T_Q . The data sets at various pressures are shifted consecutively for clarity. The solid curve for $P = 10.1$ GPa represents the fit with $\rho(T) = \rho_0 + AT^2$, with $A = 0.3 \mu\Omega \text{ cm}/\text{K}^2$. This Fermi-liquid behavior in $\rho(T)$ suggests that the splitting of the Γ_3 doublet stabilizes the Fermi-liquid state.

moments. On the other hand, upon applying pressure with glycerol, the anomalies at T_c and T_Q as well as the downward curvature above T_Q remain up to 4.9 GPa, where glycerol holds a liquid state at room temperature. However, at 6.3 GPa, where glycerol solidifies, we did not observe any transition in $\rho(T)$ down to the lowest temperature, 0.04 K. It is noted that the downward curvature in $\rho(T)$ also disappears, as shown in Fig. 1(b). The concomitant suppression of T_c and T_Q and the downward curvature in $\rho(T)$ suggest that the scattering of conduction electrons by means of the Γ_3 doublet is significantly modified under nonhydrostatic pressure. In fact, pressure gradients along the Pb manometer increase slowly from $\Delta P = 0.01$ GPa at 3.2 GPa to 0.02 GPa at 4.9 GPa but steeply to $\Delta P = 0.39$ GPa at 6.3 GPa, as demonstrated in Fig. S4 in the Supplemental Material [27] (see also Refs. [28,29] therein). This pronounced increase of the pressure gradient in the range 4.9–6.3 GPa should cause a large anisotropic strain in the sample.

The pressure dependences of T_c and T_Q are compared in Fig. 2 for argon and glycerol media together with the data for Daphne oil for $P \leq 2.1$ GPa (see Fig. S1 in the Supplemental Material for details [27]). T_c was defined as the onset temperature of the drop in $\rho(T)$ in Fig. 1, and T_Q was taken as the peak temperature of $d\rho/dT$. For $P = 3.2$ and 4.9 GPa, T_c and T_Q for glycerol are slightly lower than those for argon, suggesting the influence of the increased viscosity of glycerol near the solidification pressure. On further pressurizing with glycerol up to 6.3 GPa, both T_c and T_Q vanish. The vanishment of T_Q is attributed to the possible anisotropic strain caused by the nonhydrostatic pressure, which lowers the cubic symmetry of the Pr site. Thereby, the nonmagnetic Γ_3 doublet of the CEF ground state of Pr^{3+} splits and loses the quadrupolar degree of freedom. The concomitant disappearance of T_c with T_Q is

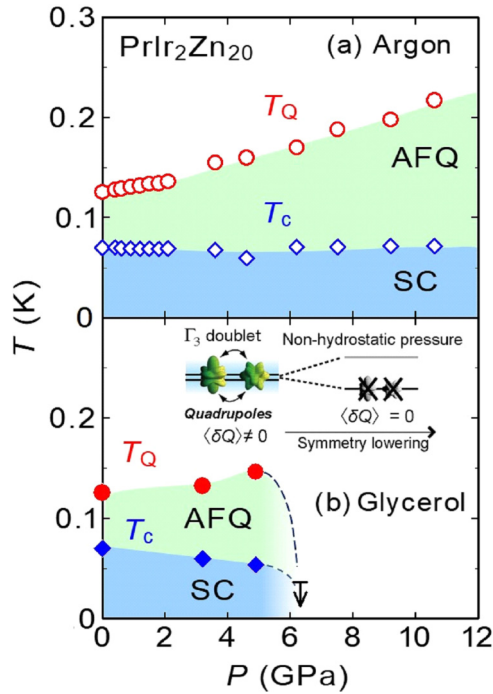


FIG. 2. Pressure dependences of the superconducting transition temperature T_c and AFQ ordering temperature T_Q using (a) argon and (b) glycerol as the pressure-transmitting media, respectively. The data for $P \leq 2.1$ GPa in (a) are taken using a piston-cylinder cell with Daphne oil 7474 (see Fig. S1 in the Supplemental Material for details [27]). The inset in (b) represents a schematic diagram for the nonhydrostatic effect on the Γ_3 doublet.

indicative of a strong correlation between the superconductivity and AFQ order.

Let us compare the above results with those for the substitution of La for Pr in $\text{Pr}_{1-x}\text{La}_x\text{Ir}_2\text{Zn}_{20}$ [24]. It was found that T_Q disappears at a small substitution level $x = 0.09$, while the superconducting transition remains in the whole range $0 \leq x \leq 1$ [24]. This fact was interpreted to be an indication of the lack of a relation between T_c and T_Q . Now, it is noteworthy that the electronic state of Pr^{3+} in the substituted system is different from that under pressure. In our previous paper [24], two mechanisms were considered to explain the suppression of T_Q in the La-substituted system. First, the expectation value of the quadrupoles is reduced when the Γ_3 doublet is split. Second, the intersite RKKY-type quadrupole interaction [30] is weakened by the breaking of coherence in the lattice due to random distribution of Pr and La atoms.

The second mechanism was considered to play the dominant role in the collapse of the AFQ order in the La-substituted system because the first one was discarded. In fact, the magnetic susceptibility and specific heat data for $\text{Pr}_{1-x}\text{La}_x\text{Ir}_2\text{Zn}_{20}$ indicated the stability of the Γ_3 doublet even in the substituted system [24]. Under this condition, quadrupolar fluctuations at each Pr site do not vanish. The quadrupolar fluctuations manifest themselves in the downward curvature of $\rho(T)$ for $x = 0$ on cooling below 2 K due to the quadrupolar Kondo effect [17,31,32]. The downward curvature in $\rho(T)$ remains for $x = 0.22$ in which no AFQ order occurs, but a superconducting transition appears below 0.07 K [33]. Therefore,

superconductivity survives in the La-substituted system if it originates from the on-site coupling between quadrupole fluctuations and conduction electrons.

On the other hand, the first mechanism is responsible for the collapse of the AFQ order under nonhydrostatic pressure applied by solid glycerol. Once on-site quadrupole fluctuations are quenched, superconductivity no longer survives. The disappearance of the downward curvature in $\rho(T)$ at 6.3 GPa as shown in Fig. 1(b) strongly indicates the quenching of the quadrupole fluctuations. This scenario is consistent with the coexistence of AFQ order and superconductivity under hydrostatic pressures applied by argon. Furthermore, according to this scenario, superconductivity may survive until the uniaxial stress becomes large enough to stop on-site quadrupole fluctuations. In fact, by using another pressure medium, a Fluorinert 70/77 = 1:1 mixture, we have observed both the AFQ order and superconducting transitions up to 9.6 GPa beyond the solidification pressure of 1 GPa, as shown in Figs. S2 and S3 in the Supplemental Material [27]. The transition happened because the uniaxial stress in the solid of Fluorinert was small compared with that of solid glycerol at $P > 6$ GPa (see Sec. 3 of the Supplemental Material for details [27]). All observations for $\text{PrIr}_2\text{Zn}_{20}$ under hydrostatic and nonhydrostatic pressures as well as for the La-substituted system are consistent with the idea that on-site coupling between quadrupole fluctuations and conduction electrons induces superconductivity in $\text{PrIr}_2\text{Zn}_{20}$.

To shed light on the pairing mechanism for the superconductivity, we compare the observed pressure effect on $\text{PrIr}_2\text{Zn}_{20}$ with theories and those on other systems. The above argument is consistent with the theoretical calculation showing that the superconductivity for the f^2 state of the Γ_3 non-Kramers doublet system appears only in the quadrupole ordered phase [34]. In this theory, the on-site pairing state composed of the Γ_3 doublet state is indispensable to the superconductivity. The pressure dependence of T_c of $\text{PrIr}_2\text{Zn}_{20}$ is distinguished from that of $\text{PrOs}_4\text{Sb}_{12}$ with the Γ_1 CEF ground state. In the latter, T_c of 1.85 K at $P = 0$ decreases continuously to 1.3 K by applying pressure up to 8 GPa even with the glycerol transmitter. Note that there is no anomaly in $T_c(P)$ around 5 GPa [18], unlike the case of $\text{PrIr}_2\text{Zn}_{20}$. Furthermore, as shown in Fig. 2(a), under hydrostatic pressure, T_c of $\text{PrIr}_2\text{Zn}_{20}$ hardly changes up to 10 GPa. For $\text{PrTi}_2\text{Al}_{20}$, T_c also hardly changes up to 6 GPa [35]. In the BCS-type superconductor $\text{LaIr}_2\text{Zn}_{20}$, however, T_c of 0.6 K at $P = 0$ is suppressed with a ratio of -0.12 K/GPa, as shown in Fig. 3. This contrasting behavior in $T_c(P)$ between $\text{PrIr}_2\text{Zn}_{20}$ and $\text{LaIr}_2\text{Zn}_{20}$ strongly suggests a non-BCS pairing mechanism for the superconductivity in $\text{PrIr}_2\text{Zn}_{20}$. If on-site interaction between quadrupoles and conduction electrons at the Pr site plays an important role in the pairing mechanism as proposed by theories [34,36,37], the superconductivity would be robust against hydrostatic pressure.

We turn our attention to the hydrostatic pressure effect on $\rho(T)$ for $T > T_Q$. On cooling, $\rho(T)$ at ambient pressure transforms from a T -linear line to a curve with a downward curvature, as shown in Fig. 4(a). We denote the crossover temperature as T_R , as indicated by the arrow, which is defined as the temperature where $\rho(T)$ starts deviating from the linear dependence on cooling. A theory based on the two-channel

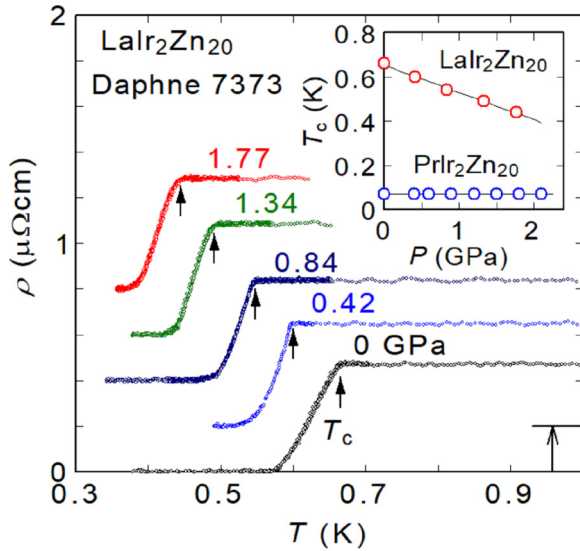


FIG. 3. Temperature dependence of the electrical resistivity of $\text{LaIr}_2\text{Zn}_{20}$ under various constant pressures applied with Daphne oil 7373. Data sets are shifted upward consecutively by $0.2 \mu\Omega\text{cm}$ for clarity. The inset shows the pressure dependence of the onset temperature T_c of the superconducting transition for $\text{LaIr}_2\text{Zn}_{20}$ and $\text{PrIr}_2\text{Zn}_{20}$.

Anderson lattice model shows that T_R increases as the c - f hybridization is increased [17]. In fact, an experimental study substituting Cd and Ga for Zn in $\text{PrIr}_2\text{Zn}_{20}$ confirmed that T_R is controlled by the strength of c - f hybridization [38].

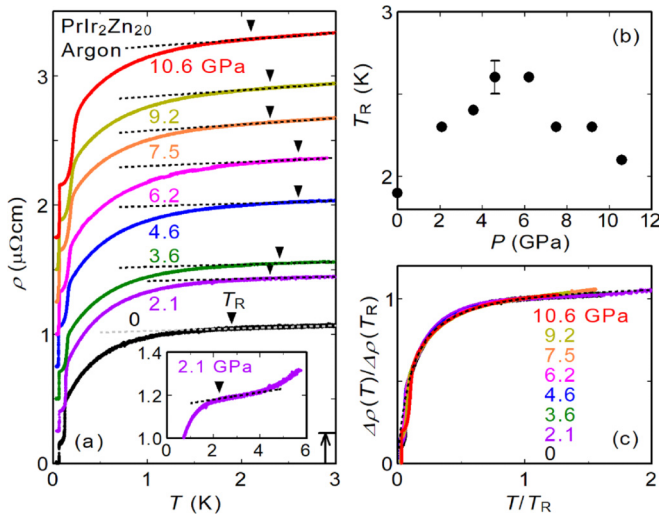


FIG. 4. Temperature dependence of the electrical resistivity $\rho(T)$ of $\text{PrIr}_2\text{Zn}_{20}$ under pressure up to 10.6 GPa applied with argon as the transmitting medium. Data sets at various pressures are shifted upward consecutively by $0.25 \mu\Omega\text{cm}$ for clarity. The arrows indicate the characteristic temperature T_R , which is defined as the temperature where $\rho(T)$ starts deviating from the linear dependence on cooling. The inset shows the data of $\rho(T)$ at 2.1 GPa in the expanded temperature range. (b) Pressure dependence of T_R . (c) Scaling plot of the differential electrical resistivity $\Delta\rho = \rho(T) - \rho_0$ under various constant pressures. In the temperature region $0.1 < T/T_R < 1.2$, $\Delta\rho$ follows the dashed curve calculated by using the two-channel Anderson lattice model [17], as shown by the dashed curve.

Therefore, T_R can be regarded as the measure of the strength of the c - f hybridization. In Fig. 4, the data for $\rho(T)$ under various constant pressures show that T_R increases with P from 1.8 to 2.6 K at around 6 GPa and decreases to 2.1 K at 10.6 GPa [see Fig. 4(b)].

Figure 4(c) displays a plot of $\Delta\rho(T)/\Delta\rho(T_R)$ vs T/T_R , where $\Delta\rho(T) = \rho(T) - \rho_0$ and ρ_0 is the residual resistivity. Note that all the data taken under pressures from 0 to 10.6 GPa fall on one curve in a rather wide temperature range of $0.1 < T/T_R < 1.2$. The dashed curve is a fit of the following form derived from the two-channel Anderson lattice model [17]:

$$\Delta\rho = \frac{a_1}{1 + a_2 \frac{T}{T_R}},$$

where a_1 and a_2 are parameters. This scaling underlines that the NFL behavior in $\rho(T)$ characterized by the downward curvature is the manifestation of the quadrupolar Kondo lattice.

Let us discuss the pressure dependence of T_R . The increment of T_R for $P < 6$ GPa is understood as a result of the enhancement of the c - f hybridization under pressure. However, this relation does not hold for $P > 6$ GPa, where T_R decreases with pressure. On the other hand, by substituting Ga for Zn in $\text{PrIr}_2\text{Zn}_{20}$, T_R is increased as a result of the increase in the $4p$ electron density of states of the conduction bands [38]. Therefore, the decrease in T_R for $P > 6$ GPa may result from the decrease of the density of states at the Fermi level. Consequently, the pressure-induced enhancement of c - f hybridization gives way to suppression of c - f hybridization by the decrease of the density of states. To confirm this scenario, the modification of the band structure under pressure needs to be calculated.

IV. CONCLUSION

In iron-based and heavy-fermion superconductors, magnetic fluctuations and/or orbital fluctuations have been discussed as the Cooper pairing interaction thus far, despite the prevailing controversy on the detailed mechanism [2,4–9,39–41]. In this work, we have measured the electrical resistivity of $\text{PrIr}_2\text{Zn}_{20}$ under pressure applied using two different pressure-transmitting media, argon (hydrostatic) and glycerol (nonhydrostatic for $P > 5$ GPa). Under hydrostatic pressure, T_c hardly changes up to 10.6 GPa, while T_c for the isostructural BCS superconductor $\text{LaIr}_2\text{Zn}_{20}$ largely decreases. This contrasting pressure dependence of T_c corroborates the unconventional nature of the superconductivity in $\text{PrIr}_2\text{Zn}_{20}$. On the other hand, the AFQ order is stabilized under the hydrostatic pressure, as manifested by the monotonic increase in T_Q from 0.11 to 0.23 K. Under nonhydrostatic pressure at $P = 6.3$ GPa, both T_Q and T_c simultaneously disappear. The comparison between the pressure effect and substitution effect on T_Q and T_c strongly suggests that on-site coupling between quadrupole fluctuations and conduction electrons induces superconductivity in $\text{PrIr}_2\text{Zn}_{20}$. Furthermore, NFL behavior in $\rho(T > T_Q)$ with the downward curvature also disappears under nonhydrostatic pressure. These observations reveal that the NFL behavior as well as the superconductivity in $\text{PrIr}_2\text{Zn}_{20}$ results from the quadrupolar degree of freedom through c - f hybridization.

Our finding will pave the way to a deeper understanding of the pairing mechanism mediated by orbital fluctuations in iron-based and heavy-fermion superconductors.

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