Pressure-induced ferromagnetic quantum criticality in the low-carrier system CeP

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Abstract. We have already obtained the tentative pressure (P)-temperature (T) magnetic phase diagram of the antiferromagnetic (AF) semimetal CeP with strongly localized 4f electrons by measurements of the only T-dependence of the electrical resistivity, \( \rho(T) \), up to 8 GPa. In order to get further reliability, we performed the simultaneous measurements of the T-dependent real part of the AC magnetic susceptibility, \( \chi'_{AC}(T) \), and \( \rho(T) \) at \( P \lesssim 2.5 \) GPa. As a result, the temperature which give the resistive anomaly was found to be the exact transition temperature from the paramagnetic (PM) phase to the magnetically ordered state, even in \( P > 2.5 \) GPa. The detailed magnetic structures (MSs) is already clarified by the neutron diffraction (ND) experiments under high pressures up to 1.7 GPa by Kohgi et al. and up to 5.6 GPa by Goncharenko et al. The careful comparison between our macroscopic P-T magnetic phase diagram and the microscopic MSs elucidated the new facts: in the high-P region, the induction of the pure ferromagnetic (FM) phase for \( P \gtrsim 4 \) GPa is revealed to cause a rapid decrease in the Curie temperature \( T_C \) and the Ce localized magnetic moment (LM); and furthermore, the \( T_C \) is observed to vanish at a critical pressure \( P_C \approx (5.5 \pm 0.1) \) GPa together with the LM. We report the convincing basis for the presence of a P-induced FM quantum critical point (FM-QCP) and the possibility of localized-delocalized transition of the Ce-4f states approximately at the FM-QCP in the semi-metallic LM system CeP with the extremely low-carrier density.

1. Introduction
When the second-order phase transition temperature arrives at \( T \to 0 \) K by tuning of a nonthermal control parameter such as pressure, magnetic field, and chemical substitution, quantum fluctuations exist even at \( T = 0 \) K. This second-order phase transition point of absolute zero is generally called a quantum critical point (QCP) [1, 2]. An overwhelming majority of such QCPs are antiferromagnetic (AF) instabilities realized in itinerant d- or f-electron systems [3, 4]. In contrast, the FM-QCP is extremely rare. It is generally thought that many d- or f-electron FM-systems do not have a QCP. The reasons for this follow. For suppression of \( T_C \) by applying pressure in Ce-based compounds, such as CeAgSb\(_2\) [5] or CeRu\(_2\)Ge\(_2\) [6], long-range AF order is induced before reaching \( P_C \); alternatively, the induction of a first-order transition at \( P < P_C \) obscures the FM-QCP, for example UGe\(_2\) [7] or ZrZn\(_2\) [8]. Other cases for the example which hints weakly the presence of a FM-QCP are: CePdSb [9] and CeSi\(_{1.8}\) [10]. In the case of chemical substitution, a sort of FM cluster-glass prevents arriving at a FM-QCP, for example, in CePt\(_{1-x}\)Rh\(_x\) [11]. Therefore, experiments performed to date strongly suggest that the majority of d- or f-electron itinerant metallic-FM systems do not contain a FM-QCP.

The compounds that indicate conclusive evidence for the presence of a FM-QCP have been discovered in the following clean 4f-electron systems FM metal: YbNi\(_4\)(P\(_{0.92}\)As\(_{0.08}\))\(_2\) [12] and Ce(Ru\(_{1-x}\),Fe\(_x\))PO with \( x \approx 0.86 \) [13]. YbNi\(_4\)(P\(_{0.92}\)As\(_{0.08}\))\(_2\) is the heavy fermion metallic LM ferromagnet and...
Ce(Ru$_{1.6}$Fe$_{0.4}$)PO is a itinerant metallic-ferromagnet. These are new compounds for studying the FM quantum criticality in f-electron systems. In particular, YbNi$_4$(P$_{0.92}$As$_{0.08}$) is very interesting, because this material hides a potential localization-delocalization transition of the Yb-4f state. However, almost all of the d- and f-electron compounds mentioned above are typical metals. Our interest is in the transition from a FM-semimetal with LM to a normal-metal at a FM-QCP, not the FM-QCP itself, which is caused by the P-induced valence fluctuating state. For this reason, we abandoned the metallic ferromagnets with no LM, which already have demonstrated the FM order at ambient pressure.

We have paid special attention to the Ce-monopnictides, CeX (X = N, P, As, Sb and Bi) with the simple NaCl-type crystal structure. Except for the valence fluctuating CeN, all of the CeX compounds are magnetic semimetals. In particular, CeP is the most attractive material among CeX compounds. This is because CeP is located in the intermediate position that connects the metallic CeN to CeSb and CeBi, indicating the characteristics of well localized 4f-electrons. The main features of CeP at ambient-P are: (a) the physical properties of CeP with extremely low carrier density are characterized by a semi-metallic band structure and localized 4f-electrons [14-16]; (b) the low carrier density is attributed to a slight overlap near the Fermi level between the top of the valence band at the Γ point and the bottom of the conduction band at the X point in the Brillouin zone [17, 18]; (c) the carrier concentration is almost on the order of 0.01/Ce, which is roughly estimated by the volume of the hall pocket at the Γ point and that of the electron at the X point by the de Haas-van Alphen (dHvA) or Shubnikov-de Haas (SdH) effect [15, 19-21]; (d) the Ce$^{3+}$ 4f$_{5/2}$ multiplet is split into the Γ$_7$ ground state doublet and the Γ$_8$ exited state quartet due to the cubic crystal field; (d) the energy difference $\Delta_\Gamma$ between the Γ$_7$ and Γ$_8$-levels is $\approx$ 162 K [22]; (e) the excited Γ$_7$-level is very well separated from the Γ$_8$ lowest level; and (f) the simple type-I AF order at the Néel temperature $T_N = 10.5$ K with its value of LM $u_{C_m} \approx (0.8\pm0.1)\mu_\text{B}/\text{Ce}$ originating from the Γ$_8$ ground state [23, 26].

The physical properties of CeP are very sensitive to an applied pressure. For example, the application of a slight-P for $P \approx 0.3$ GPa to CeP causes “various-striped” magnetic structures (MSs) consisting of ferromagnetically coupled Γ$_8$-like bilayers and Γ$_7$-single plans, which indicate the co-existence between the Γ$_8$ and Γ$_7$-states by overcoming a considerably large energy difference $\Delta_\Gamma \approx 162$ K [25]. Our goal is to search the P-induced FM-QCP in the magnetic semimetal CeP with LM and the final aim is the localized-itinerant transition accompanied by a change in valence approximately at the FM-QCP.

2. Experiments

Extremely high quality single crystalline CeP was grown in a scaled tungsten crucible using a mineralization method with a high frequency induction furnace that demonstrates clearly dHvA oscillations and a SdH signal [14, 15]. We used a piston-cylinder device below 2.5 GPa, and a cubic-anvil device up to 8 GPa [24] alternately for the $T$-region $\approx 4.2 \leq T \leq 300$ K, with a mixture of machine oil and kerosene as the pressure-transmitting medium. To keep constant pressure during the heat cycle, the load was maintained constant by controlling the oil-press equipment [24].

The $\rho(T)$ was measured using a DC four probe method. The $\chi^*_\text{AC}(T)$ was performed by a Hartshorn-type bridge with the frequency of 33 Hz.

3. Results

Figure 1 depicts the $\rho(T)$ curves at selected $P$ values for 0–6.5 GPa. The $\rho(T)$ at ambient $P$ exhibits a sharply-pointed peak at $T_N = 10.5$ K due to the
type-I AF order. For \( P \geq 0.3 \) GPa, a prominent peak splits into the two anomalies at \( T_c \) for the low-\( T \)-side and at \( T_H \) for high-\( T \) region, as indicated by arrows, open ellipses, and closed circles, respectively. The anomaly at \( T_H \) shows a first-order-like precipitous change in \( \rho(T) \) and at \( T_c \) is a small hump. To confirm the relation between the resistive anomaly at \( T_H \) and the magnetic ordering, we performed the simultaneous measurement of \( \rho(T) \) and \( \chi'(T) \) under pressures up to 2.5 GPa. Figure 2 demonstrates \( \rho(T) \), \( \chi'(T) \), and the \( T \)-derivative of \( \rho(T) \), \( d\rho/dT-T \), at 1.5 GPa as a typical example. As seen from Fig. 2, \( d\rho/dT-T \) forms a very prominent peak at \( T_H \). Consequently, \( T_H \) is strictly defined as one point of the temperature that gives the steepest gradient in \( \rho(T) \). Furthermore, the peak positions of both \( \chi'(T) \) and \( d\rho/dT-T \) completely coincides with each other, obviously indicating that \( T_H \) is the transition temperature from the paramagnetic phase to the magnetically ordered state. Therefore, we can accurately determine \( T_H \) for \( P > 2.5 \) GPa only by measurements of \( \rho(T) \) under high pressures of up to 8 GPa, without making a magnetic measurement.

We perform a careful comparison between our macroscopic \( P-T \) magnetic phase diagram shown in Fig. 3 and the microscopic MSs obtained by ND experiments under high-\( P \) up to 5.6 GPa [25, 26]. The \( P \)-dependent \( T_H \) and \( T_c \) are summarized in Fig. 3. \( T_H \) increases considerably up to 51 K around 3 GPa; for \( P \gtrsim 3.0 \) GPa, \( T_H \) starts to decrease. On the other hand, \( T_c \) is almost \( P \)-independent and tends to disappear at approximately 1.5–2 GPa. Thus, the \( P-T \) plot is divided into two regions, denoted \( \alpha \) and \( \beta \) in accordance with Ref. [25]. Our interests are only in the \( \beta \)-phase and the high-\( P \)-phase for \( P > 3 \) GPa. The \( \beta \)-phase (0.3–1.7 GPa and 12–40 K) shown in Fig. 3 accompanied by “various-striped” MSs consists of ferromagnetically coupled \( \Gamma_7 \)-like bi-planes with the \( \mu_{C_2} \approx 2 \mu_B \) and \( \Gamma_7 \)-PM planes [25]. The \( \chi'_AC(T) \) at 1.5 GPa in Fig. 2 shows divergent behavior at \( T_H \approx 35 \) K, indicating the feature of a typical second-order phase transition. This obviously detects the FM-component in the \( \beta \)-phase, which causes a precipitous decrease in \( \rho(T) \). For \( P \geq 2.3 \) GPa, the FM phase begins to appear at \( \approx 30 \) K [25, 26]: in the region of 3–4 GPa, the FM-state coexists with an extra phase at 32–45 K [26]; the pure FM-phase appears for \( P \gtrsim 4 \) GPa with a large \( \mu_{C_2} \approx 1.6 \mu_B \) [26]; and thus, we rewrite \( T_H \rightarrow T_c \) (see Fig. 1 and Fig. 3). The results that have been discussed so far are summarized in Fig. 3 as a \( P-T \) magnetic phase diagram. Our \( \rho(T) \) data at 5.3–6 GPa in Fig. 1 shows that \( T_c \) vanishes at a critical pressure \( P_c \approx 5.5 \) GPa. On the other hand, the \( \mu_{C_2} \) in the vicinity of 5.5 GPa are as follows [26]: \( \mu_{C_2} \approx 1.1 \mu_B \) at 4.9 GPa and \( 0 \leq \mu_{C_2} \leq 0.6 \mu_B \) at 5.6 GPa within the experimental sensitivity (\( \mu_{C_2} \approx 0.6 \mu_B \) at 1.5 K) [26]. This result indicates that \( P_c \) exists for the range 5–5.6 GPa. Interestingly, both \( T_c \) and LM decrease rapidly until they simultaneously disappear with the emergence of the pure FM-order. By considering our result and the ND experiments, we define the critical-\( P \) as \( P_c \approx (5.5 \pm 0.1) \) GPa.

Figure 2. \( \rho(T) \), \( \chi'_AC(T) \) and \( d\rho/dT-T \) at 1.5 GPa as a typical example.

Figure 3. \( P-T \) magnetic phase diagram of CeP. Striped MSs after Ref.[25]. MSs for \( P \gtrsim 2.5 \) GPa after Ref.[26], \( T_H \) (■), \( T_c \) (●) and \( T_c \) (○). The solid line and curve are guides to the eye.
This behavior directly indicates a distinct increase in
other hand, UGe$_2$ (excluding the F2 phase) \[29, 31, 32\] and CeRh$_2$Si$_2$ (excluding the 2nd - order transition)\[28, 29\] around the QCP, clearly indicating the absence of critical fluctuations. In the case of CeP, we consider the influence of the magnetic critical fluctuations with respect to the low-\(\rho\) behavior and forms an extremely sharp peak for 5.3 - 5.7 GPa. Note that the magnitude of \(\alpha\) in CeP is \(\approx 1000\) times smaller than that of usual heavy fermion compounds, which is attributed to the extremely low-carrier density of \(\approx 0.01/\text{Ce}\). The sharp maxima in \(\rho_0(P)\) and \(A(P)\) seem to be ubiquitous for \(f\)-electron systems near QCP, for example CeIn$_3$\[27\], CeRh$_2$Si$_2$ \[28, 29\] or CeAgSb$_2$ \[30\]. In these materials, the presence of magnetic critical fluctuations is strongly suggestive (except for the 1st - order transition in CeRh$_2$Si$_2$). On the other hand, UGe$_2$ (excluding the F2 phase) \[29, 31, 32\] and CeRh$_2$Si$_2$ (excluding the 2nd - order transition) \[28\], which exhibit the first order transition before reaching the QCP, do not have obvious maxima in \(\rho_0(P)\) and \(A(P)\) around the QCP, clearly indicating the absence of critical fluctuations. In the case of CeP, we consider the influence of the magnetic critical fluctuations with respect to \(\rho_0(P)\), \(A(P)\), and \(\alpha\).

\(\alpha\) \(\rho_0\) is expressed in most simply by the following equation: \(\rho_0 = (3 \pi^2)^{1/3} (\hbar/e^2) \ell^{-1} n^{-2/3}\), where \(\ell\) is the mean free path of electrons and \(n\) is electron density. The observed prominent peak at 5.3 GPa is caused by a significant decrease in either \(\rho\) or \(n\). Obviously, \(\rho_0\) decreases with increasing-P in all P-regions 2.5 – 8 GPa, except for the range of 5 – 5.5 GPa where a sharp pointed-peak is formed. This behavior directly indicates a distinct increase in \(n\) by pressure. This is strongly supported by our previous data \[24\]. Because the mean free path \(\ell\) at \(T = 0\) K is determined by impurities, defects, or dislocations and cannot be influenced by pressure. Consequently, an extreme decrease in \(\ell\) is most likely caused by the intense scattering of electrons due to the strong FM quantum critical fluctuations developing toward \(P_c\). This forms the very sharply pointed peak at 5.3 GPa by overcoming a
considerable increase in $n$. The theoretical viewpoint also predicts the formation of a sharp peak in $\rho_{0}(P)$ by critical fluctuations [33].

(B) $A(P)$ exhibits divergent behavior in the narrow $P$-region of 5–6 GPa. The position of the extremely sharpened peak at 5.5 GPa corresponds to the FM-instability $P_{C}$. This indicates that the FM quantum critical fluctuations correlate closely with the divergent behavior in $A(P)$. This behavior seems quite independent of the contribution of the magnetic domain.

(C) The results of $\alpha$ in the $\rho(T) \propto T^{\alpha}$ dependence are summarized as follows: (i) the region for 2.5–5.3 GPa: $\alpha \approx 3.4–3.9$; (ii) for the range 5.5–6 GPa: $\alpha \approx 2.2–2.6$; and (iii) $P > 6$ GPa: $\alpha \approx 0.95$–1.1. The $\alpha$ is strongly affected by the $T$-region for a fit to the data. Despite that the lowest arrival temperature of approximately 5 K in our experiments is too high, $\alpha$ in the regions (i) and (iii) is considered to be reliable. The $T$-variation of $\rho(T)$ for $T \leq 5$ K in region (i) and (iii) is strongly suggested to be gradual, as shown in Fig. 4; consequently, a significant change in $\alpha$ is not expected, even if we measured $\rho(T)$ for $T \leq 5$ K.

(I) The $\rho(T)$ at 2.5–5.3 GPa, in the magnetic order region (pure FM and FM+AFM-like phases), is most likely caused by the scattering of charge-carriers due to the FM magnons because the value of $\alpha \approx 3.3$–3.8 is very close to that of theoretical predictions ($\alpha = 4$) based on the dispersion relation $\omega(q) \propto q$ [34]. (II) The region 5.5–6 GPa nearest to $P_{C}$ is an uncertain area owing to the fault of our experiments. While measuring $\rho(T)$ at 5.5–6 GPa for $T \leq 5$ K, if $\alpha$ decreases from $\alpha \approx 2.2$–2.6 to $\alpha \leq 2$, Fermi liquid (FL) or NFL behavior will be observed, whereas if $\alpha > 2$, the characteristics of $\rho(T)$ are caused by the magnon contribution. (III) The $\rho(T)$ at $P > 6$ GPa exhibit a normal metallic-behavior for $T \leq 200$ K as shown in Ref. [24]. Furthermore, $\rho(T)$ at 6.5 GPa in Fig. 1 is almost invisible. These behavior strongly reflect that the metallic conductivity in $\rho(T)$ at $P > 6$ GPa is caused by the valence fluctuating state. Thus, $\alpha \approx 1$ in the region (iii) considered to be quite independent of the magnetic critical fluctuations in the vicinity of $P_{C}$ and the electronic properties.

5. Summary and conclusion
From the results based on consideration of (A), (B), and the ND experiments, and particularly (A), we emphasize that FM quantum critical fluctuations almost certainly exist around $P_{C}$; therefore, FM-QCP in the clean magnetic semimetal CeP with LM is located at $P_{C} \approx (5.5 \pm 0.1)$ GPa.

The pure FM-phase for $P \geq 4$ GPa destabilizes rapidly and is completely suppressed at $P_{C}$ with the simultaneous disappearance of $T_{C}$ and LM. It is noteworthy that the $\rho(T)$ curves at $P \geq P_{C}$ in Fig. 1 are quite similar to that of LaP with no 4f-electrons or that of metallic CeN [35]. This strongly suggests that the valence fluctuating state is realized in this $P$-region. We want to emphasize the possibility of the localization-itinerant transition of the Ce-4f states approximately at the FM-QCP.

We could not confirm the FL or NFL behavior in our experiments. If we could observe these behaviors, it would be limited to a very narrow $P$-range of 5.5–6 GPa on the $P$-$T$ plane.

The “various-striped” MSs for $P \geq 0.3$ GPa, consisting of ferromagnetically coupled $\Gamma_{8}$-like bilayers and $\Gamma_{7}$-single plans, are caused by the co-existence of the $\Gamma_{8}$- and $\Gamma_{7}$-states overcoming the large $\Delta_{7} \approx 162$ K [25]. Quite interestingly, the value of $\mu_{C} \approx 1.9\mu_{B}$ at 3 GPa [26] is very close to that of the full $\Gamma_{8}$ state $\approx 2\mu_{B}$. This strongly suggests that the ground state is very closed to the $\Gamma_{8}$ state. Such “various-striped” MSs cannot be explained by the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction. Kasuya et al. have proposed magnetic polaron effects based on strong $p_{3/2} (\Gamma_{8})$-$4f_{5/2} (\Gamma_{8})$ mixing to understand these magnetic properties [36]; moreover, the Kondo effect in a low-carrier density system is considerably questioned with the discovery of various “stripe-like” MSs [23, 25, 37]. As discussed so far, it is obviously inappropriate to describe FM quantum criticality under high-$P$ in the CeP low-carrier density system because of the normal competition between the Kondo effect and the RKKY interaction based on the Doniach model. We should discuss the $P$-induced quantum critical phenomena in CeP in greater detail based on a new concept that includes the charge degrees of freedom.
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