Non-Fermi-liquid behavior consistent with a preasymptotic disorder-tuned ferromagnetic quantum critical point in the heavy fermion system $CeTi_{1-x}V_xGe_3$

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An investigation of the thermodynamic and electrical transport properties of the isoelectronic chemical substitution series $CeTi_{1-x}V_xGe_3$ (CTVG) single crystals is reported. As x increases, the ferromagnetic (FM) transition temperature is suppressed, reaching absolute zero at the critical concentration $x = 0.4$, where a non-Fermi-liquid low-temperature specific heat and electrical resistivity, as well as the hyperscaling of specific heat and magnetization, are found. Our study suggests the presence of an FM quantum critical point (QCP) in CTVG. The obtained critical exponents indicate that CTVG falls in the preasymptotic region of the disorder-tuned FM QCP predicted by the Belitz-Kirkpatrick-Vojta theory.

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I. INTRODUCTION

The f-electron system exhibits diverse magnetic behaviors, which has attracted the attention of many researchers. This diversity in magnetic behavior arises from the competition between the magnetic Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, the nonmagnetic Kondo effect, and the crystal electric field effect. Because the system's ground state is delicately balanced among these three factors, nonthermal tuning parameters, such as magnetic field, pressure, and chemical substitution, are often used to alter the system's underlying physical behavior [1–3]. Among these, the most eye-catching physical behavior is when long-range magnetic order is continuously suppressed to absolute zero, reaching a magnetic quantum critical point (QCP).

Depending on the type of magnetic order before tuning, the magnetic QCP can be subdivided into two kinds: antiferromagnetic (AFM) and ferromagnetic (FM) QCPs. The AFM QCP seems easier to obtain than the FM QCP. The reason for this is that when suppressing the FM ordering temperature to zero, in addition to the inherent order parameter fluctuation energy, additional soft modes appear, leading to avoidance of a single fixed point [4]. The only known ferromagnet that can be tuned to a QCP by hydrostatic pressure is $CeRh_6Ge_4$ and its mechanism remains elusive [5,6]. Another more discernible approach is proposed by Belitz, Kirkpatrick, and Vojta (BKV), suggesting that it is possible to reinstate the FM QCP by introducing a suitable level of quenched disorder [4,7–9]. A handful of ferromagnets have confirmed BKV's prediction and are reported to exhibit QCPs, including $UCo_{1-x}Fe_xGe$ [10], $(Mn_{1-x}Cr_x)Si$ [11], $NiCoCr_x$ [12], $Ce(Pd_{1-x}Ni_x)_2P_2$ [13], and $Ni_{1-x}Rh_x$ [14,15]. In these studies, researchers observed the logarithmic or power-law divergence of the Sommerfeld coefficient and magnetic susceptibility as the temperature decreases. Some of these studies conducted scaling analyses to compare the critical exponents with those predicted by the BKV theory. The BKV theory also predicts the temperature dependence of the electrical resistivity, which probes how quasiparticles scatter from the order parameter fluctuations. However, studies on resistivity in disorder-tuned ferromagnets are rarely reported. One reason for this scarcity is that most of the research employs polycrystalline samples, where grain boundaries can compromise the data, leading to an ambiguous interpretation of the role of quantum fluctuations. Conducting resistivity measurements on single crystalline disorder-tuned ferromagnets are imperative, as it allows for a more comprehensive examination of the completeness of the BKV theory.

$CeTiGe_3$ has been reported as a rare FM Kondo system with a Curie temperature $T_C = 14$ K [16]. The FM order can be reduced by the hydrostatic pressure [17], suggesting that $CeTiGe_3$ is located at the FM side of the phase diagram of an

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FM Kondo lattice model [6]. This also implies that the transition is sensitive to chemical substitution, leading to the change in chemical pressure and modification of the band structure. By estimating the volume-scaled Sommerfeld coefficient, $CeTiGe_3$ sits on the fringe between a region of paramagnetism and a region of weak ferromagnetic order [18,19]. By substituting vanadium for titanium in $CeTi_{1-x}V_xGe_3$ (CTVG), researchers have successfully synthesized this compound in both polycrystalline and single crystalline forms. In both cases, the full suppression of FM order is observed at the critical concentration $x_{cr} = 0.4$. [20,21]. Fritsch *et al.* have shown logarithmic divergence of the Sommerfeld coefficient for the single crystal CTVG with $x = 0.405$, which hints at the existence of the QCP [22]. In this paper, we report electrical resistivity, magnetization, and specific heat measurements of CTVG single crystals, with a focus on the x_{cr} sample. The results provide several pieces of evidence for an FM QCP in CTVG and the obtained quantum critical exponents, as well as the temperature dependence of resistivity, are consistent with the framework of the BKV theory for disorder-tuned FM quantum criticality in the preasymptotic region.

II. RESEARCH METHODS

The CTVG single crystals were grown using the self-flux method, with varying values of x including 0, 0.1, 0.3, 0.4, 0.9, and 1.0. The detail of the growth has been described in Ref. [21]. For $0.8 > x > 0.4$, we were unable to obtain single crystals, and furthermore, these polycrystals often contain the magnetic impurity $CeGe_x$. Therefore, we omit these concentrations. A nonmagnetic reference $LaTi_{0.55}V_{0.45}Ge_3$ was grown using the arc-melting method. Specific-heat measurements employing thermal-relaxation calorimetry and the four-point-contact electrical resistivity were performed for current flow in the *a-b* plane in the temperature range from 0.05 to 300 K using a Dynacool physical properties measurement system (Dynacool PPMS, Quantum Design) equipped with a 9 T magnet. Magnetization measurements were carried out in a SQUID magnetometer (MPMS, Quantum Design) in the temperature range from 1.8 K to 300 K and in external fields up to 7 T. For all measurements the magnetic field is applied parallel to the hexagonal c axis.

III. RESULTS

A. Specific heat

Figure 1(a) shows the total specific heat of CTVG and nonmagnetic $LaTi_{0.55}V_{0.45}Ge_3$. As x increases from 0 to 0.3, the sharp FM transition around $T = 14$ K is suppressed to 7 K. As we approach lower temperatures, constant C/T is observed for $x = 0 - 0.3$, indicating Fermi-liquid (FL) behavior. For the $x = 0.4$, no transition could be observed down to 0.06 K, indicating that $x = 0.4$ is very close to a magnetic instability, i.e., the critical concentration x_{cr} at which the FM transition disappears. As we further increase the V concentration to $x = 0.9$ and 1.0, transitions appear around $T = 5 - 6$ K, below which a constant C/T indicating the FL behavior is observed. Below the transition temperatures for samples with $x = 0.9$ and 1.0, an AFM order was reported [21], and a helimagnetic

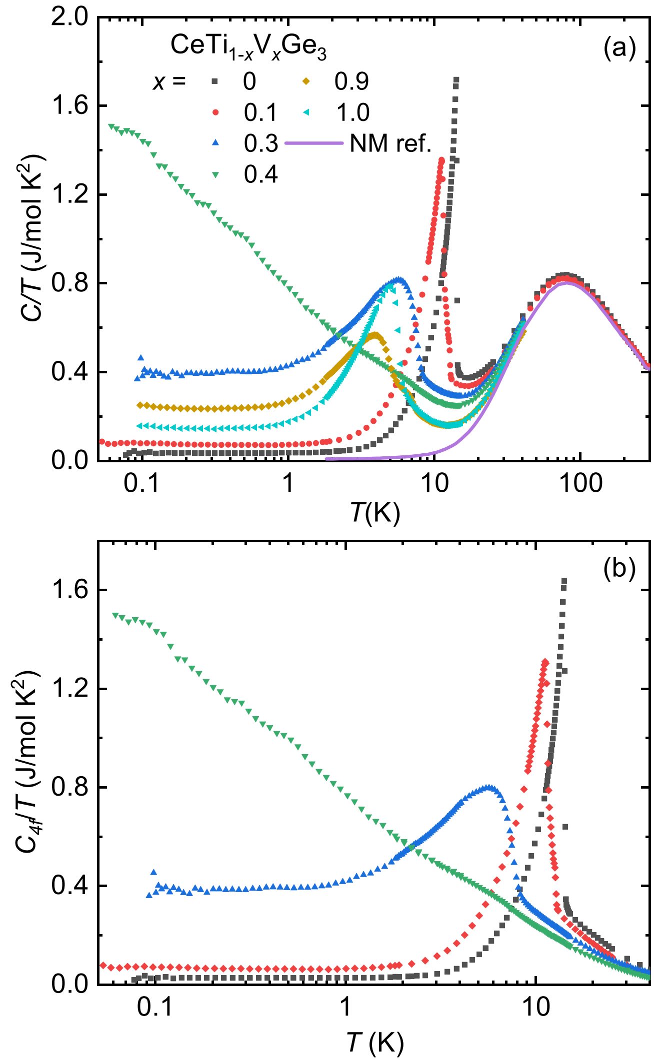

FIG. 1. (a) Zero-field specific heat of CTVG. NM ref. stands for nonmagnetic reference $LaTi_{0.55}V_{0.45}Ge_3$. (b) $4f$ contributions to the specific heat.

state characterized by moments rotating around the c axis has recently been fully elucidated [23].

We next focus solely on the FM side of CTVG ($x = 0 - 0.4$) and discuss the magnetic contribution to the specific heat. C/T of the nonmagnetic reference is subtracted from that of CTVG, and the result is shown in Fig. 1(b). For $x = 0 - 0.3$, C_{4f}/T exhibit a logarithmic increase as decreasing T from high temperature to right above respective T_C, and for $x = 0.4$ the divergence of C_{4f}/T extends widely between $T = 40$ and 0.06 K. Such an increase of C_{4f}/T at temperatures above 10 K can be described by the CEF splittings of the Ce^{3+} ground state doublet, as evidenced by the comparison of the experimental data and the simulated CEF curves for $x = 0 - 0.3$ in Figs. S2(a)–(c) in the Supplemental Material (SM) [19]. For $x = 0.4$, we have analyzed the CEF splittings based on the temperature dependence of the inverse magnetic susceptibility derived from the same piece of the sample for the specific heat measurement, and the constraint for the high temperature magnetic entropy. We found the first excited state $\Delta_1 = 12$ K and the second excited state $\Delta_2 = 65$ K (see Fig. S2(d) in

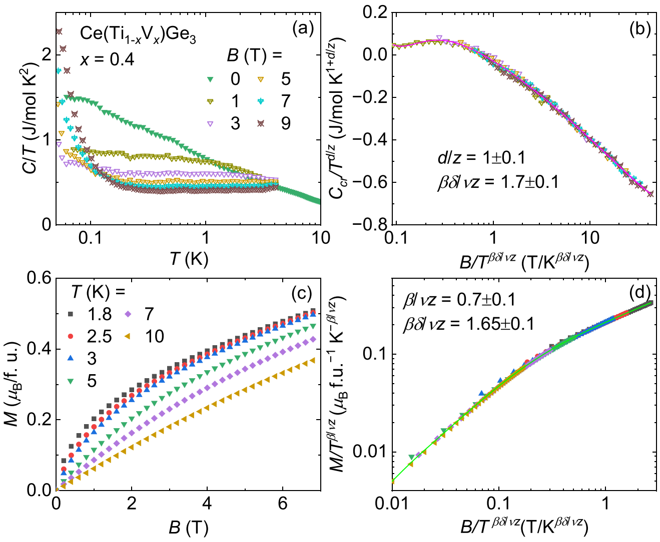

FIG. 2. (a) Specific heat of CTVG with $x = 0.4$ under different magnetic fields. (b) Scaling of C_{cr} as a function of T and B. (c) Isothermal magnetization curves measured at different temperatures. (d) Scaling of magnetization M as a function of T and B. The solid lines in (b) and (d) represent fits of polynomials to test the quality of the scaling.

Ref. [19]). Between $T = 10 - 40$ K, C_{4f}/T matches C_{CEF}/T quite well. Below 10 K, C_{4f}/T cannot be explained by the C_{CEF}/T. Instead, we observe non-Fermi-liquid (NFL) behavior, i.e., logarithmic divergence $C_{4f}/T = a \ln T/T_0$ with $a \sim -0.25$ J mol^{-1} K^{-2} and $T_0 \sim 26$ K, between 0.06 and 10 K, i.e., over more than two orders of magnitude in T. Our data is consistent with that reported by Fritsch *et al.* [22]. The magnetic entropy $S_{4f}(T)$ obtained from C_{4f} is shown in Fig. S1 in the SM [19]. S_{4f} exceeds $R\ln 2 = 5.76$ J/mol K at 16 K expected for a ground-state doublet. This is again due to the relatively small crystal-electric-field (CEF) splittings. If we express C_{4f}/T as $\gamma + C_{\mathrm{CEF}}/T$, where γ is the Sommerfeld coefficient, the observation of logarithmic divergence in C_{4f}/T between 0.06 and 10 K, even with the presence of the low CEF splitting, implies the strong hybridization between the conduction and f electron states, i.e., γ is dominant in C_{4f}/T [24].

A straightforward way to qualitatively explain the NFL behavior at a magnetic instability is to consider the presence of numerous low-energy magnetic excitations when the $T_c \rightarrow 0$ [25]. This conjecture is supported by the recovery of FL behavior in high magnetic fields. Figure 2(a) shows the magnetic field dependence of C/T of the $x = 0.4$ sample. After subtraction of the nuclear contribution to the specific heat C_{nuclear} which causes an upturn at the lowest temperature, Fig. S3 in the SM shows the tendency toward FL behavior $(C - C_{\mathrm{nuclear}})/T$ = constant is strengthened as the field increases [19].

B. Hyperscaling analysis on magnetization and specific heat

Due to the insensitivity to low CEF splittings in specific heat, we are prompted to conduct further studies on the quantum criticality in CTVG. At a QCP below the upper critical dimension, the hyperscaling relation of the quantum critical part of the specific heat C_{cr} is given by

$$\frac{C_{\mathrm{cr}}(T,B)}{T^{d/z}} = \Psi\left(\frac{B}{T^{\beta\delta/\nu z}}\right), \tag{1}$$

where d is the spatial dimension, and z and $\beta\delta/\nu$ are critical exponents associated with the tuning parameters T and B, respectively [2,15]. Equation (1) indicates that C_{cr}/T is a universal function of $B/T^{\beta\delta/\nu z}$. To eliminate noncritical quasiparticle contribution to the electronic specific heat, we determine $C_{cr} = C(T,B) - C(T,0)$ (C_{nuclear} subtracted) and plot $C_{cr}/T^{d/z}$ vs $B/T^{\beta\delta/\nu z}$. Excellent scaling over more than two orders of magnitude of $B/T^{\beta\delta/\nu z}$ with $d/z = 1 \pm 0.1$ and $\beta\delta/\nu z = 1.7 \pm 0.1$ is shown in Fig. 2(b) (also see Fig. S2 in [19]). As C is tightly bound to the Gibbs free energy $\mathcal{F}$, from which the isothermal magnetization M can be obtained, it is expected M follows similar scaling behavior near a QCP, i.e.,

$$\frac{M(T,B)}{T^{\beta/\nu z}} = \Phi\left(\frac{B}{T^{\beta\delta/\nu z}}\right). \tag{2}$$

Figure 2(c) shows $M(B)$ measured at different temperatures. Using Eq. (2), excellent scaling over more than two orders of magnitude of $B/T^{\beta\delta/\nu z}$ with $\beta/\nu z = 0.7 \pm 0.1$ and $\beta\delta/\nu z = 1.65 \pm 0.1$ is obtained and shown in Fig. 2(d) (also see Fig. S2 in the SM [19]). Below 1 T, noncritical contributions to M cause deviations from scaling behavior, and hence the data are omitted. While the exact forms of Ψ and Φ in Eqs. (1) and (2), respectively, determined by the details of $\mathcal{F}$ are not clear at the present stage, the scaling results still strongly suggest the existence of an FM QCP in CTVG with $x = 0.4$.

C. Electrical resistivity

In addition to the NFL behavior in the thermodynamic properties, at low temperatures, the scattering caused by quantum fluctuations drives quasiparticles critical, and this phenomenon is best represented by electrical resistivity measurements, as shown in Fig. 3. For $x = 0$ and 0.1, below the FM transition the $\rho(T)$ data can be described by

$$\rho = \rho_0 + AT^2 + BT\Delta\left(1 + 2\frac{T}{\Delta}\right)e^{-\frac{\Delta}{T}}, \tag{3}$$

where ρ_0 is the residual resistivity, A is the coefficient for electron-electron scattering, B is the coefficient for electron-magnon scattering, and Δ is an energy gap in the magnon excitation spectrum [21]. The fits of Eq. (3) are shown as dashed lines in Fig. 3(a). For $x = 0.3$ and 0.4, Eq. (3) failed to fit the low-T data, indicating a complicated evolution of the interplay between the Kondo effect, the CEF effect, and the quantum fluctuations. The low-T part of $\rho(T)$ for $x = 0.4$ is shown in Fig. 3(b). The data shows appreciable NFL behavior, i.e., $\rho(T) = \rho_0 + aT^{\alpha}$ with $\alpha \sim 1.2$, in the temperature range between 1 K and 0.1 K. The power α evolves to $\sim$0.88 between 5 K and 1 K. At such a low temperature, the CEF effect is negligible as evidenced by the isotropic magnetic susceptibility [19]. Therefore, the evolution of α may be due to the coupling of the Kondo effect to the ferromagnetic fluctuations. To quantitatively determine the contribution to ρ due to the Kondo effect is beyond the scope of our work.

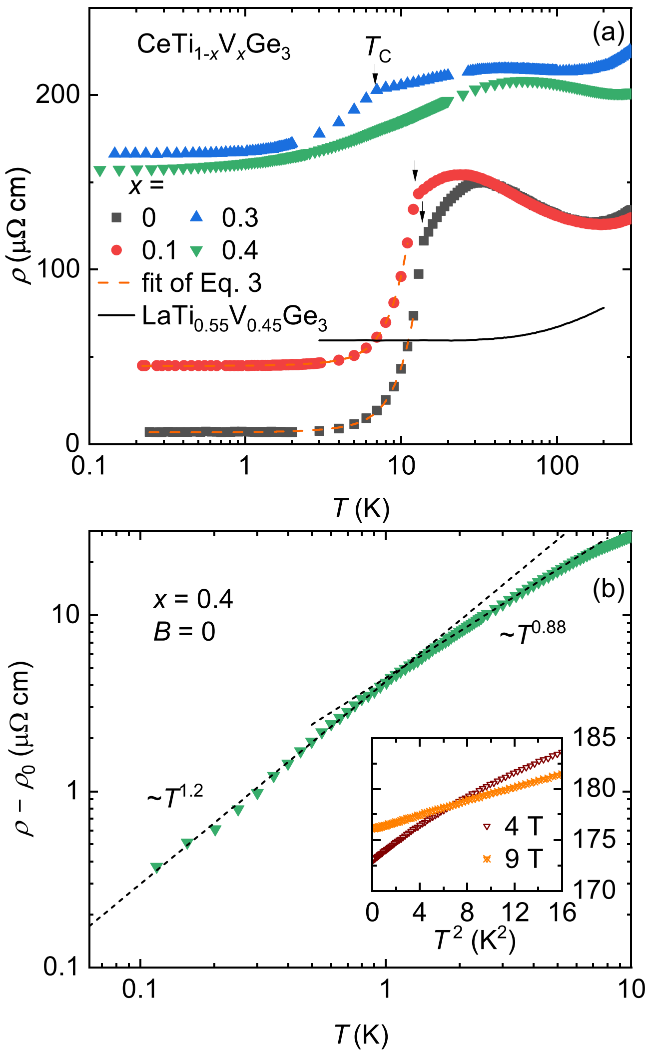

FIG. 3. (a) Zero-field electrical resistivity of $CeTi_{1-x}V_xGe_3$ with $x = 0, 0.1, 0.3, 0.4$ and $LaTi_{0.55}V_{0.45}Ge_3$. Arrows indicate the FM ordering temperature. (b) Low-temperature electrical resistivity of $x = 0.4$, obeying $\rho - \rho_0 = AT^\alpha$, with $\rho_0 = 156.85\ \mu\Omega$ cm, $\alpha \sim 1.2$ between 1 and 0.1 K, and $\alpha \sim 0.88$ between 5 and 1 K. The inset shows $\rho(T)$ vs T^2 for $B = 4$ and 9 T.

A large enough magnetic field should suppress the Kondo effect and the low-lying magnetic excitations, leading to a recovery of the FL behavior. The inset of Fig. 3(b) shows that $B = 9$ T indeed leads to a recovery of a $\rho(T) = \rho_0 + aT^2$ law. Our $\rho(T)$ data reinforces the evidence of the FM QCP in CTVG with $x = 0.4$. The $\rho(T)$ of the nonmagnetic reference is temperature independent below 30 K, as shown in Fig. 3(a). Therefore, the temperature dependence of $\rho(T)$ of $x = 0.4$ sample is irrelevant of electron-phonon scattering. The relatively high $\rho_0 = 156.85\ \mu\Omega$ cm observed for $x = 0.4$ may be attributed in part to quantum fluctuations close to the QCP [26].

IV. DISCUSSIONS

The source of NFL behavior within the CTVG system, as previously explained, can be linked to its proximity to a magnetic instability occurring at $x = 0.4$ at absolute zero. In some doped systems, the NFL behavior could be attributed to a distribution of the Kondo temperature T_K originated from inhomogeneities [27]. Such an effect could be neglected in CTVG as the NFL behavior only exists at the $x = 0.4$ sample.

Next we discuss the nature of quantum criticality in CTVG. In $4f$ heavy fermion systems upon increasing tuning parameters, the competition between the RKKY coupling among the moments and the Kondo interaction varies. Two classes of QCPs have been identified [28]: the Kondo-breakdown QCP, where the critical fluctuations encompass both the fluctuations of the magnetic order parameter and the breakdown of the Kondo effect, and the spin-density-wave (SDW) QCP, where critical fluctuations involve only fluctuations of the magnetic order parameter. Quantum critical fan is a generic feature of Kondo breakdown QCPs [5,29]. Since CEF can strongly affect the resistivity in the paramagnetic state due to small CEF splittings and there is no data for x>0.4 of CTVG, it is not clear whether there is a quantum critical fan in this compound. To clarify this issue, one needs single crystals of CTVG for x>0.4, which goes beyond the scope of our present work and requires further study. Nevertheless, to proceed our analysis, in the following we assume the SDW scenario within the BKV theory of an itinerant picture to play the dominant role here, and try to understand our results within this framework.

For a clean ferromagnet, the SDW-type QCP proposed originally by Hertz [30] is inherently unstable so that the QCP is disrupted by competing phases or first order phase transitions as $T_C \to 0$ [3,31]. The BKV theory suggests it is only possible to stabilize an FM QCP by introducing disorder. For CTVG, the fact that the ferromagnetism is destroyed by increasing the vanadium concentration could logically classify the QCP in CTVG within the framework of the BKV theory. Our hyperscaling of $C_{cr}(T, B)$ gives rise to the exponent $d/z = 1 \pm 0.1$ [Fig. 2(b)]. The ratio of the 2 K magnetic susceptibility with the magnetic field applied parallel and perpendicular to the c axis is $\sim$1 for $x = 0.4$, suggesting $d = 3$, as shown in Fig. S4 [19]. Therefore, $z = 3$ for CTVG. The hyperscaling analysis result, i.e., $d/z = 1$, is consistent with the prediction of the BKV theory in the dirty limit [9]. The critical exponent $\delta = 2.4$ deduced from the hyperscaling of $M(T, B)$ [Fig. 2(d)] is slightly larger than the value predicted by the BKV theory ($\delta = 1.8$), which implies CTVG is in the preasymptotic region, i.e., strong disorder. Given the fact that the ρ_0 amounts to $\sim$157 $\mu\Omega$ cm for $x = 0.4$, much larger than the values for $x = 0$ and 0.1, our system indeed falls in the strong-disorder regime of the BKV theory. Moreover, the resistivity data shows $\Delta\rho \propto T^{1.2}$ at the QCP in CTVG, as shown in Fig. 3(b). The power of 1.2 is the value one expects for Hertz's fixed point in the presence of strong disorder [9]. We notice that the NFL behavior in C_{4f}/T and ρ is observed in different temperature windows. A similar phenomenon has been observed in an AFM QCP $YbRh_2(Si_{0.95}Ge_{0.05})_2$, where quasiparticles in the quantum critical region may break up into spin (spinon) and charge (holon) excitations, dominating in different temperature regions for different measurements [29,32]. Whether this scenario applies here needs further investigation.

V. CONCLUSION

To conclude, CTVG has revealed several pieces of evidence of the FM QCP where the NFL behavior is registered in temperature dependence of C_{4f}/T and ρ, as well as the existence of the hyperscaling on M and C_{cr}. The obtained critical exponents and the temperature dependence of ρ both agree well with the BKV theory in the strong-disorder regime. In the future, inelastic neutron scattering measurements on CTVG will be indispensable which allow us to quantitatively resolve the CEF splittings, especially for the samples in the vicinity of the FM QCP. This will help individually clarify the roles of different Ce^{3+} doublets in terms of the quantum criticality.

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