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Ferromagnetic quantum critical behavior in heavy-fermion compounds CeTi$_{1-x}$Ni$_x$Ge$_3$

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Abstract

The measurements on magnetization ($M$), resistivity ($\rho$) and specific heat ($C$) were carried out for the ferromagnetic CeTi$_{1-x}$Ni$_x$Ge$_3$ (0.0 $\leq$ $x$ $\leq$ 0.45) system. It was found that the Curie temperature, $T_C$, decreases with increasing Ni content, $x$, and reaches zero kelvin near a critical content $x_{cr} = 0.44$. A new phase diagram is constructed based on these measurements. The non-Fermi liquid (nFL) behavior in $\rho(T)$, and log($T_C/T$) relationship in $C/T$ in the samples near $x_{cr}$, demonstrate that strong spin fluctuation emerges in these samples, indicating that they are near a quantum critical point (QCP). Our results indicate that CeTi$_{1-x}$Ni$_x$Ge$_3$ may provide another platform to study exotic quantum phenomena near ferromagnetic QCP.

1. Introduction

Quantum criticality continuously raises scientists’ research interest because it is believed to be the origin of the many exotic emergent phenomena in modern condensed-matter physics, such as quantum phase transition (QPT), new hidden order, unconventional superconductivity and the breakdown of the Fermi liquid model [1, 2]. The QPT, which occurs at zero kelvin, is driven by quantum fluctuations, in contrast to the phase transition occurring at finite temperature, driven by thermal fluctuations. The existence of ferromagnets with a lower Curie-temperature ($T_C$) makes the ferromagnetic quantum criticality visible because their $T_C$ are easily suppressed to zero kelvin by tuning the external parameters, such as chemical composition, or pressure $P$. Most of the experimental studies on the QPT were devoted to the stoichiometric compounds based on ytterbium, uranium or cerium, which show antiferromagnetic correlation [1, 3]. On the other hand, ferromagnetic QPT is quite unusual both from an experimental and theoretical point of view [1, 3, 4]. Therefore it is especially attractive to investigate ferromagnetic alloys in which $T_C$ can be driven to zero kelvin by tuning external parameters. For example, in URhGe, [5] UCoGe [6] and UGe$_2$ [7], applying pressure tuned ferromagnetic QPT has been realized. In CePt$_{1-x}$Rh$_x$ [8], CePt$_{1-x}$Ni$_x$ [9], CeTi$_{1-x}$V$_x$Ge$_3$ [10], YbNi$_2$P$_2$ [11], URu$_{2−x}$Re$_x$Si$_2$ [12] and UCo$_{1−x}$Fe$_x$Ge [13], the $T_C$ can be driven to zero kelvin by a chemical substitution.

CeTiGe$_3$ is an anisotropic ferromagnetic Kondo-lattice system with $T_C = 14$ K at ambient pressure [10]. It crystallizes in the BaNiO$_3$ type structure (hexagonal perovskite, $P6_3/mmc$), which consists of one-dimensional chains of face shearing Ti-centered octahedra stacked along the c-axis. The short distance between Ti–Ti atoms has been taken as a sign of a weak metal–metal bonding [14, 15]. The spins of Ce$^{3+}$ ions with a moment of (1.5 $\pm$ 0.1)$\mu_B$/Ce order are ferromagnetic at low temperatures. It was found [10] that the ferromagnetic order in CeTiGe$_3$ is difficult to suppress by applying hydrostatic pressure. However, CeNiGe$_3$, is an antiferromagnet with a Neel temperature $T_N = 5.5$ K due to the localized 4$f$ magnetic moment. CeNiGe$_3$ crystallizes in the SmNiGe$_3$ type orthorhombic crystal structure [16]. Recently, Kittler and his coworkers performed a detailed investigation on the ferromagnetic QPT in the CeTi$_{1−x}$V$_x$Ge$_3$ system [10]. Here we show that the ferromagnetic order can be suppressed by the partial substitution of Ni for Ti in CeTi$_{1−x}$Ni$_x$Ge$_3$ system. It was found that $T_C$ decreases with
increasing Ni content, and reaches zero kelvin near a critical content $x_{c} = 0.44$. A new phase diagram is constructed based on the measurements of magnetization ($M$), resistivity ($\rho$) and specific heat ($C$) for this system. The non-Fermi liquid (nFL) behavior in $\rho(T)$, and the log($T_{C}/T$) relationship in $C/T$ in the samples near $x_{c}$ demonstrate that strong spin fluctuation emerges in these samples, indicating that they are near a quantum critical point (QCP).

2. Experimental details

Alloys of CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0.0 \leq x \leq 0.45$) were synthesized by an arc-melting method under an argon (Ar) atmosphere. The starting elements of Ce ingot (99.9%), Ti powder (99.9%), Ni slugs (99.9%) and Ge pieces (99.999%) were stoichiometrically mixed and put in a water cooled copper hearth. Each sample was re-melted several times with overturn to improve its homogeneity. After arc-melting, the samples were sealed in a Ta tube with a half Ar atmosphere and finally sealed in quartz glass tubes under high vacuum. The quartz tubes were then annealed for 10 days at 950 °C. The weight loss was found to be below 1% for each sample during the above processes. The x-ray powder diffraction (XRD) pattern for all the samples was recorded at room temperature with a PANalytical x-ray diffractometer (Model EMPYREAN). Analysis of the XRD data was made by using the GSAS suite of Rietveld programs [17]. The magnetization measurements were carried out by a Quantum Design MPMS (SQUID). The magnetic susceptibility of all the samples were measured at a 1000 Oe magnetic field with a process of field-cooling (FC). The resistivity and heat capacity measurements were carried out in a Quantum Design Physical Properties Measurement System.

3. Results and discussions

3.1. Crystal structure

Figure 1(a) shows the powder XRD pattern of the un-doped CeTiGe$_3$ sample, together with its Rietveld refinement (weighted profile factor, $R_{wp} = 2.4\%$, and the goodness-of-fit, $\chi^2 = 1.67$). All peaks could be well indexed with the hexagonal structure (BaNiO$_3$-type, space group P6$_3$/mmc), the lattice parameters of $a = 6.345 (\pm 0.001)$Å and $c = 5.912 (\pm 0.001)$Å were obtained. Figures 1(b)–(e) show the XRD patterns for CeTi$_{1-x}$Ni$_x$Ge$_3$ ($x = 0.1, 0.4, 0.44$ and $0.45$) samples, respectively, indicating that all the alloys of CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0.0 \leq x \leq 0.44$) are a single phase. A small amount of unknown impurity emerges in the $x = 0.45$ sample, indicating that there is a substitution limit of Ni for Ti. As shown in figures 1(f) and (g), with increasing Ni content, the lattice parameters, $a$ and $c$, values, decrease monotonically, implying that Ni$^{2+}$ can uniformly be substituted for Ti$^{3+}$ in the CeTi$_{1-x}$Ni$_x$Ge$_3$ lattice, as $x \leq 0.44$. The following Vegard’s law of lattice parameters demonstrates that the Ce$^{3+}$ ion remains its valency in this composition range. The relative change, $\Delta a/a$ and $\Delta c/c$, within the range of $0 \leq x \leq 0.45$ is about 0.072 and 0.017.

3.2. Magnetization

Figure 2(a) shows the temperature dependence of magnetic susceptibilities, $\chi(T)$, measured with an FC process for CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0.0 \leq x \leq 0.45$) samples. For the un-doped sample CeTiGe$_3$, a ferromagnetic transition occurs at $T_C = 14$ K, which is characterized by a sharp increase in susceptibility near $T_C$. Partial substitution of Ni for Ti results in the decrease of $T_C$, and ferromagnetic order disappearing in the $x_{c} = 0.44$ sample. Since the radius of Ni$^{2+}$ is smaller than that of Ti$^{3+}$, it is natural to suggest that the chemical pressure due to the partial substitution of Ni$^{2+}$ for Ti$^{3+}$ results in the decrease of $T_C$ in this system. However, no realization by applying hydrostatic pressure [10] indicates that the shrinking of unit cell volume here may not be the main reason for the suppression of ferromagnetism. It is possible that the increment of the state density at the Fermi level due to Ni$^{2+}$ (3$d^8$) replacing Ti$^{3+}$ (3$d^2$) enhances the Kondo coupling. In order to get much information about the magnetic properties in these samples, we fitted the susceptibility data above 50 K using the modified Curie–Weiss law [18]

$$\chi_T \approx \chi_0 + \frac{C}{T - \theta} = \chi_0 + \frac{N_m \mu^2_{\text{eff}}}{3k_B(T - \theta)}$$

(1)

where $\chi_0$ denotes the temperature-independent susceptibility, $\mu_{\text{eff}}$ is the effective magnetic moment of Ce, $N_m$ the number of Ce atoms per mole, $k_B$ the Boltzmann constant and $\theta$ the Curie–Weiss temperature. As shown in figure 2(b), the estimated Curie–Weiss temperature ($\theta$) increases from $-38.5$ K for CeTiGe$_3$ to $-26.1$ K for CeTi$_{0.95}$Ni$_{0.05}$Ge$_3$, while the effective moment $\mu_{\text{eff}} = 2.639 \mu_B$, being close to the value expected for Ce$^{3+}$ ions, is almost independent of Ni content. The negative $\theta$ values have been found in most Kondo compounds, and the absolute values of $\theta$ are associated with Kondo temperature ($T_K$) for overall crystal field levels, such as $T_K \sim |\theta|/2$ [16]. Using this relation, we estimate the Kondo temperature ($T_K$) to be $19.2 \text{ K}$ and $13.1 \text{ K}$ for $x = 0.0$ and $0.44$, respectively.
and 0.45 samples, respectively. In addition, the 4f electrons of Ce in CeTi$_{1-x}$Ni$_x$Ge$_3$ are partially delocalized, as found in most Ce-based intermetallic compounds. Because the effective moment ($\mu_{\text{eff}}$) remains a constant for all the partial substituting samples, it can be concluded that the degree of delocalization of the Ce 4f electrons may unchanged by the partial replacing of Ni$^{2+}$ for Ti$^{3+}$.

In order to precisely determine $T_C$ for CeTi$_{1-x}$Ni$_x$Ge$_3$ samples, we measured the isothermal magnetization $M(H)$ curves at different temperatures. As shown in figure 3, the isothermal magnetization Arrott plot ($M^2$ versus $H/M$) curves for CeTi$_{1-x}$Ni$_x$Ge$_3$ samples exhibit a linear behavior at high magnetic fields. A similar behavior has also been observed in various itinerant ferromagnets, such as Sr$_{1-x}$Ca$_x$RuO$_3$ [19], Y(Co$_{1-x}$Al$_x$)$_2$ [20], ZrZn$_2$ [21] and Ni$_3$Al$_{1-x}$Ga$_x$ [18]. It is easy to determine the $T_C$ by the temperature at which the Arrott plots at higher magnetic field extrapolate linearly to the origin point ($H = 0, M = 0$). Using this method, we determined precisely the $T_C$ of 14, 11.5, 6, 2 and $<2$K for $x = 0.0, 0.1, 0.3, 0.4$ and 0.44 samples, respectively. It is

Figure 1. (a) X-ray diffraction pattern of CeTiGe$_3$ with its Rietveld refinement. (b)–(e) Powder XRD pattern of CeTi$_{1-x}$Ni$_x$Ge$_3$ ($x = 0.1, 0.4, 0.44$ and 0.45) alloys, respectively. Lattice parameter (f) $a$ and (g) $c$ as a function of the Ni content $x$ for CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0.0 \leq x \leq 0.45$).
obvious that $T_C$ decreases monotonously with increasing $x$. It should be noted that $T_C$ for the $x = 0.44$ sample is lower than 2K, which is the lowest temperature in our measurement machine, indicating that the $x = 0.44$ sample may locate near a ferromagnetic QCP.

### 3.3. Electrical resistivity

Figure 4 shows the temperature dependence of electrical resistivity, $\rho(T)$, for CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0.0 \leq x \leq 0.45$) alloys. The $\rho(T)$ curves for the $x < 0.44$ samples exhibit typical behavior for the ferromagnetic Kondo-lattice system. For the pure CeTiGe$_3$ ($x = 0$) sample, with decreasing temperature, the resistivity decreases at first, reaches a minimum at about 130 K, then increases to a maximum ($\sim 182 \, \mu\Omega$ cm) at about 25 K, and finally drops sharply to $\sim 48 \, \mu\Omega$ cm at 2 K. This sharp drop in $\rho$ is related to the ferromagnetic transition, as observed in the ferromagnetic Kondo-lattice compounds CeAgSb$_2$ [22] and CeVGe$_3$ [23]. As shown in the inset of figure 4(a), the temperature at which the sharp drop in $\rho$ starts to occur, decreases with increasing Ni content, and becomes invisible in the $x = 0.44$ and 0.45 samples. Only for the $x = 0.45$ sample does its $\rho(T)$ exhibit a metallic behavior in the whole temperature range, i.e. its resistivity decreases monotonously with decreasing temperature.

In order to explore the influence of spin fluctuation near QCP on $\rho(T)$ behavior as $T_C$ approaches zero kelvin, we analyze the $\rho(T)$ data for all the CeTi$_{1-x}$Ni$_x$Ge$_3$ samples. As discussed by Sidorov et al [22] for the ferromagnetic Kondo-lattice compounds CeAgSb$_2$, and by Inamdar et al [23] for CeTiGe$_3$, in the ferromagnetic state, the $\rho(T)$ data over a wider temperature range can be described by the expression:

$$\rho(T) = \rho_0 + AT^2 + B \Delta(1 + 2T/\Delta)e^{-\Delta/T}$$  \hspace{1cm} (2)$$

where $\rho_0$ is the residual resistivity, $A$ is the coefficient of the Fermi-liquid $T^2$ term, the third term is the contribution due to an energy gap in the magnon dispersion relation [24]. In equation (2), $B$ involves the electron-magnon scattering and $\Delta$ is the magnitude of the gap. As shown in figures 4(b)–(f), it was found that, for all the ferromagnetic samples ($x \leq 0.40$), the $\rho(T)$ data below $T_C$ can be described by equation (2). For the $x = 0.0$ sample, the energy gap in the spin wave excitation spectrum $\Delta = 25$ K was inferred from the fitting.
which is consistent with the result reported by Inamdar et al [23]. They found that [23] the energy gaps for a CeTiGe$_3$ single crystal were 28 K and 23 K along the a and c axis, respectively. In addition, we found that the energy gap $\Delta$ value decreases with increasing Ni content, i.e. $\Delta = 16, 12, 10, 9$ K for $x = 0.1, 0.2, 0.3$ and 0.4 samples, respectively. The $\rho_0$ and $A$ values increase with increasing Ni content, i.e. $\rho_0 = 48, 86, 109, 130, 137 \mu\Omega$ cm and $A = 0.081, 0.295, 0.48, 0.81,$ and $1.58 \mu\Omega$ cm/$K^2$ for $x = 0.1, 0.2, 0.3$ and 0.4 samples, respectively. The good fitting of equation (2) to the $\rho(T)$ data in the ferromagnetic state indicates that there are two types of electrons in these samples. One is an itinerant part which exhibits Fermi-liquid behaviors, and the other is localized whose spins are ordered ferromagnetically. As discussed above, the increase of the state density at the Fermi level due to Ni$^{2+}$ ($3d^8$) replacing Ti$^{3+}$ ($3d^2$) enhances the Kondo coupling, then results in the decrease of $T_C$. But no change in $\mu_{\text{eff}}$ for all the partial substituting samples implies that the degree of delocalization of the Ce

![Figure 3. The Arrott plots, $M^2$ versus $H/M$ for the CeTi$_{1-x}$Ni$_x$Ge$_3$ ($x = 0.0, 0.1, 0.3, 0.4, 0.44$ and $0.45$) samples at different temperatures.](image)
4f electrons may be unchanged. For the $0 < x \leq 0.44$ samples, in which the ferromagnetic order disappears, it was found that the $\rho(T)$ data, even in the lowest temperature range ($0.5 \leq T \leq 2.5$ K), cannot be described by both Fermi-liquid behavior, $\rho(T) = \rho_0 + AT^2$, and equation (2). Then we try to fit the $\rho(T)$ data at lower temperatures using a power-law: $\rho(T) = \rho_0 + A'T^n$, where $A'$ is the temperature-coefficient and $n$ is the power-exponent, as shown in figure 4(g). It was found that the values of $n$ for $x = 0.44$ and 0.45 samples are smaller than 2.0. In particular for the $x = 0.44$ sample, $n$ reaches a minimum value of 1.6. Except for the $n$ value deviating from the Fermi-liquid value, both the residual resistivity $\rho_0$ and the temperature coefficient $A'$ values reaches a maximum. For example, $\rho_0 = 48 \mu \Omega$ cm for the $x = 0.0$ sample, while $\rho_0 = 141.9 \mu \Omega$ cm for the $x = 0.44$ sample, which is almost three times the former, and $A = 0.081 \mu \Omega$ cm/K$^n$ ($x = 0.0$), and $1.821 \mu \Omega$ cm/K$^n$ ($x = 0.44$). The maximum in $\rho_0$ and $A'(A')$ appear to be ubiquitous [1] for heavy-fermi materials in the vicinity of the QCP, such as the pressure-tuned YbFe$_2$Ge$_2$ system [25].

Figure 4. (a) Temperature dependence of the electrical resistivity, $\rho(T)$, for the CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0.0 \leq x \leq 0.45$) samples. The inset shows the low temperature data of $\rho(T)$, and the arrows correspond to the $T_C$. (b)-(g) The $\rho(T)$ at lower temperature for $x = 0.0$, 0.1, 0.2, 0.3, 0.4 and 0.44 samples, the red lines are the fitting of equation (2) or a power law, as discussed in the text.
Figure 5. (a) Temperature dependence of specific heat, $C(T)$, for CeTi$_{1-x}$Ni$_x$Ge$_3$ $(0.0 \leq x \leq 0.45)$ samples. Red line: the fitting to $C(T)$ data for $x = 0.0$ sample. Navy blue: the fitting to $C(T)$ data for the $x = 0.44$ sample, as discussed in the text.

3.4. Specific heat

Figure 5(a) shows the temperature dependence of specific heat, $C(T)$, for CeTi$_{1-x}$Ni$_x$Ge$_3$ $(0.0 \leq x \leq 0.45)$ samples. It is clear that a $\lambda$-type anomaly in $C(T)$, corresponding to the ferromagnetic transition, emerges at $T_C$ in the CeTi$_{1-x}$Ni$_x$Ge$_3$ $(0.0 \leq x \leq 0.4)$ samples, and the $T_C$ decreases with increasing Ni content. For the $x = 0.44$ and 0.45 samples, no anomaly in $C(T)$ was observed above 0.5 K. In order to get more information regarding $C/T$ near QCP, we replotted the $C/T$ data as $C/T$ versus $\log T$, as shown in figure 5(b). A small peak at 2 K was also observed for the $x = 0.40$ sample, indicating its $T_C \sim 2$ K, which was also observed in $\rho(T)$, as shown in figure 4(f). First, we discuss the $C(T)$ data in the ferromagnetic state for the $x = 0.0, 0.1, 0.2, 0.25$ and 0.3 samples. As discussed by Inamdar et al [23] for CeTiGe$_3$ and by Sidorov et al [22] for CeAgSb$_2$, their $C(T)$ data below $T_C$ could be fitted to the magnetic spin wave gap expression [23, 26]:

$$C(T) = \gamma_0 T + \beta T^3 + \alpha (\Delta^2 / \sqrt{T} + 3\Delta \sqrt{T} + 5\sqrt{T}^3) e^{-\Delta/T}$$ (3)

where the first term is the specific heat from electrons, the second term is the contribution from lattice vibration, the third term represents the contribution to the specific heat of magnons with a gap in the ferromagnetic state. For clarity, we plotted only the fitting line (red) below 10 K for the $x = 0$ sample in figure 5(b). The fitting parameters, the electronic specific heat coefficient $\gamma_0$, the lattice specific heat coefficient $\beta$, the magnon specific heat coefficient $\alpha$ and its gap $\Delta$, are listed in table 1. For the CeTiGe$_3$ ($x = 0$) sample, the values of $\gamma_0, \alpha$ and $\Delta$ are consistent with that reported by Inamdar et al [23] and the value of gap, $\Delta = 29$ K, is comparable to the value derived from the resistivity. Combined with the $A$ value inferred from the resistivity, we calculated the Kadowako–Woods ratio, $A/\gamma^2 = 1.35 \times 10^{-5}$ $\mu\Omega$ (mol K$^2$ mJ)$^2$ for the $x = 0$ sample, which is consistent with the typical value $\sim 10^{-5}$ $\mu\Omega$ (mol K$^2$ mJ)$^2$ for the standard heavy-fermion systems [27, 28], and the value $1.33 \times 10^{-5}$ $\mu\Omega$ (mol K$^2$ mJ)$^2$ for the CeTiGe$_3$ single crystal [23],
where $\alpha = 0.075 \mu\Omega \cdot \text{cm} \cdot \text{K}^{-2}$ and $\gamma_0 = 75 \text{mJ/mol K}^2$. From table 1, it is obvious that $\gamma_0$ increases, while $\alpha$ decreases with increasing $x$, indicating that the partial substitution of Ni for Ti results in the enhancement of electronic specific heat and the decrease in magnon specific heat.

Now, turn to the $C(T)$ data for the $x = 0.44$ and 0.45 samples in which the long range ferromagnetic order disappears. As shown in figure 5(b), their $C/T$ versus $\log T$ exhibits a linear behavior, and does not show a saturation at the lowest temperature, which is a typical behavior for the system with strong spin fluctuation, indicating that the $x = 0.44$ and 0.45 samples are indeed located near a QCP. We try to fit the $C(T)$ data for both samples using the expression [29, 30]

$$C/T = \frac{\gamma_0}{T} + \frac{\beta T^2}{T} + \delta \log(T_0/T)$$

where $\delta$ is a coefficient and $T_0$ presents a characteristic temperature related to the spin fluctuation term. For clarity, only the fitting line (navy blue) below 10 K for the $x = 0.44$ sample is shown in figure 5(b). It was found that the $C(T)$ data for both samples at low temperatures can be very well described by equation (4). The fitting parameters, $\gamma_0$, $\beta$, and $\delta$ for the $x = 0.44$ and 0.45 samples are listed in table 2. Similar behavior was also observed at the ferromagnetic QCP in the Ni$_x$Pd$_{1-x}$ system [30] and the antiferromagnetic QCP in CeCu$_{5.9}$Au$_{0.1}$ [31].

For all the CeTi$_{1-x}$Ni$_x$Ge$_3$ ($0 \leq x \leq 0.45$) samples, from tables 1 and 2, it was clear that the $\gamma_0$ value increases with increasing Ni content, and reaches a maximum in the $x = 0.44$ sample, then decreases a little in the $x = 0.45$ sample. The large value of $\gamma_0$ (77.5 mJ/mol K$^2$ for $x = 0$) and 310.1 mJ/mol K$^2$ for $x = 0.44$) originates from the heavy fermion behavior of the 4f electrons in this system, as found in most Ce and Yb compounds [1, 32]. The change in the $\beta$ value is not so large, without a systematic dependence on $x$, indicating that the partial substitution of Ni for Ti has little influence on its Debye temperature, for example, using the fitting $\beta$ values, and the Debye expression $\theta_D = [(12/5)N R \pi^4 / \beta]^{1/3}$, where $N$ is the number of atoms per unit cell, $R$ is the molar gas constant, the Debye temperature $\theta_D$ was calculated to be $\theta_D = 291$ and 281 K for $x = 0.0$ and 0.45 samples, respectively, which are comparable with the $\theta_D = 290$ K for the Ce-analog LaTiGe$_3$ without f-electrons reported by Manfrinetti et al [33]. In addition, the density of states at Fermi level, $N(E_F) = 3\gamma_0 / (\pi k_B)^2$, where $k_B$ is the Boltzmann constant, was estimated to be 33, 131 and 125 ev$^{-1}$ for $x = 0.0, 0.44$ and 0.45, respectively, by using the fitting values of $\gamma_0$, which indicates that the partial substitution of Ni$^{2+}$(3d$^2$) for Ti$^{3+}$(3d$^3$) results in the increase of carrier concentration, as discussed above.

### 3.5. Phase diagram and discussions

Based on the results of $\rho(T)$, $M(H)$ and $C(T)$ measurements discussed above, we constructed a phase diagram of temperature as a function of Ni content, $x$, for the CeTi$_{1-x}$Ni$_x$Ge$_3$ system, as shown in figure 6(a). $T_C$ decreases with increasing Ni content, and reaches zero kelvin at a critical content $x_{cr} = 0.44$. Figures 6(b)–(e) present the Ni content $x$ dependencies of $A$($A''$), $n$, and $\rho_0$ values, which are the fitting parameters to the $\rho(T)$ data at lower temperatures by equation (2) or a power-law, as well as the electronic specific heat coefficient $\gamma_0$ value, estimated from the fitting to the $C(T)$ data at lower temperatures by equation (3) or equation (4). It is obvious that all four parameters exhibit anomalies near $x_{cr} = 0.44$. For example, the power exponent $n = 2$ in the $0 \leq x \leq 0.40$ samples, indicating that the Fermi–liquid behavior occurs in this region, whereas for the $x = 0.44$ and 0.45

| $x$ | $\gamma_0$ | $\beta$ | $\alpha$ | $\Delta$ |
|-----|------------|---------|----------|---------|
| 0.0 | 77.5       | 0.394   | 0.170    | 29      |
| 0.1 | 115.1      | 0.421   | 0.137    | 24      |
| 0.2 | 169.2      | 0.441   | 0.115    | 17      |
| 0.25| 194.5      | 0.439   | 0.104    | 15      |
| 0.3 | 232.1      | 0.432   | 0.090    | 13      |

| $x$ | $\gamma_0$ | $\beta$ | $\delta$ | $T_0$ |
|-----|------------|---------|----------|------|
| 0.44| 310.1      | 0.473   | 0.066    | 9    |
| 0.45| 298.0      | 0.452   | 0.034    | 8    |

Table 2. The fitting parameters $\gamma_0$(mJ/mol K$^2$), $\beta$(mJ/mol K) and $T_0$(K) by equation (4) to the $C(T)$ data in the low-temperature range for $x = 0.44$ and 0.45 samples.
samples, their \( n \) value is less than 2.0, and reaches a minimum value of 1.6 in the \( x_{cr} = 0.44 \) sample, signaling that nFL behavior emerges in this sample (see figure 6(c)). In addition, all the \( A, \rho_0 \) and \( g_0 \) values reach maximum at the \( x_{cr} \), as shown in figures 6(b), (d) and (e), respectively. All these anomalies in \( n, A, \rho_0 \) and \( g_0 \) at \( x_{cr} \), in addition to \( C/T \sim \log(T_0/T) \) behavior near \( x_{cr} \), indicate that strong spin-fluctuation emerges in the samples near \( x_{cr} \).

The Hertz–Millis–Moriya model \([34–36]\) of quantum criticality predicts for the dependence of \( T_C \) on the control parameter tuning the QCP, in our case the Ni content \( x \), \( T_C(x) \sim |x - x_c|^\mu \), a critical exponent \( \mu = 3/4 \) and \( \mu = 1 \) for a ferromagnetic QCP in three dimensions (3D) and two dimensions, respectively. Both \( T_C(x) \) dependencies, \( \mu = 3/4 \) and \( \mu = 1 \), with \( x_c = 0.44 \) and 0.46 are shown in figure 6. It is clear that the \( T_C(x) \) dependence with \( \mu = 3/4 \) could better describe the \( T_C \) values obtained from the experiments, indicating that the 3D ferromagnetic QCP occurs in the \( \text{CeTi}_{1-x}\text{Ni}_x\text{Ge}_3 \) system. Similar scaling laws were also found in other weak itinerant ferromagnetic systems near the critical point, for example, the Nb-substituted system \( \text{Zr}_{1-x}\text{Nb}_x\text{Zn}_3 \) \([37]\), MnSi under high pressure for \( P \leq P^* = 12 \) kbar, \([38]\) and Ga-substituted \( \text{Ni}_x\text{Al}_{1-x}\text{Ga}_x \) \([18]\) system. Although no new collective phases, such as unconventional superconductivity near the QCP, have been observed up to now, we are looking forward to finding them after getting a high quality single crystal in the future, and studying the deep quantum-fluctuation behavior near the QCP.

![Figure 6](image_url)

**Figure 6.** Ni content \( x \) dependence of (a) \( T_C \) estimated from the \( \rho(T), M(H) \) and \( C(T) \) data for \( \text{CeTi}_{1-x}\text{Ni}_x\text{Ge}_3 (0.0 \leq x \leq 0.45) \). Red point: \( T_C^{43} \), determined by Arrott plots in \( M(H) \) measurements. Red line: \( T_C^{43} \sim |x - x_c|^{3/4} \) (b) the temperature coefficient, \( A \) or \( A' \), (c) the power-exponent, \( n \), (d) the residual resistivity, \( \rho_0 \), the fitting parameters to \( \rho(T) \), see the text. (e) The electronic specific heat coefficient, \( g_0 \), obtained from \( C(T) \) data, as discussed in the text.
4. Summary

In summary, we successfully synthesized ferromagnetic CeTi$_{1-x}$Ni$_x$Ge$_3$ (0 $\leq x \leq 0.45$) heavy-fermion alloys. Based on the measurements of magnetization, resistivity and specific heat, the magnetic phase diagram was constructed for this system. It was found that $T_C$ decreases with increasing Ni content, $x$, and reaches zero kelvin near $x_c = 0.44$. nFL behavior in $\rho(T)$, and the $C/T \propto \log(T_0/T)$ relationship in the samples near $x_c$ indicate that a ferromagnetic QPT occurs at $x_c$. These results imply that CeTi$_{1-x}$Ni$_x$Ge$_3$ may provide another platform to search for the new exotic collective phases, such as unconventional superconductivity, near the QCP.

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