Cu-doping effects on the ferromagnetic semimetal CeAuGe

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\textbf{A B S T R A C T}

Keywords:
Ce-based intermetallic compounds, in which localized moments of Ce\textsuperscript{3+} ion form Kondo lattice, exhibit exotic quantum phenomena such as heavy-fermion superconductivity \cite{1–3}, topological magnetism \cite{4}, intriguing magnetic ordering \cite{5,6}, and quantum phase transition \cite{7,8}. The ground states of the Ce-based metals are controlled by competing interactions between the spin anisotropy, multiple exchange interactions, and Kondo hybridization. The Kondo hybridization between localized Ce moment and itinerant spins can be tuned by non-thermal parameters such as pressure, magnetic field, and chemical doping. Chemical doping modifies the density of states at the Fermi level, thereby exchange interactions between localized and itinerant spin change the ground state of the system as described in the Doniach phase diagram \cite{9}.

Table 1 summarizes Ce-based hexagonal compounds of Ce\textit{MX} (\textit{M} = Cu, Ag, Au, and \textit{X} = Si, Ge, Sn). All systems exhibit magnetically ordered states. Fig. 1 shows a linear relation between transition temperatures of magnetic ordering (\textit{T}_C, \textit{T}_N) and an anisotropic parameter \( a = d_\parallel/d_\perp \), when \( d_\parallel \) and \( d_\perp \) are Ce-Ce distance along the crystallographic \( c \)- and \( c \)-axis, respectively. When the magnetic exchange interactions between Ce-spins are anisotropic, \( 0.80 < a < 0.875 \), the magnetic ground state is antiferromagnetic (AFM) one, while isotropic interactions, \( 0.875 < a < 0.95 \), favour a ferromagnetic (FM) ordering. As shown in Table 1, all Ce\textit{MX} compounds have similar crystalline-electric-field ground states (GS\textit{CEF}) of mainly \( |\pm 1/2 \rangle \) with mixing of \( |\pm 5/2 \rangle \) doublet.

An unusual non-collinear FM structure has been reported in CeAlSi, presumably due to the non-centrosymmetric crystal structure \cite{20}. In our case, both non-centrosymmetric end member CeAuGe and the centrosymmetric one CeCuGe exhibit ferromagnetism with transition temperature near 10 K. This is in accordance with a previous neutron powder diffraction study that has shown that the magnetic ordering in CeAuGe is a collinear FM, and we note that the magnetic structure of CeCuGe has not been reported \cite{21}. Cu-doping of the non-
centrosymmetric CeAuGe is the way of investigating the relationship between the magnetic ordering, crystal symmetry, and the bulk physical properties, while the Curie temperature remains similar. Herein, we report the crystal and magnetic properties of the polycrystalline series of CeAu₁₋ₓCuₓGe using x-ray and neutron diffraction and the macroscopic physical properties such as electrical resistivity and magnetization as a function of temperature and magnetic field.

2. Material and methods

Polycrystalline samples of CeAu₁₋ₓCuₓGe were synthesized by the arc-melting technique, and then the arc-melted samples were sealed in the evacuated silica tubes with a Ta-foil, then the tubes were quenched in the cold water after annealed at 800 °C for 10 days to improve the homogeneity of the samples. Phase purity and crystal structures of CeAu₁₋ₓCuₓGe were checked by powder x-ray diffraction measurements (PXRD) using a Bruker D8 Advance with Cu-cathode. Chemical composition was confirmed by x-ray micro-fluorescence (XRF) analysis using an Orbis micro-XRF analyzer from EDAX. The crystal and magnetic structures were studied by neutron powder diffraction using the high-resolution powder diffractometer for thermal neutrons (HRPT) [22] at the Swiss Spallation Neutron Source SINQ at Paul Scherrer Institut (PSI), Switzerland. About 2.5 g of each powder was loaded in 6 mm vanadium containers. Diffraction patterns were collected at temperatures of 1.5 and 15 K using neutrons with wavelengths of 1.494 and 2.45 Å. All diffraction data were analyzed using the programs of the FullProf software suite [23]. The symmetry analysis of the magnetic structures was done using the Bilbao crystallographic server [24] and the ISODISTORT tool based on ISOTROPY software [25,26]. Electrical resistivity measurements were performed using the standard four-probe (25 μm Pt wires) technique applying a current of 1 mA on the polished surface of bar-shaped specimens. A physical property measurement system (PPMS-9, Quantum Design) was used for applying magnetic fields up to 90 kOe and controlling the temperature in the range from 1.8 to 300 K. A 3He-pumping system with an Oxford vertical magnet cryostat was adopted for the resistivity measurements in the temperature range from 0.28 to 2 K. The magnetization measurements were performed on a superconducting quantum interference device (SQUID) installed in the magnetic property measurement system (MPMS-7, Quantum Design), in the temperature and magnetic field ranges from 1.8 to 300 K and 0 to 70 kOe, respectively.

3. Results

3.1. Crystal structure of CeAu₁₋ₓCuₓGe

Fig. 2(a) shows crystal structures of CeAuGe and CeCuGe and representative PXRD patterns for CeAu₁₋ₓCuₓGe (x = 0.0, 0.5, 1.0). Both end-member compounds adopt hexagonal structure, wherein the two different hexagonal layers of Ce – Ce and M – Ge (M = Cu and Au) stack alternately along the crystallographic c-axis. While the puckered hexagonal layer of Au – Ge breaks the centrosymmetric symmetry (P6₃mc, no. 186), the flat Cu – Ge layer conserves the centrosymmetric symmetry (P6₃/mmc, no. 194) of the hexagonal structure [12,27]. Fig. 2 (b) and (c) show lattice parameters of CeAu₁₋ₓCuₓGe determined from so-called Le Bail fits of the x-ray diffraction patterns using the P6₃mc and P6₃/mmc space groups for 0 ≤ x ≤ 0.5 and 0.5 < x < 1.0 compounds, respectively. In the Le Bail fit, only the crystal metrics and resolution parameters are refined, whereas the integrated Bragg peak intensities are refined independently without any structure model. The crystal structures were determined from the neutron diffraction data (see below). Cu-doping (x) linearly shortens the a-lattice parameter as x increases, decreasing about 3 % at x = 1. On the other hand, the c-lattice parameter exhibits two different linear regimes with an abrupt change near x = 0.5, which can be interpreted by the different crystal symmetry. The unit cell volume linearly decreases as x increases because the a-lattice parameter change is dominant.

Table 1

<table>
<thead>
<tr>
<th>M</th>
<th>Space group</th>
<th>Type</th>
<th>T_C, T_N (K)</th>
<th>GSₐₓᵧ₂</th>
<th>α (Å)</th>
<th>ε (Å)</th>
<th>dₐ/dₗ</th>
<th>Ref.</th>
</tr>
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<tr>
<td>CeMSi</td>
<td>Cu</td>
<td>P6₃/mmc</td>
<td>FM</td>
<td>15.5</td>
<td>0.87ₚ̅₋₋₀.₄₈q</td>
<td>4.22ₚ̅</td>
<td>7.98ₚ̅</td>
<td>0.94₀</td>
</tr>
<tr>
<td>CeMG Ge</td>
<td>Cu</td>
<td>P6₃/mmc</td>
<td>FM</td>
<td>10.2</td>
<td>0.₉ₚ̅₋₀.₀₃₁q</td>
<td>4.₃₁₁</td>
<td>7.₉₃₃</td>
<td>0.₉₂₀</td>
</tr>
<tr>
<td>Ag</td>
<td>P6₃/mmc</td>
<td>AFM</td>
<td>4.₈</td>
<td>--</td>
<td>4.₅₄₄</td>
<td>7.₁₁₁</td>
<td>0.₈₅₀</td>
<td>[14,15]</td>
</tr>
<tr>
<td>Au</td>
<td>P6₃/mmc</td>
<td>FM</td>
<td>6.₀</td>
<td>--</td>
<td>4.₅₃₆</td>
<td>7.₇₄₆</td>
<td>0.₈₅₀</td>
<td>[16]</td>
</tr>
<tr>
<td>CeMSn</td>
<td>Cu</td>
<td>P6₃/mmc</td>
<td>FM</td>
<td>9.₇</td>
<td>--</td>
<td>4.₄₆₀</td>
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<td>0.₇₉₀</td>
</tr>
<tr>
<td>Ag</td>
<td>P6₃/mmc</td>
<td>AFM</td>
<td>4.₅₈₃</td>
<td>--</td>
<td>4.₇₇₄</td>
<td>7.₇₄₇</td>
<td>0.₈₁₀</td>
<td>[17,18]</td>
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<tr>
<td>Au</td>
<td>P6₃/mmc</td>
<td>AFM</td>
<td>4.₁</td>
<td>--</td>
<td>₄.₇₂</td>
<td>₇.₇₀</td>
<td>₀.₈₁₅</td>
<td>[17,18]</td>
</tr>
</tbody>
</table>

Tₐ, Tₐ: magnetic ordering temperature, dᵢ (dₗ): distance between Ce atoms along the c-axis (a-axis) direction. GSₐₓᵧ₂: crystalline-electric-field ground state with p = 1/2 and q = 5/2 ×. Mantid software [19] was used for simulating GSₐₓᵧ₂ with the Rₐₓ crystal field parameters given in the references.

3.2. Magnetic properties of CeAu₁₋ₓCuₓGe

Magnetic susceptibility (M/H) of CeAu₁₋ₓCuₓGe is plotted as a function of temperature in Fig. 3(a), and all compounds show FM-like phase transition. Fig. 3(b) shows that the magnetic susceptibility in all samples well follows the Curie-Weiss behaviour at temperatures above 100 K. From the least-squares fits using an equation of $M/H = M/\theta + C/(T - \theta)$ (M/H(0): temperature-independent term, C: Curie-constant,
Fig. 2. (a) Schematic drawing of the centrosymmetric CeAuGe (P63/mmc) and non-centrosymmetric CeCuGe (P63/mc) crystal structure (upper panel). Powder x-ray diffraction patterns of CeAu$_{1-x}$Cu$_x$Ge ($x$ = 0, 0.5, 1.0). Black dot, red line, blue and green bar represent observed data, fit result using Le Bail method (explained in the text), nuclear diffraction positions of P63/mmc and P63/mc, respectively. (b, c) $a$- and $c$-lattice parameters as a function of $x$ (d) The unit cell volume as a function of $x$. The error bars are smaller than the symbol size.

Fig. 3. (a) Magnetic susceptibility ($M/H$) of CeAu$_{1-x}$Cu$_x$Ge as a function of temperature, the inset shows the data at low temperatures. (b) $H/M$ of CeAu$_{1-x}$Cu$_x$Ge is plotted as a function of temperature, and the inset shows each effective magnetic moment deduced from the Curie-Weiss fit at temperatures above 200 K. (c) Magnetization as a function of field of CeAu$_{1-x}$Cu$_x$Ge at 2 and 20 K. (d) Curie temperature ($T_C$, square symbol) and the room temperature $c$-lattice parameter (circles) as a function of $x$ on the left and right ordinate, respectively. Inset shows the magnetization derivative $\partial M/\partial T$ as a function of temperature, which has a minimum at $T_C$. 
3.3. Neutron powder diffraction results of \( \text{CeAu}_1\text{Cu}_3\text{Ge} \)

Magnetic and crystal structures of \( \text{CeAu}_1\text{Cu}_3\text{Ge} \) (where \( x = 0.0, 0.2, 0.5, 0.8, 1 \)) were studied using neutron powder diffraction. Table 3 summarizes the results of the Rietveld refinements of the crystal structures from the neutron diffraction patterns measured with neutron wavelength \( \lambda = 1.494 \) Å at 15 K. The best fits were obtained in the space groups \( \text{P6}_3\text{mmc} \) and \( \text{P6}_3\text{mmc} \) for \( 0 \leq x < 0.5 \) and \( 0.5 \leq x < 1.0 \), respectively (for details see Fig. S1 in Supplemental Material), in accordance with x-ray diffraction data shown above. For the magnetic structures, the longer wavelength, \( \lambda = 2.54 \) Å, was used to obtain higher resolution for the Bragg peaks at low scattering angles \( 2\theta \). Fig. 4(a) and (b) shows representative diffraction patterns of \( \text{CeAuGe} \) and \( \text{CeCuGe} \), respectively. As shown in each upper panel of Fig. 4(a) and (b), the Bragg peak positions at the temperatures 15 and 15 K are practically identical, while the Bragg peak intensities at 1.5 K are enhanced. The difference patterns (1.5 – 15 K) for \( \text{CeAuGe} \) and \( \text{CeCuGe} \) are shown in the lower panel of Fig. S5 (a) and (b). Le Bail fits of the difference patterns of \( \text{CeAu}_1\text{Cu}_3\text{Ge} \) show that the propagation vector \( k \) of all compounds is zero, \( k = 0 \), with the goodness of fit \( \chi^2 \approx 1 \) (see details Fig. S2 of SM).

3.4. Magnetic structure of \( \text{CeAu}_1\text{Cu}_3\text{Ge} \)

For \( \text{P6}_3\text{mmc} \) (\( 0 \leq x < 0.5 \)) and the propagation vector \( k = 0 \), there are four irreducible representations (irreps) for non-zero Ce moments allowed by symmetry. Assuming that the only one irrep is active at the magnetic transition, eight possible magnetic Shubnikov subgroups (labeled as 1, 2, 4–7, 12, and 13 in Fig. S3 (a)) are allowed, and four of them (labeled as 1, 2, 5, and 7) have maximal symmetry. For \( \text{P6}_3\text{mmc} \) (\( 0.5 < x < 1.0 \)) with \( k = 0 \), four irreps and eight possible magnetic subgroups (labeled as 1, 2, 4, 7, 12, and 13 in Fig. S3 (b)) are allowed and four of them (labelled as 1, 2, 5, and 7) have maximal symmetry. All Shubnikov magnetic space groups (MSG) were sorted to fit the difference patterns, starting from maximal symmetry subgroups. The best fit results were obtained in the orthorhombic \( \text{Cmc}_{21} ' \) (no. 36.175, labeled as 5 in Fig. S3 (a)) and monoclinic \( \text{P2}_1/\text{m}^* \) (no. 11.54, labeled as 12 in Fig. S3 (b)) MSG for \( \text{CeAuGe} \) and \( \text{CeCuGe} \) with the goodness of fit \( \chi^2 \approx 1 \) (see the details in Fig. S4 of SM). The unit cell metrics and atomic coordinates in the fits with the magnetic space groups were fixed by the values refined from the patterns in paramagnetic state at 15 K. We note that the goodness of fit \( \chi^2 \) is the same as for the Le Bail fit, implying that there is no room for further improvement because Le Bail fit is model-independent.

The schematic drawing of the magnetic structures of \( \text{CeAuGe} \) and \( \text{CeCuGe} \) is shown in Fig. 4 (c) and (d), respectively. The unit cell transformations from parent paramagnetic space groups are given by the following relations \( A - b, B = 2a + b, \) and \( C - c \) in \( \text{Cmc}_{21} ' \) and \( A = a, B = c, \) and \( C = -b \) in \( \text{P2}_1/\text{m}^* \), where the capital letter and lower case are the basis vectors of the magnetic and the parent paramagnetic space group, respectively. Structure parameters obtained from the fits of neutron diffraction patterns are summarised in Tables S2 and S3. In \( \text{Cmc}_{21} ' \), the Wyckoﬀ positions for Ce is 4a (0, 0, 0) with magnetic moments allowed only along the crystallographic \( c \)-axis. Thus, the magnetic symmetries impose strong constraints on the possible magnetic configurations. On the other hand, the magnetic moments of Ce in Wyckoﬀ position 2a (0, 0, 0) in \( \text{P2}_1/\text{m}^* \) are allowed along the arbitrary direction. The best result of Rietveld refinement, however, was obtained for the moment directed along the \( a \)-axis. For both MSGs, the collinear ferromagnetic structure is fixed by symmetry. The refined values of ordered magnetic moments amounted to 1.05(2) \( \mu_B \) for \( \text{CeAuGe} \) and 1.11 (3) \( \mu_B \) for \( \text{CeCuGe} \). They are consistent with the saturated moments in Fig. 3(c) and the previous neutron experiment result of \( \text{CeAuGe} \) [21]. Fig. 4(e) shows the ordered magnetic moments (\( \mu_{\text{EFG}} \)) as a function of \( x \) deduced from the fits of the neutron diffraction patterns (details are given in Table S2 in SM). The samples of \( \text{CeAu}_1\text{Cu}_3\text{Ge} \) with non-centrosymmetric MSG, \( 0 < x < 0.5 \), exhibit the orthorhombic \( \text{Cmc}_{21} ' \) FM structure, while the compositions with centrosymmetric MSG, \( 0.5 < x < 1.0 \), show that the propagation vector \( k \) of all compounds is zero, \( k = 0 \), with the goodness of fit \( \chi^2 \approx 1 \) (see details Fig. S2 of SM).

Table 3 Crystal structure parameters refined from the neutron powder diffraction patterns of \( \text{CeAu}_1\text{Cu}_3\text{Ge} \) measured with neutron wavelength \( \lambda = 1.494 \) Å at 15 K.

<table>
<thead>
<tr>
<th>( x )</th>
<th>Space group</th>
<th>( a ) (Å)</th>
<th>( c ) (Å)</th>
<th>( M ) (W, 1/3, 2/3, z)</th>
<th>( \text{Ge} ) (W, 1/3, 2/3, z)</th>
<th>( R_{\text{wp}} )</th>
<th>( R_{\text{exp}} )</th>
<th>( x^2 )</th>
</tr>
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<tbody>
<tr>
<td>0.0</td>
<td>( \text{P6}_3\text{mmc} )</td>
<td>4.4570</td>
<td>7.8517</td>
<td>2b, 0.7640</td>
<td>2b, 0.2179</td>
<td>16.9</td>
<td>4.61</td>
<td>0.01</td>
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<tr>
<td>0.2</td>
<td>( \text{P6}_3\text{mmc} )</td>
<td>4.4564</td>
<td>7.8481</td>
<td>2b, 0.7846</td>
<td>2b, 0.2415</td>
<td>30.5</td>
<td>16.4</td>
<td>0.01</td>
</tr>
<tr>
<td>0.5</td>
<td>( \text{P6}_3/ \text{mmc} )</td>
<td>4.3931</td>
<td>7.8784</td>
<td>2c, 0.75</td>
<td>2c, 0.25</td>
<td>33.0</td>
<td>4.61</td>
<td>0.01</td>
</tr>
<tr>
<td>0.8</td>
<td>( \text{P6}_3/ \text{mmc} )</td>
<td>4.3420</td>
<td>7.9252</td>
<td>2c, 0.75</td>
<td>2c, 0.25</td>
<td>46.6</td>
<td>27.0</td>
<td>0.01</td>
</tr>
<tr>
<td>1.0</td>
<td>( \text{P6}_3/ \text{mmc} )</td>
<td>4.2859</td>
<td>7.9285</td>
<td>2c, 0.75</td>
<td>2c, 0.25</td>
<td>50.7</td>
<td>27.0</td>
<td>0.01</td>
</tr>
</tbody>
</table>

The goodness of fit \( x^2 \) is defined as \( (\text{R}_{\text{wp}}/\text{R}_{\text{exp}})^2 \), \( \text{R}_{\text{wp}} \) and \( \text{R}_{\text{exp}} \) represent a reduced chi-square, weighted profile factor, and expected weighted profile factor, respectively [23]. The atomic coordinate of Ce 2a (0, 0, z) in \( \text{P6}_3\text{mmc} \) is fixed to zero for maintaining crystal symmetry. W represents Wyckoﬀ position in relevant crystal symmetry.
3.5. Electrical resistivity of CeAu$_{1-x}$Cu$_x$Ge

Fig. 5(a) shows the zero-field electrical resistivity as a function of temperature ($\rho(T)$) of CeAu$_{1-x}$Cu$_x$Ge with showing high resistivity values and metallic behaviour, which are consistent with the previous report [30]. The residual-resistance ratio (RRR) coefficients of CeAuGe and CeCuGe are of range from 1.2 to 10.6 (details are given in Table S4 in SM). The transition temperatures of CeAuGe (3.968(1) Å) and CeCuGe (3.967(1) Å) are similar. Band structures are similar but have different crystallographic symmetries.

$x \leq 1.0$, adopt P2$_1$/m’ monoclinic symmetry. We note that the FM structures are similar but have different crystallographic symmetries.

Fig. 4. Neutron powder diffraction data collected at 15 and 1.5 K (upper panel) and their difference pattern (lower panel) of CeAuGe (a) and CeCuGe (b) are representatively displayed. The red line in the lower panel represents the best fit results using the Shubnikov magnetic space group Cmc $2'_1$ for CeAuGe and P2$_1$/m’ for CeCuGe. The schematic magnetic structures of CeAuGe and CeCuGe are displayed in (c) and (d), respectively. (e) The sizes of ordered magnetic moments obtained from the fits on the difference (1.5 - 15 K) pattern are plotted as a function of x.

Distances between the hexagonal Ce-layers ($d_c$, as defined in Fig. 1) of CeAuGe (3.968(1) Å) and CeCuGe (3.967(1) Å) are similar. Band structures of ScAuGe, CeAuGe, and LuAuGe calculated using TB-LMTO-ASA (Tight-Binding, Linear Muffin-Tin Orbital, Atomic-Sphere Approximation) program show that the larger spacing $d_c$ leads to the smaller inter-layer interaction [27]. As shown in Fig. 3(d), the maximum $d_c$ is observed at $x \sim 0.5$, where the Curie temperature $T_C$ reaches the maximum value of 12 K. The higher $T_C$ = 15.5 K in CeCuSi [10] can be interpreted by the larger $d_c$ = 3.994 Å than the one in CeAuGe and CeCuGe. Similar behaviour was reported in CeCuGe$_{1-x}$Sn$_x$ that the $T_C$ is proportional to the interlayer distance. On the other hand, $T_C$ is inverse proportional to the interlayer distance in CeAu$_{1-x}$Al$_x$Ge, in which Al-doping was expected to screen Ce moments with a higher electron density. $T_C$ in CeAu$_{1-x}$Cu$_x$Ge may be correlated with not only the interlayer interaction but also conduction electron density by the chemical doping.

The similar anisotropic parameters $a = d_c/d_x$ of CeAu$_{1-x}$Cu$_x$Ge are responsible for the similar magnetic properties and collinear ferromagnetic structure. While the magnetic properties of CeAu$_{1-x}$Cu$_x$Ge compounds are similar, the electrical resistivity exhibits different behaviours depending on doping concentration $x$. CeCuGe exhibits the local
minimum of $\rho(T)$ at 17.5 K due to the Kondo scattering and $T$-linear behaviour in the FM state. On the other hand, $\rho(T) \sim T^3$ behaviour in the FM state was observed in CeAuGe without the local minimum of $\rho(T)$. Note that the fit by a gap function, $\rho(T) \sim T^2\exp(-\Delta/k_B T)$, does not work for both cases. Since the temperature exponent of resistivity in CeCuGe and CeAuGe polycrystals are different from the one expected in the ferromagnetic metals [32,33], $\rho(T)$ measurements on single crystals and at lower temperatures might help to understand the scattering mechanism in the FM state. As shown in Fig. 5(b), the small Cu-doping in CeAuGe suppresses FM transition temperature to 8 K, while other doping cases show $T_C$ higher than $x = 0.0$ and 1.0. Further experiments with $x = 0.2$ concentration and other small doping contents can be useful to elucidate the origin of the broad transition at 12.5 K and the low-temperature transition-like feature at 2.5 K that are seen only in the electrical resistivity.

5. Conclusion

Cu-doping effects in the hexagonal ferromagnet CeAuGe have been studied on polycrystalline series of CeAu$_{1-x}$Cu$_x$Ge ($x = 0.0, 0.2, 0.4, 0.5, 0.6, 0.8, 1.0$) using x-ray and neutron (except $x = 0.4$ and 0.6) diffraction, electrical resistivity and magnetization as a function of temperature and magnetic field. The doping changes the crystal structure from the non-centrosymmetric ($P6_3/mc$) to the centrosymmetric ($P6_3/mmc$) space group at the critical concentration $x_C \sim 0.5$. The magnetic moments of Ce are ferromagnetically ordered in the Shubnikov magnetic space groups $Cmc_21$ and $P2_1/m$ below and above $x_C$, respectively. The Curie temperature $T_C$ depends on concentration and reaches the maximum value $T_C = 12$ K at $x = x_C$, where the distance between Ce-layer shows the maximal value. The magnetic moments are oriented perpendicular to the hexagonal c-axis with the sizes slightly dependent on concentration amounting to 0.95(2) - 1.16(1) $\mu_B$ at 1.5 K. The electrical resistivity exhibits metallic temperature dependence and a pronounced sharp change at the ferromagnetic transition for all concentrations except for $x = 0.2$. The resistivity $\rho(T)$ has a local minimum in the paramagnetic state due to Kondo effects at high doping levels $x > 0.5$. The sample with the small doping $x = 0.2$ shows different from other concentrations behaviour of electrical resistivity. The transition seen by resistivity is broadened and shifted to 12.5 K, which is higher than the bulk $T_C$ and in addition, there is a transition-like feature in the ferromagnetic state at 2.5 K. Further experiments with the low doping content samples can be useful to elucidate the origin of the above peculiarities.

The data that support this study are available via the Zenodo repository [34].

CRedit authorship contribution statement

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jmmm.2022.169147.

References