



Revisiting the physical properties of $\text{Ce}_2\text{Ru}_3\text{Ga}_9$: Intermediate valence, or Kondo lattice system?

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ABSTRACT

In this paper we discuss in details the experimental results regarding to the crystal structure, magnetic, electronic and thermal transport properties of the strongly correlated compound $\text{Ce}_2\text{Ru}_3\text{Ga}_9$. Based on these data we report the observation of coexistence and competition between intermediate valence state on Ce ions and Kondo-like behaviour in a high-quality polycrystalline alloy. Intermediate valence behaviour manifests itself in the temperature dependence of the magnetic susceptibility (χ), which reveals a broad maximum at around 335 K as is frequently observed for Ce compounds with mixed-valent $\text{Ce}^{3+/4+}$. The electrical resistivity (ρ) behaves in a manner characteristic of systems with strong electronic correlations. First varies in a metallic manner, typical for intermediate valence compounds and then demonstrating a behaviour pointing on the onset of coherence that develops above the room temperature. The thermoelectric power (S) shows a broad maximum with a relatively large value of $S_{max} = 35.7 \mu\text{V/K}$, which is typical for the Ce-based systems that exhibit a strong $c-f$ hybridization. Both $\rho(T)$ and $S(T)$ show the $-\ln T$ behaviour at high temperatures thereby indicating on the presence of incoherent electron scattering on a local moment of Ce^{3+} . Moreover, a particular attention has been paid to the low- T behaviour in the investigated physical quantities, as it is one of the focuses of this paper.

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1. Introduction

Intermetallic Ce-based compounds being on the border between localized $4f^1$ and non localized $4f^0$ state have attracted vast attention in recent years. As it is universally recognized, the low- T properties of most metallic systems classified as the Fermi liquids are qualitatively similar, i.e., show the linear temperature dependence of the electronic specific heat, the quadratic temperature dependence of the electrical resistivity, and frequently reveal a weakly temperature-dependent Pauli susceptibility. In the case of Ce compounds where the presence of strong hybridization between the $4f$ -electron and the conduction electrons ($c-f$) can lead to the nonmagnetic intermediate valence (IV) state of $\text{Ce}^{3+/4+}$, or to formation of the heavy fermion Kondo lattice ground state with a localized character of Ce^{3+} ($J = 5/2$) ions, the physical quantities are strongly modified. In particular the occurrence of a broad maximum in $\chi(T)$ represents a hallmark of the IV behaviour on Ce

ions. The hybridization due to the overlap of the Ce $4f$ -electron wave function with the conduction electron wave functions causes the delocalization (fluctuation $4f^1 \rightarrow 4f^0$) of the f -electron for some period of time, and consequently disappearance of the Ce local moment [1]. On the other hand, there is another alternative for isolated Ce^{3+} magnetic impurities [2] that for temperature less than the Kondo temperature T_K , predicts also a strong $c-f$ coupling, where the local $4f$ -spin is compensated by the conduction electrons (Kondo effect) and the local moment is quenched. In general, this $4f$ -electron duality in Ce compounds is primarily considered as a basic ingredient of quantum critical behaviours at very low temperatures such as unconventional (magnetically driven) superconductivity or non-Fermi liquid behaviour, but also at higher temperatures may be responsible for competition or an interplay of the localized and itinerant degrees of freedom of the $4f$ -electrons. This issue seems to be particularly interesting because the Ce $4f$ shell contains just one electron, but the correlation effects among a lattice of Ce atoms in effect pose a many body problem that is notoriously difficult to solve.

$\text{Ce}_2\text{T}_3\text{X}_9$ compounds, where T stands for a transition metal Rh, Ru, Ir, whereas X is Al and Ga have the orthorhombic $\text{Y}_2\text{Co}_3\text{Ga}_9$ -

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structure with space group $Cmcm$ [3–5]. In this structure Ce atoms occupy only one crystallographic site at 8g. In the case of Ru the atoms are located in positions 8e and 4a, while for Ga the atoms are located in four crystallographic positions, namely 16h, 8g, 8f and 4c [4]. These compounds attract an interest due to the variety of remarkable physical properties, i.e., coexistence and competition between IV behaviour and Kondo interaction [3,6,7], as well as a deviation from the Fermi liquid behaviour at low temperatures observed in the case of $Ce_2Rh_3Al_9$ [3,8].

The titled compound of $Ce_2Ru_3Ga_9$ also appears to belong to the category of highly correlated materials, acting as a new example of competing energy scales. As far as we know, there are two reports on the physical properties of this compound [5,7], however, they are inconclusive as to the ground state of Ce ions. Previously the possibility of the coexistence of the IV state and formation of the Kondo lattice behaviour, two different with respect to the energy scales cooperative phenomena were already indicated in this compound [7]. Therefore, motivated by these considerations in this paper we seek to verify this scenario by studying the crystallographic, magnetic and transport properties. Furthermore, our earlier investigation on $Ce_2Ru_3Ga_9$ has indicated anomalous feature in the thermal conductivity measurements around $T^* \approx 203$ K that suggested the possible occurrence of the phase transition, probably of phononic or lattice origin [5]. Thus in this work we have also attempted to clarify this issue based on the low- T x-ray diffraction study. Moreover, to our knowledge no thermoelectric properties have been reported on this compound to date, and hence the motivation for this study.

2. Sample and experimental details

A polycrystalline sample was prepared by arc-melting stoichiometric amounts of the elements (purity in wt.%) Ce (99.99), Ru (99.99), and Ga (99.999) in ultra-high purity argon gas, followed by annealing at 600 °C for two weeks in an evacuated quartz ampoule. The x-ray diffraction (XRD) data, as well as the x-ray energy dispersive microanalysis (EDX) proved the sample to be single-phase. The crystal structure was established to be that of the expected orthorhombic $Y_2Co_3Ga_9$ -type with space group $Cmcm$. No parasitic or unreacted phases were detected from the EDX analysis taken at room temperature, obtaining a nominal stoichiometric ratio of the elements composition, i.e., Ce: 13.5 at.%, Ru: 21.9 at.% and Ga: 64.6 at.% [5]. In addition, the low- T XRD measurements on the powdered

specimen were carried out. All physical properties presented in this paper were measured on a physical property measurement system (PPMS) from *Quantum Design*, San Diego. Thermoelectric power was measured in the T range between 2 – 350 K in zero magnetic field followed by measurements in a static field of 9 T parallel to the heat current. The experiment was performed on slow cooling continuous mode at the rate of 12 K/h. The heat capacity was measured using a standard adiabatic heat-pulse method, employing the relaxation technique. Electrical resistivity measurements were carried out using a conventional dc four-probe technique with gold conducting wires spot welded onto the bar-shaped specimen with the same size as for thermal transport measurements. The temperature and magnetic field dependences of magnetization were measured using a VSM option on PPMS-9T apparatus equipped with an oven option enables high- T measurements up to 1000 K.

3. Results and discussion

3.1. Crystallographic properties

The low- T x-ray diffractometry was performed with a refurbished Siemens D500 system equipped with a 1D Mythen 1K detector and a closed cycle He refrigerator allowing to perform diffraction experiments down to 2.3 K. The sample was measured in the Bragg-Brentano geometry using $Cu K\alpha_{1,2}$ radiation ($Cu K\beta$ line was suppressed by the Ni filter). We performed the diffraction measurements for temperatures between 300 K and 2.3 K in a closed heating/cooling loop. No hysteresis in any parameters was observed, i.e., the diffraction patterns were not dependent on the temperature history.

The experimental data were refined using the MStruct software [9] (see Fig. 1). The initial guess for the structure was taken as a prototype structure $Dy_2Ru_3Ga_9$ (space group $Cmcm$, No. 63) from Ref. [4]. From the data fitting, we were able to refine the lattice parameters and atomic positions in the unit cell, while the resulted Goodness of Fit was 1.4506 and weighted R-factor was 0.0215. No indication of any structural transition or crystal lattice modifications was found for the investigated T range.

The refined atomic positions are summarized in Table 1, the orthorhombic unit cell of $Ce_2Ru_3Ga_9$ is visualized in Fig. 2. No temperature dependence of the atomic fractional coordinates was observed within the error. The lattice parameters of the orthorhombic unit cell at room temperature are $a = 12.9716(5)$ Å,

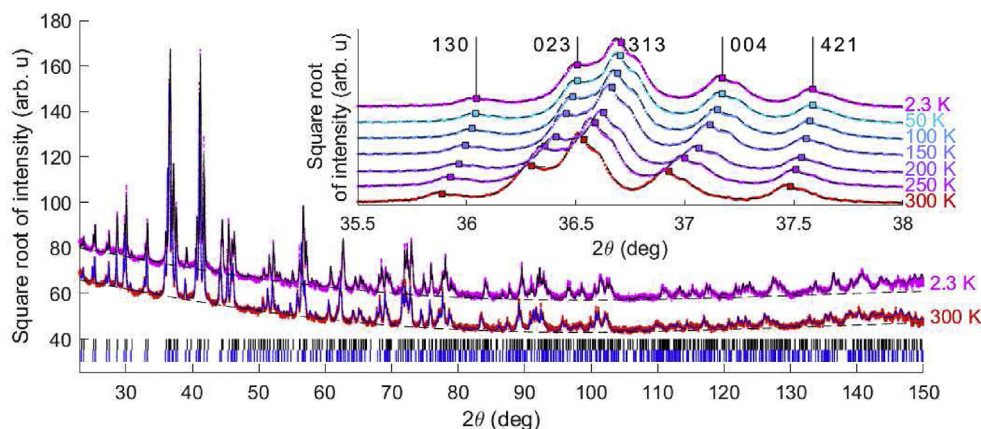


Fig. 1. XRD powder patterns of $Ce_2Ru_3Ga_9$ collected at room temperature (red curve) and at 2.3 K (magenta curve) fitted by the numerical simulation (blue and black lines, respectively). The peak positions are indicated by the vertical lines below the data. The diffraction patterns are vertically shifted for the clarity. The temperature evolution of powder pattern is depicted for several temperatures in the inset. The peak positions are indicated by squares marked by the vertical ticks with the diffraction indices hkl . (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

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