

Physical Properties Study of the CeOsGa₄ Compound

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A polycrystalline sample of CeOsGa₄ was prepared by arc-melting stoichiometric quantities of high purity elements of 4N and greater, in an arc furnace. The data was confirmed to crystallize in an orthorhombic structure, with a space group *Pmma* (Number 51). Measurements of the magnetic susceptibility showed a distinct anomaly around $T_o = 3.8$ K, while the high temperature data (above 100 K) obeys Curie-Weiss law. The calculated effective moment of $2.44 \mu_B$, was obtained, which was a bit less than the theoretical moment for a Ce³⁺ ion of $2.54 \mu_B$. Magnetization data at $T = 2$ K depicts that CeOsGa₄ does not saturate up to applied magnetic field of 7 T, while the data measured at 9 K indicates that the compound is purely paramagnetic at that temperature. The low temperature specific heat data is characterized by an anomaly at $T_o = 3.8$ K confirming the $\chi(T)$ data. Electrical resistivity shows a metallic behaviour at high temperatures with a deviation at low temperatures, signalling the cooperative behaviour which results to ordering. In this work we present the initial studies of the physical properties measurements performed on this compound.

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1. Introduction

The study of gallium (Ga) rich compounds has drawn some attention due to the complexities related to the crystal structures in these compounds. Amongst the first reported Ga-rich compounds are RE₂Co₃Ga₉ (RE: Y, Ho) [1], RE₂T₃Ga₉ (RE: Y, Sm, Gd-Yb; T: Rh, Ir) [2]. The structures of these members in these families of gallium-rich lanthanide (Yb₂Ru₃Ga₁₀ and Lu₂Ru₃Ga₁₀) vary from tetragonal structure, with space group *P4/mbm* where gallium atoms form a two dimensional infinite double layer that extends orthogonally to the entire axis of the structure [3]. Other stoichiometries such as LnRu₂Ga₈ (Ln = La – Nd) were found to crystallize in an orthorhombic CaCo₂Al₈-type structure, exhibiting the space group, *Pbam*, and also orthorhombic structures belonging to *Cmcm* space group [4].

Here we report on the physical properties of a polycrystalline CeOsGa₄ compound. This compound was confirmed to crystallize in an orthorhombic structure belonging to the space group *Pmma* [5]. The structure exhibits two types of two dimensionally alternating layers that are slightly puckered, having OsGa₃ layer sandwiched between CeGa layers.

2. Experimental procedure

Polycrystalline sample of the CeOsGa₄ compound was prepared by arc-melting high-purity elemental constituent elements on a water-cooled copper plate under ultra-high pure argon atmosphere. Metals of the following purity in wt% were used: Ce, 99.99%; Os, 99.95% and Ga, 99.999%. The button was turned over and remelted

several times to ensure good homogeneity of the sample. The calculated mass loss after final melting was less than 1%. The sample was characterised by powder x-ray diffraction (XRD) at room temperature using a Rigaku powder diffractometer with a CuK_α radiation ($\lambda = 1540698 \text{ \AA}$).

Magnetic susceptibility measurements of the polycrystalline sample was carried out using Quantum Design MPMS — a superconducting quantum interface device magnetometer, in the temperature range 380–2 K. Magnetization in applied fields up to 7 T was also measured in three different isotherms. Specific heat measurements were measured in the temperature range 275–2 K, using Physical Properties Measuring System (PPMS) from San Diego. Electrical resistivity was carried out using DC option in PPMS, in temperature ranges between 300 K and 2 K.

3. Results and discussion

3.1 Magnetic susceptibility

Figure 1 depicts the inverse magnetic susceptibility, $\chi^{-1}(T)$ data measured in the field of 0.05 T in temperature range of 2 K till 380 K. The high temperature data (> 100 K) obey Curie Weiss (CW) relationship following the relation:

$$\chi^{-1}(T) = \frac{3k_B(T - \theta_p)}{N_A \mu_{eff}^2}, \quad (1)$$

where k_B is the Boltzmann constant, θ_p is the paramagnetic Curie temperature, N_A is the Avogadro number and μ_{eff} , is the effective magnetic moment. The least square (LSQ) fit of $\chi^{-1}(T)$ data is shown in the main panel of the figure using a red line.

The fit yields the values of the effective magnetic moment, $\mu_{eff} = 2.44(2) \mu_B$ and $\theta_p = 6.5(2)$ K. The fitted value of μ_{eff} is close to the expected value of the free Ce³⁺ ion for $g[j(j+1)]^{1/2} = 2.54 \mu_B$, although the slight

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reduction in the effective moment can be ascribed to few possibilities which include the presence of the crystal electric field (CEF) effects.

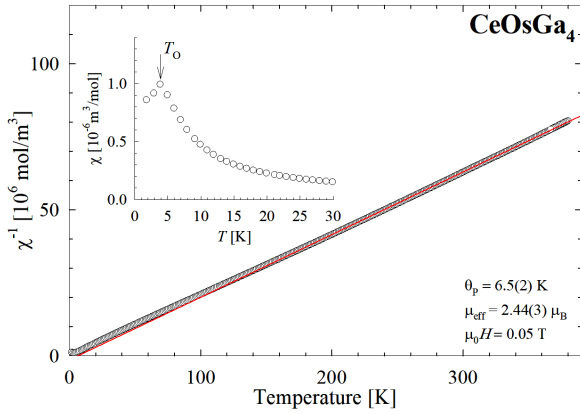


Fig. 1. The graph of the inverse magnetic susceptibility vs temperature, with the red solid line indicating a fit according to Eq. 1. The inset depicts low temperature region of the magnetic susceptibility where the arrow indicates the ordering temperature, $T_o = 3.8$ K.

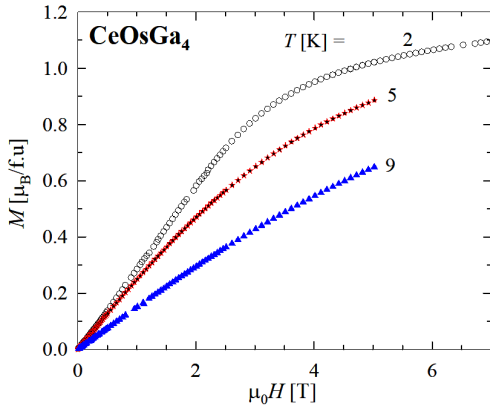


Fig. 2. The magnetization data measured deep within the ordered region, 2 K, above the ordering temperature, 5 K, and well within the paramagnetic region, 9 K.

The positive value of θ_p reveals the net ferromagnetic-like interactions within the paramagnetic region although the ground state of CeOsGa₄ at low temperatures exhibits antiferromagnetic-type of ordering. The anomaly at low temperatures of $\chi^{-1}(T)$ data (below 100 K), deviates from the CW behaviour, likely due to magnetocrystalline anisotropy or to thermal depopulation of CEF split of Ce $4f^1$ states. The inset on the top left corner of Fig. 1 depicts low temperature data with an arrow indicating an ordering temperature at $T_o = 3.8$ K.

Figure 2 depicts magnetization data, where it is observed that as from zero field up to about 2 T, the antiferromagnetic linearity is shown for isotherm measured at 2 K. This data confirms the type of ordering represented by the anomaly at 3.8 K. However, above this

field (2 T), a curvature is observed which turns to saturation at higher applied magnetic field. The figure also depicts the magnetization data measured above the ordering temperature, 5 K, and well within the paramagnetic region, 9 K. The data at 9 K is more linear which confirms the paramagnetic state of the system at this temperature.

3.2 Specific heat

Specific heat measured is shown in Fig. 3. The data monotonously decreases down to low temperatures and a peak is observed at $T_o = 3.8$ K. The estimation of the location of this anomaly was taken from the maximum peak as indicated by an arrow on the inset of Fig. 3(a). The location of this peak corresponds well with the data obtained in magnetic susceptibility. This data reaches the Dulong-Petit value at 149.652 J/molK, reached at about 150 K, and it is less than the maximum specific heat obtained at room temperature, which is 170 J/molK.

Fig. 3(b) shows the C_p/T vs T^2 curve, with the Sommerfeld coefficient (γ) fit for the data just above the ordering temperature. The red line is the fit according to the expression:

$$\frac{C_p}{T}(T) = \gamma + \beta T^2, \quad (2)$$

where γ is the Sommerfeld coefficient and β is the constant associated with Debye temperature, θ_D . The calculated Debye temperature was found to be 226(1) J/molK, while the obtained Sommerfeld coefficient, $\gamma = 0.098(2)$ J/molK².

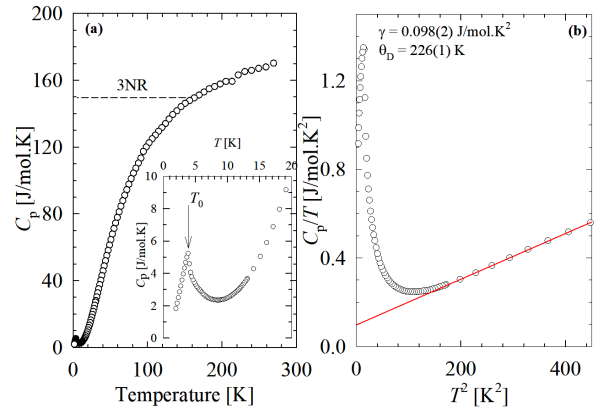


Fig. 3. (a) Specific heat of CeOsGa₄ showing data from 275K down to 2K, with the inset showing the low temperature data with an arrow pointing at the peak associated with ordering temperature. (b) C_p/T vs T^2 curve with Sommerfeld coefficient fit to the specific heat on the data just above T_o .

3.3 Electrical resistivity

The electrical resistivity, $\rho(T)$ measured in zero applied magnetic field is displayed in Fig. 4. The data is characterised by a metallic behaviour at high temperatures where deviations around T_o are observed with the

decrease in temperatures. The drop in $\rho(T)$ is ascribed to the scatter on the long-range ordered magnetic moments. The inset shows the low temperatures where position of the ordering temperature is expected. The arrow points at temperature that corresponds to anomaly that exhibited ordering temperature in $C_p(T)$ and $\chi(T)$ data.

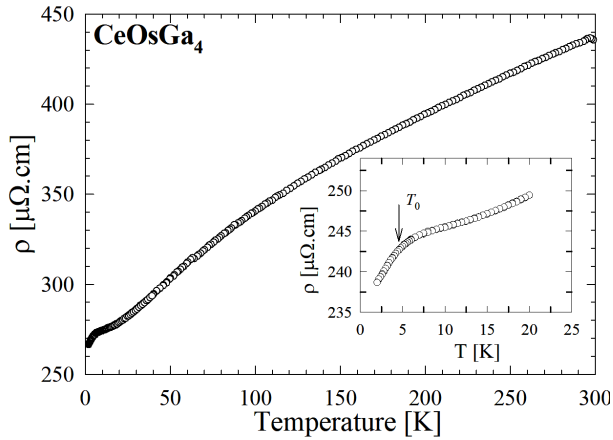


Fig. 4. Electrical resistivity measured from 300 K down to 2 K at zero applied magnetic field. The inset reveals the low temperature region, where the arrow indicates the location of the ordering temperature.

4. Conclusion

The physical properties of CeOsGa_4 were studied through magnetic studies ($\chi(T)$, $M(B)$), specific heat and electrical resistivity, $\rho(T)$. An antiferromagnetic anomaly was observed at $T = 3.8$ K. The compound shows signs of ferromagnetism at high applied magnetic field. This finding is further investigated and will be reported elsewhere.

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