

Magnetic and Physical properties of the layered compound

Introduction

R₃T₄X₁₂ type of compounds are of particular interest among intermetallics because the crystal structure contains layers as well as triangular and distorted Kagomé lattice features [1-3]. The arrangement of the atoms carrying magnetic moments at the vertices of the structure and the competition between ferro- and antiferromagnetic interactions can lead to the appearance of magnetic frustration phenomena. For instance, in Gd₃Ru₄Al₁₂, geometrical frustration together with the formation of ferromagnetic trimers due to the long-range RKKY interaction is observed at low temperatures [4]. Several studies have been done on Ru-based compounds in this series of aluminides. However, no physical and magnetic properties have been reported yet on Os-based compounds (except on Gd₃Os₄Al₁₂) synthesized for the first time by Niermann [5]. This work is the first report on physical properties of Ce₃Os₄Al₁₂ with the focus on the possible effects of the geometric frustration.

Experimental

The metals used as starting materials were cerium (solid bar, 99.99 mass% purity), Osmium (powder, 99.9+ mass% purity), and Aluminium (granules, 99.999 mass% purity). A polycrystalline sample of 1.5 g, was prepared by arc-melting under the argon atmosphere in an Edmund Bühler arc-melting furnace. After melting the sample was annealed in a resistance furnace at 900 °C for two weeks and finally water quenched. Powder x-ray diffraction was performed on a powdered specimen using the Rigaku diffractometer with Cu- α radiation. Data of physic properties were recorded using a Dynacool physical and magnetic properties measurement system from Quantu Design (San Diego).

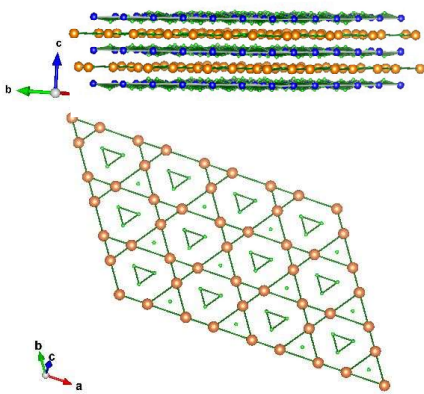


Fig. 1. (a): Layered representation of the crystal structure Ce₃Os₄Al₁₂ with Ce (Orange spheres), Os (blue spheres) and Al (green). (b): The Ce₃Al₄ layer showing the distorted Kagome nets.

References

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- [2] M. S. Henriques *et al.*, Phys. Rev. B **97** (2018) 014431
- [3] D. I. Gorbunov *et al.*, Phys. Rev. B **90** (2014) 094405
- [4] S. Nakamura *et al.*, Phys. Rev. B **98** (2018) 054410
- [5] J. Niermann *et al.*, Z. Anorg. Allg. Chem. **628** (2002) 2549-2556

Results

Formation and structure. The powder x-ray diffraction spectrum of this sample was successfully refined on the basis of the hexagonal Gd₃Ru₄Al₁₂ structure type with P6₃/mmc space group. The obtained lattice parameters are $a = 0.889$ (1) nm and $c = 0.953$ (1) nm. These values are in good agreement with an earlier report [5]. The whole system is a layered structure. The R atoms are arranged as a distorted Kagomé net in the Ce₃Al₄ layer, and the Al atoms occupy two different sites in the Os₄Al₈ puckered layer (see Fig. 1).

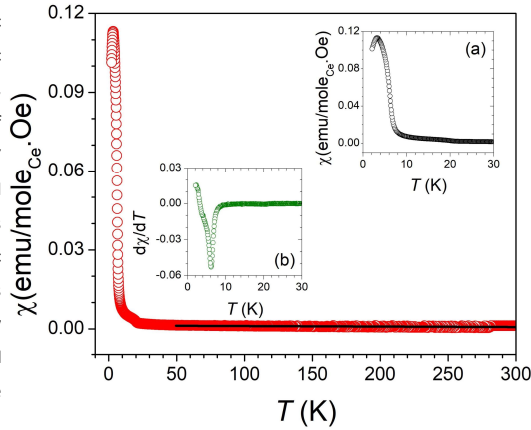


Fig. 2. Main panel: magnetic susceptibility of Ce₃Os₄Al₁₂ measured in a constant dc-magnetic field of 0.1 T. The black line represents the least-squares fit of the modified Curie-Weiss relation ($\chi(T) = \chi_0 + N_A \mu_{\text{eff}}^2 / 3k_B(T - \theta_p)$). The obtained positive paramagnetic Weiss temperature ($\theta_p = 5.33$ K) indicates the dominant presence of ferromagnetic interactions in the high temperature region. The obtained effective magnetic moment ($\mu_{\text{eff}} = 0.94 \mu_B/\text{Ce}$) is around one third the full free-ion moment value $2.54 \mu_B/\text{Ce}$. Inset (a): highlight of the low-temperature region. Inset (b): represents the first derivative of $\chi(T)$. The sharp peak in the plot of $d\chi/dT$ indicates the presence of a magnetic phase transition.

Fig. 3. Isothermal magnetization at temperatures between 2 K and 20 K. Broad curvatures are observed below 6 K with tendency to saturation at 9 T. The saturation magnetization is only about 0.06 μ_B/Ce which is considerably reduced compared to the free ion saturation value $2.16 \mu_B/\text{Ce}$. The quasi-linear behavior down to 6 K indicates a paramagnetic state.

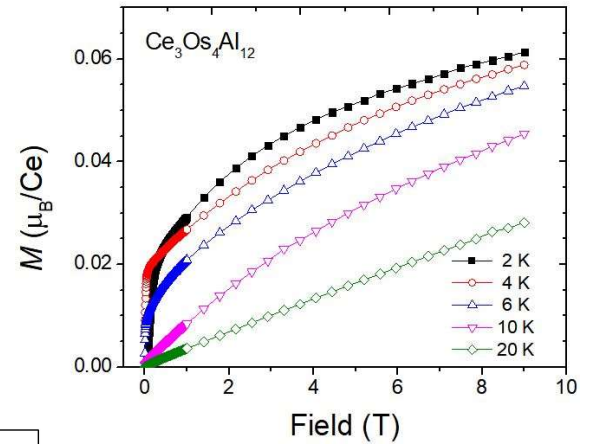
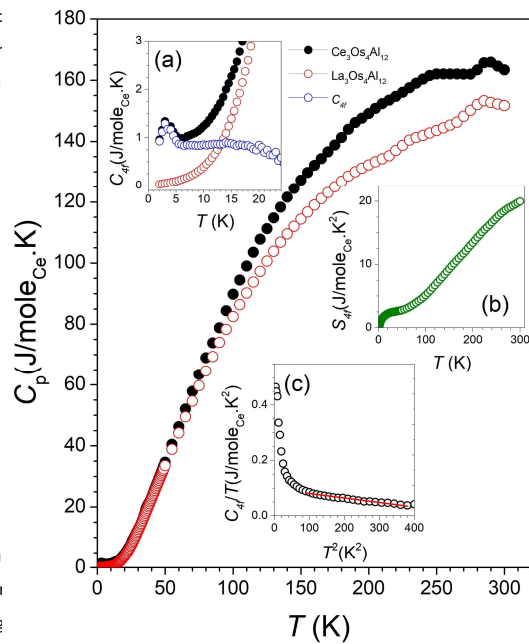


Fig. 4. Main panel: Specific heat of Ce₃Os₄Al₁₂ and La₃Os₄Al₁₂ against temperature. Inset (a) represents the low-temperature region. The blue symbols represent the magnetic contribution to the specific heat obtained by subtracting the specific heat of La₃Os₄Al₁₂ from that of Ce₃Os₄Al₁₂. The kink observed around the transition temperature $T_c = 3$ K is a sign of a short-range order-like transition. Inset (b) shows the 4f contribution to the entropy per Ce as a function of temperature. The magnetic contribution released at T_c is about $0.6 \text{ J/mole}_{\text{Ce}} \cdot \text{K}^2$ which is only a fraction of $R \ln 2$. A linear behavior observed from about 100 K to 300 K confirms that there is no tendency to saturation. Inset (c): specific heat $C_p(T)/T$ vs T^2 . The red line represents a linear fit which gives an estimation of the Sommerfeld coefficient $\gamma = 0.097(1) \text{ J/mole}_{\text{Ce}} \cdot \text{K}^2$. This value is an enhancement of about 20 times over that of a normal metal.



Conclusions

Ce₃Os₄Al₁₂ is a new example of layered Kagome structure with possible effects of the geometric frustration. The $\chi(T)$ and $C_p(T)$ data confirms the presence of a short-range order-like transition. A field study will be carried out further to confirm the nature of the transition.

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