Magnetic properties and magnetocaloric effect of a distorted Kagomé lattice: $Gd_3Os_4Al_{12}$

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Abstract. We report on the magnetic properties and magnetocaloric effect of a new distorted Kagomé lattice compound $Gd_3Os_4Al_{12}$. The temperature dependent dc-magnetic susceptibility ($\chi(T)$) reveals that the compound undergoes successive ferromagnetic and antiferromagnetic orderings below 30 K. The estimated effective magnetic moment from magnetic susceptibility confirmed that the 4f-shell fully contributes to the magnetic properties. The obtained positive paramagnetic Weiss temperature indicates the presence of strong ferromagnetic interactions. The isothermal magnetic entropy change was evaluated from the magnetization isotherms and yielded a value of 4.6 J/kg-K at 9 T.

1. Introduction

Magnetocaloric effect is an interesting topic in the physics of rare-earth based magnetic compound due to the application in refrigeration and the environmental friendly application of materials with large magnetocaloric effect (MCE). Gadolinium (Gd) and Gd-based compounds have drawn particular attention on this regard, because Gd - itself shows a large MCE around room temperature. Typically, a good MCE material has large isothermal magnetic entropy change, adiabatic temperature change and cooling power close to a magnetic phase transition temperature. It is also proposed that emerging large MCE can be obtained in some magnetically frustrated systems [1, 2]. In these latter systems, the magnetic spin type of ordering is frustrated due to aspects of symmetries in the lattice. The system is unable to satisfy all the pairwise interactions simultaneously. In a certain class of frustrated system, the rare-earth atom form distorted Kagomé nets and triangles. For this purpose, we have synthesized a Gd - based distorted Kagomé lattice compound namely $Gd_3Os_4Al_{12}$ which was first reported by J. Niermann and W. Jeitschko in 2002 [3]. The magnetic properties and magnetocaloric effect were investigated using dc-magnetization measurements and the results are reported in details.

2. Synthesis and Experimental Details

A polycrystalline sample was synthesized by arc-melting stoichiometric amounts of high - purity (99.99 mass % purity or better) elements (Gd, Os and Al) under argon atmosphere in an Edmund Buhler arc-melting furnace. The Rietveld refinement of the powder x-ray diffraction pattern indicated that the compound crystallizes in the $Gd_3Ru_4Al_{12}$ type hexagonal structure with space group $P6_3/mmc$ (not shown in this paper). DC-magnetic susceptibility and isothermal magnetization measurements were performed using a commercial Dynacool physical properties

measurement system from Quantum Design, USA. The measurement was carried out in the temperature range between 1.8 K to 300 K and fields upto a maximum value of 9 T.

3. Results and Discussion

Figure 1 shows the dc-magnetic susceptibility as function of temperature (left axis). The data was obtained during the cooling process from 300 K to 1.8 K under a magnetic field of 0.1 T. As seen in the inset of figure 1, the compound exhibits two magnetic phase transitions at approximately 6 K and 18 K and is marked by arrows in the inset. It is also observed that the $\chi(T)$ has another transition around 150 K. Further investigation is required to find the details for that transition at high temperatures.

The inverse of $\chi(T)$ is plotted and shown on the right axis. As seen from the same figure, the high-temperature region T > 200 K is followed by the Curie-Weiss law, which is given by: $\chi = N_A\mu_{eff}^2/3k_B(T-\theta_P)$ where N_A and k_B are respectively the Avogadro number and Boltzmann constant. The fitting is shown as a black line on the $\chi^{-1}(T)$ data. The least-squares fit yields an effective magnetic moment $\mu_{eff} = 8.0\ \mu_B/Gd^{3+}$. This results indicate that the 4f-shell of Gd^{3+} is fully contributing towards the magnetic state of this compound. The positive sign of the paramagnetic Curie temperature $\theta_P = 60$ K suggests the presence of dominant ferromagnetic exchange interactions in the paramagnetic region. A strong deviation from the high-T Curie-Weiss behavior is observed at about 200 K as the temperature is lowered. This is due to the gradual presence of short-range correlations.

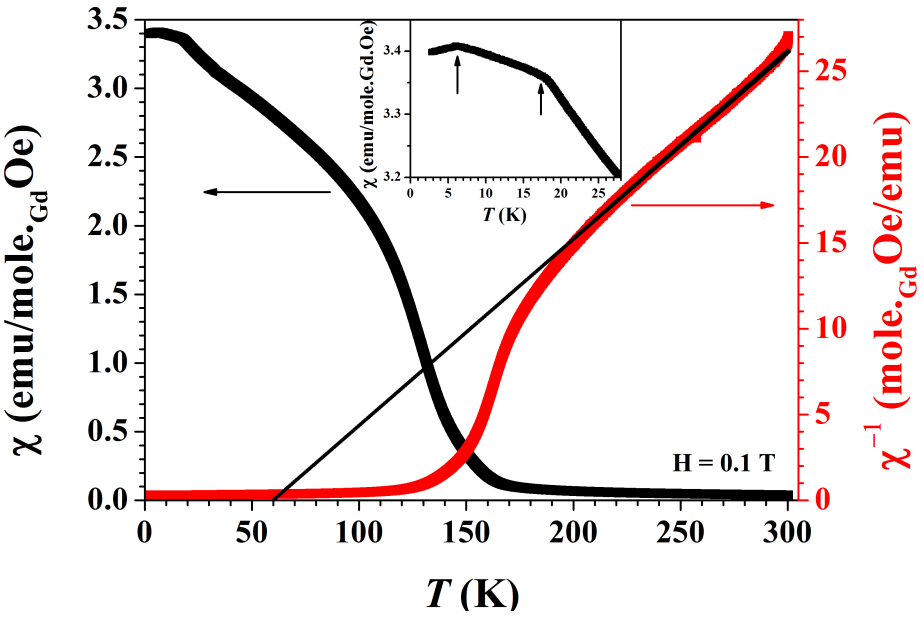

Figure 1. Left axis: temperature dependence of dc-magnetic susceptibility of $Gd_3Os_4Al_{12}$ in the temperature range 1.8 K - 300 K measured in a magnetic field of 0.1 T. Right axis: the inverse magnetic susceptibility of $Gd_3Os_4Al_{12}$. The black solid line represents the Curie-Weiss fit as described in the text.

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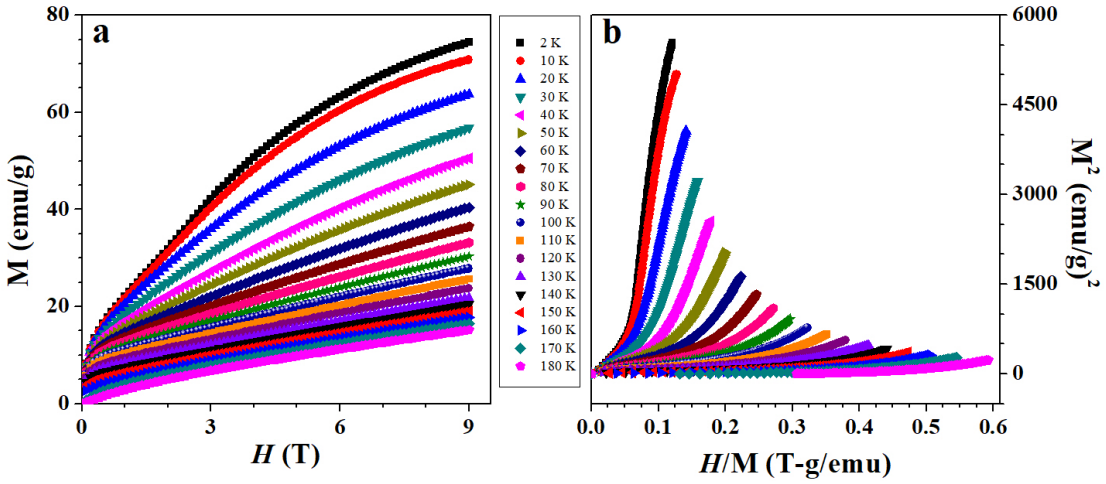

Figure 2. **a**) Isothermal magnetization curves of $Gd_3Os_4Al_{12}$ as function of magnetic field measured at selected temperatures up to 180 K. **b**) Standard Arrott - plots of isotherms corresponding to those in (a).

To further investigate the magnetic behavior of the compound, the isothermal magnetization measurements were performed from 2 K to 180 K in fields up to 9 T and is shown in figure 2**a**. As seen from the figure, the magnetization curves below 20 K have a tendency to saturate in high magnetic fields. The obtained magnetic moment reaches approximately 7.0 μ_B/Gd^{3+} at 2 K for 9 T, which is equal to the saturation magnetic moment of Gd^{3+}. This indicates that the magnetic moments of Gd^{3+} are nearly fully aligned parallel to the applied field. Arrott plots (M^2 *vs.* H/M) were derived from the magnetization isotherm curves to find the order of magnetic phase transitions. According to the Banarjee criterion [4]; a positive slope in the M^2 *vs.* H/M plots indicates second order magnetic phase transition, whereas, negative slope represents first order magnetic phase transition. In this case, positive slopes are observed (see figure 2**b**) meaning that the transition in this compound is of second order.

The isothermal magnetic entropy change ($-\Delta S_M$) was estimated from the isothermal magnetization curve using the Maxwell relation [5]; $\Delta S(T,H) = \int_0^H (\partial M/\partial T)_H dH$, where H is the applied magnetic field. Figure 3 shows the $-\Delta S_M$ changes as function of temperature for different magnetic fields. The maximum value of $-\Delta S_M$ is 4.6 J/kg-K for the change of field 9 T around 20 K. Another important factor is the refrigeration capacity (RC), which measure the amount of heat transfer between the cold and hot reservoir in an ideal refrigerator cycle [6, 7]. This RC value was calculated by integrating the area under the curve of $-\Delta S_M$ *vs.* T using the formula RC = $\int_{T_1}^{T_2}(-\Delta S_M)dT$, where T_1 and T_2 are the temperature corresponding to the left and right side at the half-maximum value of $-\Delta S_M$ respectively [8]. The obtained value of RC for a field change of 9 T is 177.804 J/kg. These obtained results are relatively small in comparison with other Gd-based ternary compounds [9, 10, 11].

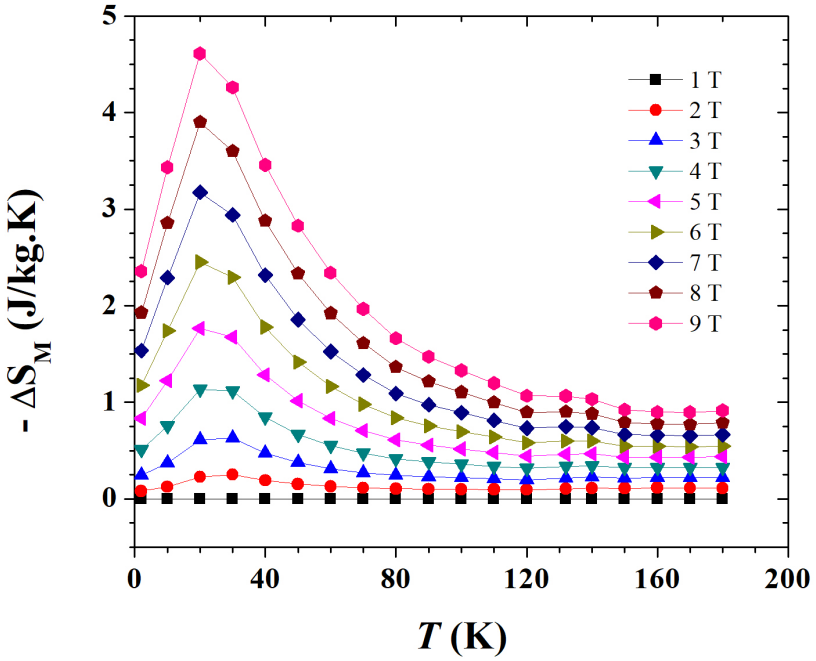

Figure 3. Temperature dependence of the isothermal entropy changes measured in fields up to 9 T as described in the text.

4. Conclusion

In summary, the magnetic properties and magnetocaloric effect of the compound $Gd_3Os_4Al_{12}$ have been studied experimentally. The initial magnetic study revealed that it possesses two magnetic transitions below 30 K and another transition above 150 K more likely being a structural transition. The Arrott - plots of magnetization suggest that the transition is of second order. The magnetocaloric effect of this compound shows a relatively small value of $-\Delta S_M = 4.6$ J/kg-K for a change of field 9 T compared to other Gd compounds [11, 12] with comparable values to profitable magnetic refrigerant materials. As the structure is likely to be subject to geometrical frustration, further investigations on this compound may lead to a better understanding of distorted Kagomé lattice and frustrated magnetism.

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