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Insight into the magnetism of a distorted Kagome lattice, $Dy_3Ru_4Al_{12}$, based on polycrystalline studies

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ABSTRACT

The layered compound with distorted Kagome nets, $Dy_3Ru_4Al_{12}$, was previously reported to undergo antiferromagnetic ordering below (T_N=) 7 K, based on investigations on single crystals. Here, we report the results of our investigation of *AC* and *DC* magnetic susceptibility (χ), isothermal remnant magnetization (M_{IRM}), heat-capacity, magnetocaloric effect and magnetoresistance measurements on polycrystals. The present results reveal that there is an additional magnetic anomaly around 20 K, as though the Néel order is preceded by the formation of ferromagnetic clusters. We attribute this feature to geometric frustration of magnetism. In view of the existence of this phase, the interpretation of the linear-term in the heat-capacity in terms of spin-fluctuations from the Ru 4d band needs to be revisited. Additionally, in the vicinity of T_N, *AC* χ shows a prominent frequency dependence and, below T_N, M_{IRM} exhibits a slow decay with time. This raises a question whether the antiferromagnetic structure in this compound is characterized by spin-glass-like dynamics. In contrast to what was reported earlier, there is a change in the sign of the magnetoresistance (MR) at the metamagnetic transition. A butter-fly-shaped (isothermal) MR loop (interestingly spanning over all the four quadrants) is observed at 2 K with distinct evidence for the magnetic phase co-existence phenomenon in zero field after travelling through metamagnetic transition field. The results on polycrystals thus provide additional information about the magnetism of this compound, revealing that the magnetism of this compound is more complex than what is believed, due to geometric frustration intrinsic to Kagome net.

1. Introduction

Recently [1], single crystals of the compound, $Dy_3Ru_4Al_{12}$ [Ref. [2]], crystallizing in $Gd_3Ru_4Al_{12}$-type hexagonal structure (space group: $P6_3/mmc$), were reported to exhibit interesting magnetic properties owing to certain structural features. The crystal structure [Refs. [2–3]] contains planar layers of Gd_3Al_4 and puckered layers of Ru_4Al_8 stacked alternately along the *c*-axis. The magnetic atoms occupy the vertices of distorted Kagome nets and triangles, thereby favoring magnetic frustration for intersite antiferromagnetic coupling. Many interesting anomalies were reported [1], which require further understanding. These are: (i) *DC* magnetization (*M*) data taken with a magnetic field (*H*) of 10 kOe provided evidence for the onset of long range magnetic order at (T_N =) 7 K, Neutron diffraction results at 1.8 K reveal a complex antiferromagnetic structure. Interestingly, this magnetic transition was proposed to be first-order on the basis of heat-capacity (*C*) studies, in contrast to the expectation of second-order magnetic ordering. (ii) Magnetization jumps at different magnetic fields for different orientations of the single crystals favoring field-induced ferromagnetism have been found. There are corresponding jumps in magnetoresistance (*MR*). However, a *positive* magnetoresistance was observed even at high fields, which is unusual for the high-field ferromagnetic state. (iii) On the basis of large linear term in heat-capacity (*C*) above 7 K, it was proposed that Ru 4d band exhibits spin-fluctuations. We have carried out exhaustive investigations on polycrystalline samples by *AC* and *DC* magnetic susceptibility (χ) with relatively low-fields, isothermal magnetization, heat-capacity and magnetoresistance studies. We obtain additional information, which are reported in this article. We think that the understanding of this compound with interesting crystallographic features (as mentioned above) favoring geometrically frustrated magnetism is still in its infancy.

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2. Experimental details

A polycrystalline specimen of the compound, $Dy_3Ru_4Al_{12}$, was prepared by melting together stoichiometric amounts of high purity (>99.9%) constituent elements in an arc furnace in an atmosphere of argon. The ingot was subsequently vacuum-annealed at 800 C for a week. The Rietveld-fitted powder x-ray diffraction pattern (Cu K_α), shown in Fig. 1, confirms single phase nature of the sample. Scanning electron microscopy (SEM) was used to further characterize the sample. The temperature (T) dependence of *AC* as well as *DC* χ was measured (1.8–300 K) on a specimen of 8 mg with a commercial (Quantum Design) SQUID magnetometer. For *AC* measurements, the field employed was 1 Oe. *M(H)* curves were also obtained at several temperatures. The electrical resistivity (ρ) as a function of T in the presence of several magnetic fields and $\rho(H)$ curves at several temperatures were obtained with a commercial Physical Property Measurements System, PPMS (Quantum Design); heat-capacity was also studied with the same PPMS. Unless otherwise stated, all the measurements were performed for the zero-field-cooled (ZFC) condition of the specimen. An analogue, $Y_3Ru_4Al_{12}$, was also prepared by arc melting constituent elements to obtain lattice contribution to heat-capacity.

3. Results

3.1. Dc magnetic susceptibility

Magnetic susceptibility measured in the presence of 5 kOe as a function of T in the form of χ and inverse χ is plotted in Fig. 2a. There is a distinct peak at 7 K, consistent with antiferromagnetic order [1]. From the Curie-Weiss fitting of the high temperature data, we find that the value of the paramagnetic Curie temperature (θ_p) is 20 ± 1 K. The positive sign of θ_p implies the presence of ferromagnetic correlations, as emphasized in Ref. [1], though the compound orders antiferromagnetically. Above 7 K, inverse χ exhibits a deviation from the high temperature (>150 K) Curie-Weiss behavior as the T is lowered and is attributed to short-range magnetic correlations gradually building below 150 K. These are in good agreement with Ref. [1]. However, measurements in small fields (20 and 100 Oe) reveal a bifurcation of the curves, obtained for ZFC and field-cooled (FC) conditions, below about 18 K (see Fig. 2b). We have measured χ in the presence of additional fields and the curves thus obtained tend to show dominant bifurcation around this temperature (Fig. 2b). The sharpness of the change in the slope of the curve around 20 K in Fig. 2b is large for low field measurements. [AC χ data (see below) also show a feature around 18 K]. These findings suggest that there is another magnetic feature above T_N. The effective moment obtained from the high temperature linear region (that is, ignoring temperature independent component) in the plot of inverse $\chi(T)$ is 10.5 ± 0.03 μ_B per Dy, in close agreement with that expected for trivalent Dy (10.63 μ_B). However, if we fit to modified Curie-Weiss form (i.e., adding a temperature independent component, χ_0), one gets a value of 3.4 × 10^{-3}emu/mol for χ_0 with a more satisfactory value for the effective moment.

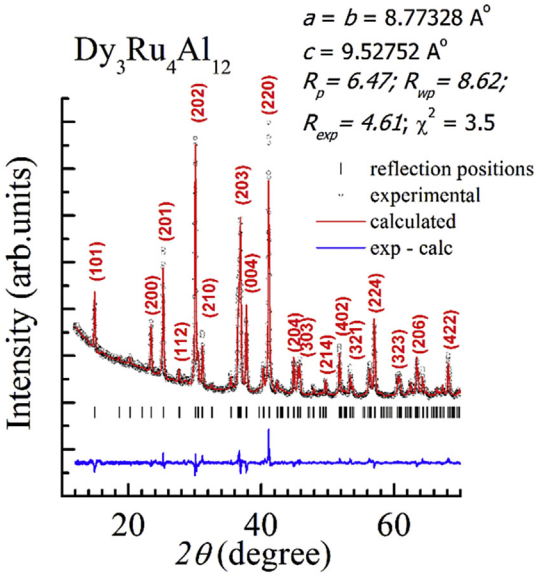

Fig. 1. X-ray diffraction (Cu K_α) pattern for $Dy_3Ru_4Al_{12}$. Rietveld fitting, along with fitted parameters and lattice constants, is also included.

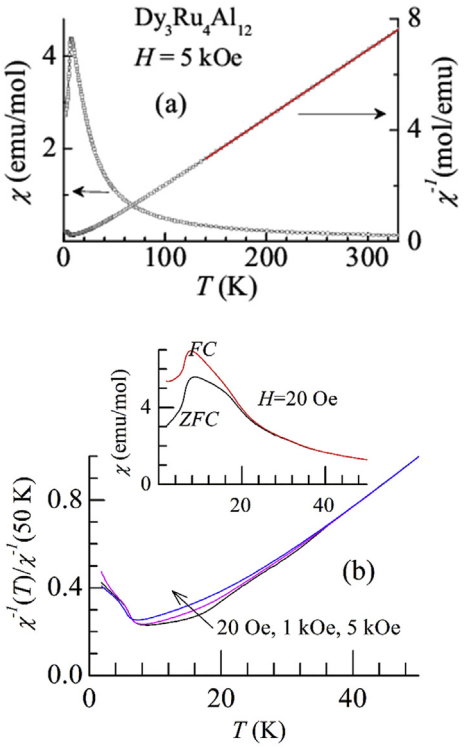

Fig. 2. (a) Magnetic susceptibility and inverse susceptibility as a function of temperature measured in a field of 5 kOe for $Dy_3Ru_4Al_{12}$. The continuous line in the inverse χ plot represents modified Curie-Weiss fit above 150 K. (b) Inverse susceptibility as a function of temperature below 50 K measured in various fields; in the inset, zero-field-cooled and field-cooled susceptibility taken in a low-field is plotted to show the bifurcation of the curves.

3.2. Isothermal dc magnetization

We have measured isothermal magnetization in 2 K temperature steps between 2 and 90 K, in order to see the behavior of isothermal entropy change, $\Delta S = S(H) - S(0)$, at low temperatures, to offer support to the magnetic ordering behavior presented above. Derivation of ΔS employing Maxwell's thermodynamic relationship between magnetization and entropy and its relationship to the nature of magnetic ordering have been discussed at several places in the literature [4]. *M(H)* curves at selected temperatures are shown in Fig. 3. The values of ΔS obtained are shown in Fig. 4 for a change of the magnetic field from zero to a selected field (5, 10, 30, or 50 kOe). It is evident from this figure that the magnitude of ΔS gradually increases with decreasing T, attaining a peak, e.g., at a reasonably large value of 6.5 J/kg-K for a final field of 50 kOe. It is