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Physical properties of antiferromagnetic single crystal $GdIn_3$

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ABSTRACT

A sample of $GdIn_3$ was obtained via the metallic flux technique and its structural, magnetic, and thermodynamic properties were studied. X-ray diffraction (XRD) measurements realized at room temperature were performed in order to verify whether the desired crystalline phase had been obtained. The experimental results show that single crystals were grown successfully by the employed growth technique. The magnetic characterization was carried out through magnetic susceptibility data as a function of temperature ($2 \leq x \leq 300$ K) and magnetic field ($-7 \leq H \leq 7$ T). The specific heat was measured using a commercial small mass calorimeter that employed the thermal relaxation technique in the temperature range of 2–200 K.

1. Introduction

The synthesis of intermetallic rare-earth-based compounds has been a wide and promising area of development, and studies of interesting physical phenomena of condensed matter have been carried out. The systems of the type RM_3, where R corresponds to elements of the lanthanide series and in general Me represents a transition metal, have been largely synthesized and characterized as exhibiting diversified physical behaviors such as the magnetic ordering, influence of crystalline electric field (CEF) on the magnetic properties, unconventional superconductivity, fluctuating valence, highly correlated electron systems, and non-Fermi-liquid (NFL) behavior, among others [1–4].

The compounds of the RIn_3 (R=earth rare) family show interesting magnetic properties that are associated with unpaired electrons of the layer 4*f*. For R=Gd, Tm, Pr, Er, Tb, Dy, Ho, Nd, and Ce, a magnetic transition temperature T_N that indicates the onset of the antiferromagnetic (AFM) ordering is observed. To R=Ce is a composed heavy fermion with a temperature of antiferromagnetic ordering of $T_N=10$ K that also presents Kondo-type behavior. Specifically, the compound $GdIn_3$ is antiferromagnetically ordered with a Néel temperature of $T_N \approx 45$ K and crystallizes in the cubic structure of the $AuCu_3$ type (space group *Pm*-3*m*) with a lattice parameter value of 4.6068 Å [5]. When the value of T_N for $GdIn_3$ is compared with the Gd-based tetragonal of type Gd_xMIn_y (x=1 or 2; y=5 or 8; *M*=Rh. Ir, Co), a slight change of the ordering temperature is observed, with a maximum value for the compound $GdIrIn_5$ ($T_N=42$ K) and a minimum for $GdCoIn_5$ ($T_N=30$ K). A similar feature is observed for isostructural $SmIn_3$ when compared with its relative tetragonal [6–8].

The great interest in studying magnetic Gd-based materials arises from the ground state characteristic *S*-purely ($S=7/2$, $L=0$) presented by the magnetic ion Gd^{3+} no having in first order effects of crystalline electric field as well as the effects that appear due to the spin-orbit coupling, with the magnetic properties being determined by interactions of the type RKKY and Fermi surface effects. Moreover, study of this type of compound (e.g. R_xMIn_y; x=1 or 2; y=5 or 8; *M*=Rh, Ir series) has generated considerable attention in the scientific community because the isostructural members with R=Ce were found to be heavy fermions in superconducting state. For example, the compound $CeIn_3$ shows superconductivity below about 0.2 K and 26.5 kbar of hydrostatic pressure. The related tetragonal (eg. $CeIrIn_5$, $CeRhIn_5$) believed that the unconventional superconductivity of these compounds is mediated by magnetic interactions and not by electron–phonon interactions as in the case of conventional superconductors [9,10].

In this work, we revisited the magnetic, structural, and thermodynamic properties of single crystals of $GdIn_3$ grown from an In flux. The *T*-dependence of the magnetization shows a peak magnetic ordering temperature $T_N=45$ K, which is consistent with the value observed in the measurement of specific heat. The

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magnetization measurements $M(H)$ taken at 2 K show a linear increase in fields of up to 7 T.

2. Experimental details

Single crystals of $GdIn_3$ were grown from In flux melt as described elsewhere [11]. The structure and phase purity were confirmed by X-ray powder diffraction (XRD) measurements taken at room temperature. The Rietveld method [12] was applied to estimate the lattice parameter. Magnetization measurements in function of the field ($-7\,T \leq H \leq 7\,T$) and of the temperature ($2\,K \leq T \leq 300\,K$) were accomplished using a commercial superconducting quantum interference device (SQUID). The data of specific heat were obtained by a physical properties measurement system (PPMS-Quantum Design) in the temperature range between 2 and 200 K.

3. Results and discussion

Fig. 1 shows the XRD pattern taken at room temperature from crushed fragments of the single crystal $GdIn_3$. The powder XRD pattern confirms the cubic structure of the $AuCu_3$-type space group Pm-3m (221). The experimental pattern was compared to the pattern calculated from structural refinement using the Rietveld method. The obtained value of the lattice parameter was a=4.6014 (4) Å.

Fig. 2 shows the temperature-dependent magnetic susceptibilities (M/H) taken at an applied field of 1 kOe. These data show a robust antiferromagnetic transition of T_N=45 K that arises from the interaction of the RKKY-type exchange between the unpaired 4f electron-mediated electron conduction. From Curie–Weiss fits of the data at $T > 100$ K, we have extracted the parameters of the paramagnetic Curie temperature θ_p and the effective magnetic moment (μ_{eff}). The calculated value of μ_{eff} was 8.1μ_B, which is consistent with the theoretical value, considering the free ion, for $Gd^{3+}\{(J=7/2);\ \mu_{eff} = g[(J(J+1))]^{0.5}\mu_B = 7,\ 9\mu_B\}$. The value of 8.1$\mu_B$ is slightly higher than the theoretical value, possibly due to the additional contribution from 5d-site moments parallel to moments from ions $-4f$ already know thing in intermetallic Gd-based [13, 14]. The paramagnetic Curie-Weiss temperature was $\theta_P = -93$ K.

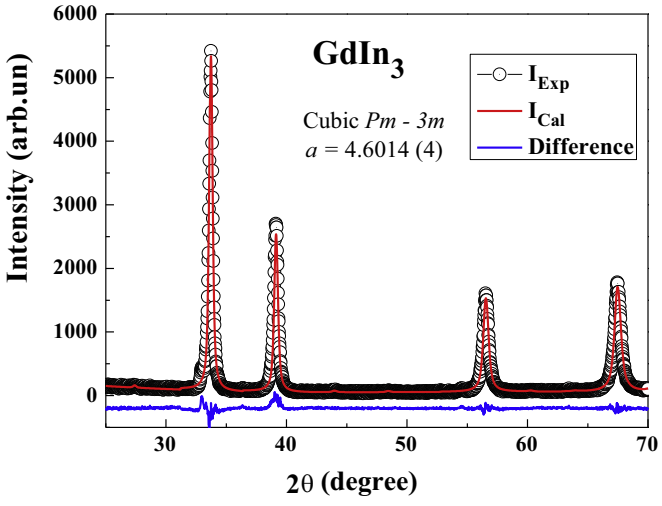

Fig. 1. XRD patterns taken at room temperature from crushed single crystal $GdIn_3$. The solid lines are the best fit obtained by the Rietveld method (red lines) and the difference (blue lines) between the experimental and calculated patterns. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

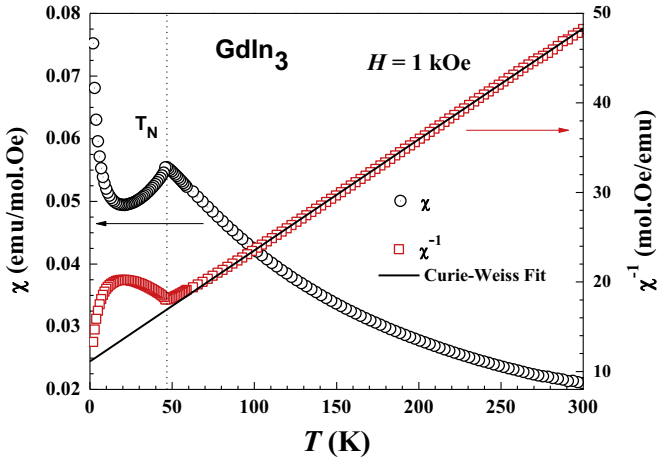

Fig. 2. T-dependence on magnetic susceptibility taken at a field of 1 kOe. The black solid line represents the Curie-Weiss fit at high temperature.

Therefore, $|\theta_P| > T_N$ indicates the existence of magnetic frustration with the figure of merit $f=|\theta_P|/T_N \approx 2$. In this case the frustration could be related to competition between the exchange interactions among the first neighbors and second-nearest neighbors. According to reference [15], the magnetic structure of $GdIn_3$ is comprised of FM chains along the c-direction and AFM exchange interactions in the ab plane. Therefore, the magnetic competition between these phases is certainly responsible by magnetic frustration. In fact, the effects of the crystalline electric field and spin orbit interaction can be disregarded because $L=0$ and 4f functions from Gd^{3+} are symmetric. Besides, the increase of the magnetization to $T < T_N/3$ may indicate that magnetic competition causes an increased contribution the ferromagnetic component at low temperatures or even a spin reorientation process at low temperatures.

Furthermore, when compared to T_N from $GdIn_3$ with respect to tetragonal (eg. $GdIrIn_5$, $GdRhIn_5$), a significant change of magnetic ordering temperature is not observed. If we consider the spatial dependence of the effective exchange parameter, $J_{Gd\text{-}Gd}$, we observe that the symmetry change cubic-tetragonal does not significantly affect the magnetic interaction from compounds. This simple scenario is inconsistent with compounds that are strongly affected by the effects of the CEF $NdIn_3$-like when compared with their respective tetragonal. In this case, although there was an increase in the distance of the Nd-Nd ions, a significant increase of the magnetic ordering temperature was observed, which could be attributed to changes in the crystal field scheme of tetragonal and $NdIrIn_5$ $NdRhIn_5$ and the influence of frustration from the exchange parameter between nearest neighbors [16].

The magnetization isotherms M(H) taken at T=2 K are show in Fig. 3. It is possible to see that the magnetization in any of compounds not reaches magnetic saturation showing a lineal increase without to reveal any anomaly in the curve of M (H) being characteristic from compounds with AFM ordering without induced transition by field.

The total specific heat of the $GdIn_3$ sample is shown in Fig. 4 as well as the curve of specific heat from the isostructural nonmagnetic compound YIn_3. In the measurement of the $GdIn_3$ sample, a well-defined anomaly of the λ-type can be observed, which characterizes the temperature from the onset of AFM magnetic ordering at T_N=47.6 K, which is consistent with the obtained value of the measure of magnetic susceptibility. The clear peak like exhibited at T_N is indicative of a single magnetic transition with a weak character of first order, similar to the transition from the tetragonal compound Gd_2CoGa_8 with T_N=20 K [17].

At low temperature, the data of specific heat can be described