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Generation of magnetic phase diagram of $HoRu_2Si_2$ using magnetocaloric effect

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ABSTRACT

To get complete knowledge about the evolution of different magnetic phases on application of temperature and magnetic field magnetic, magnetotransport and magnetocaloric properties of $HoRu_2Si_2$ have been investigated from 2 to 100 K in magnetic fields ranging from 0 to 70 kOe in detail. The complex magnetic phase diagram of $HoRu_2Si_2$ has been generated by studying magnetocaloric effect (MCE). Three ordered phases have been observed. With lowering of temperature in the presence of lower magnetic field (<10 kOe) two different types of ordered antiferromagnetic phases have been observed. Both of the phases show metamagnetic transition on application of external magnetic field greater than the required critical field. An efficient procedure to construct magnetic phase diagram using MCE has been described. The overlap of the data points obtained from the magnetization and magnetotransport data at the phase boundaries with those generated by MCE indicates the strength of this powerful technique.

1. Introduction

Magnetic phase diagram (MPD) elucidates the magnetic properties of a magnetic materials. The total information about its magnetic ground state as well as modification of magnetic phases induced by external perturbations namely magnetic field and temperature can be extracted by studying the magnetic phase diagram. For that reason, the generation of MPD of magnetic materials is an important subject of research [1].

Magnetocaloric effect (MCE) is the isothermal entropy change or change of temperature of magnetic materials when subjected to external magnetic field variation under adiabatic condition [2]. Moreover MCE reflects the transformation taking place in spin configuration of magnetic materials. The dependence of MCE on temperature and magnetic field is strongly dependent on the nature of the corresponding magnetic phase transition [2]. It is very useful to obtain information about the magnetic state and magnetic phase transformations in magnetic materials by MCE study. For example, we can get valuable information about magnetic materials like the nature of magnetic ordering [3] and metamagnetic transitions [4]. One relatively better technique to generate MPD is the study of MCE. This is a much more flexible and quicker technique and can detect feeble magnetic transitions due to its superior probing sensitivity than magnetization [5].

Great diversity in magnetic properties has been observed due to complex magnetic structure [6] of the compounds of RT_2X_2 (R=Rare-earth; T=4d or 5d electron metal and X=silicon or germanium) series. In most of the materials of this series more than one magnetic structure [8,7,6] has been observed at low temperature region. Though much attention has been paid to construct MPD of other members of this series [9,5], no report exists about the MPD of $HoRu_2Si_2$ in the literature.

In this paper the MPD of large magnetocaloric and large inverse magnetoresistive intermetallic compound $HoRu_2Si_2$ has been generated by using MCE. A procedure for extracting transition temperatures using MCE in the presence of external magnetic field has been described. The MPD generated from MCE is compared with the phase diagram obtained from magnetization and magnetotransport data. Similar phase boundaries obtained from MCE, magnetization and magnetotransport (MT) data establish the procedure to generate MPD by using MCE on firm ground.

2. Sample preparation and characterization

The ternary polycrystalline $HoRu_2Si_2$ sample was prepared by arc melting in argon atmosphere of constituent elements having purity 99.9% followed by annealing at 800 °C for one week in

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evacuated sealed quartz tube. The X-ray diffraction (XRD) spectrum using Philips PW1710 diffractometer with Cu Kα radiation at 300 K confirms the formation of the compound. The XRD data have been recorded in the 2θ range 10–110° with a step size 0.020°. Detailed magnetization measurement was carried out as a function of magnetic field (H) up to 70 kOe as well as function of temperature (T) up to 100 K using Quantum Design Physical Property Measurement System (PPMS). The temperature dependence of resistivity ($\rho(T)$) measurement was carried out in the temperature range 3–100 K by the conventional four probe method in the absence of magnetic field as well as in the presence of 5, 10, 20, 50 and 70 kOe magnetic fields. The resistivity measurement was performed in zero field cooled (ZFC) condition using conventional four probe technique. MR of the compound at 4, 10, 20, 40 and 80 K and up to 75 kOe magnetic field were calculated by conventional definition [MR=((R(H)−R(0))/R(0))∗100%] in longitudinal configuration.

3. Experimental results and discussion

The room temperature XRD spectrum of the annealed sample has been shown in Fig. 1. The lattice structure of the bulk crystalline sample has been analyzed by standard profile fitting method using FULLPROF 2009 program. The XRD study confirms the single phase nature of the compound which crystallizes in the body centered tetragonal $ThCr_2Si_2$ type structure with space group I4/mmm. The refined lattice parameters are a=4.1462(6) Å and c=9.515(1) Å. The Bragg R-factor and the RF-factor are 1.62% and 2.01% respectively. The lattice parameters (a and c) of the sample are in agreement with the previously reported values [10].

The temperature dependence of ZFC dc magnetization under 500 Oe externally applied magnetic fields is shown in Fig. 2. Under lower magnetic field value (H=500 Oe) the temperature dependence of magnetization shows sharp peak around 19 K and a hump around 3 K. The sharp peak around 19 K indicates antiferromagnetic (AFM) nature of the transition. Besides the long range AFM interaction, strong ferromagnetic interaction in the layer structure has also been confirmed from both experiments and theoretical investigations [11]. The hump around 3 K indicates the presence of a spin reorientation transition [12]. Using the slope of $1/\chi$ in the PM region for 500 Oe magnetic field the effective

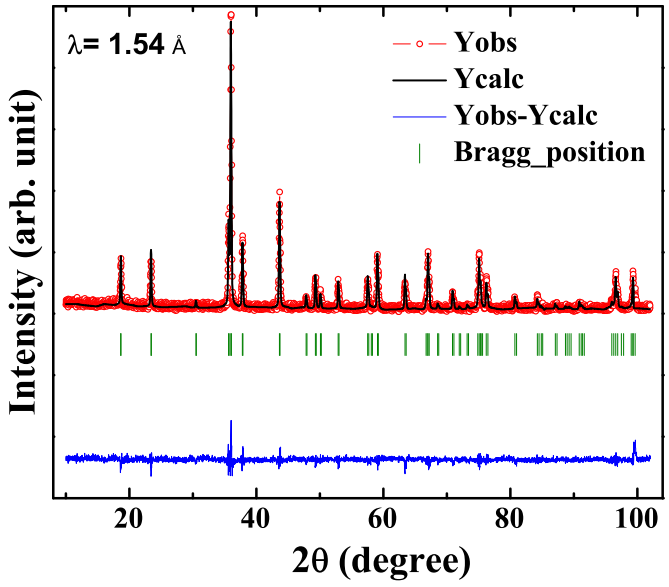

Fig. 1. Profile fitting of XRD spectrum for the annealed sample.

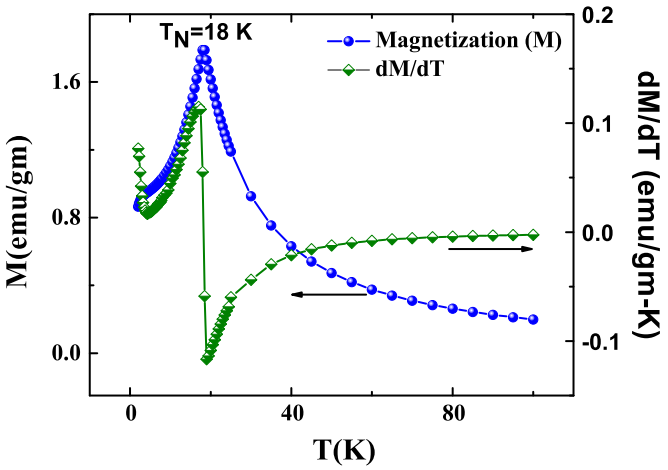

Fig. 2. Temperature dependence of ZFC magnetization (M) and differential magnetization of $HoRu_2Si_2$ for 500 Oe magnetic field.

magnetic moment (μ_{eff}) has been estimated and the value comes out to be 11.97 μ_B/Ho-atom and the paramagnetic (PM) Curie temperature (θ_P) comes out to be +11.2 K. The excess value of μ_{eff} over its theoretical value (10.6 μ_B/Ho-atom) probably arising due to the partial polarization of the conduction electrons [12,13]. The positive value of θ_P indicates the presence of ferromagnetic (FM) interactions within the layer structures of AFM $HoRu_2Si_2$ compound [11,14]. Under application of higher values of magnetic fields the field-induced modification of magnetic transitions was observed in the temperature dependence of susceptibility [see Fig. 1 of Ref. [12]]. With the increase of external magnetic field the AFM–PM transition shifts towards lower temperatures, as expected for typical AFM material [15].

Fig. 3(a) shows magnetization isotherms measured at 4 K and 14 K for applied magnetic fields up to 70 kOe. In low temperature region for lower magnetic field, M changes linearly as a function of magnetic field, indicating antiferromagnetic nature of the magnetic ordering. The presence of AFM to FM spin–flip FO metamagnetic transition (see Fig. 1(b) of Ref. [12]) above certain value magnetic field has been reported in our previous report. Magnetization isotherms for some selected temperatures (for conciseness) are shown in Fig. 3(b).

In equilibrium state, the magnetic entropy change (ΔS_M) of the system due to the application of a magnetic field can be derived from Maxwell relations by integrating over the magnetic field [2]:

$$\Delta S_M(T) = \int_{H_I}^{H_F} \left(\frac{\partial M(H)_T}{\partial T}\right)_H dH \quad (1)$$

where H_I and H_F represent the initial and final values of magnetic field, respectively. Normally ΔS_M is calculated by taking H_I=0 T, i.e.,

$$\Delta S_{0\,T}(T) = \int_{0}^{H_F} \left(\frac{\partial M(H)_T}{\partial T}\right)_H dH \quad (2)$$

The isothermal magnetic entropy changes for different magnetic field changes as a function of temperature ($-\Delta S_{0\,T}(T)$) is shown in Fig. 4. For lower magnetic field values 5 kOe and 10 kOe $-\Delta S_{0\,T}$ is positive in the PM region and its magnitude increases as temperature decreases down to the T_N. After showing a maximum around T_N, the value of $-\Delta S_{0\,T}$ decreases as temperature is lowered it changes sign for lower temperature value and shows a minimum indicating the dominance of AFM nature [16] of the ordering. With further decrease in temperature, $-\Delta S_{0\,T}$ again changes sign at ~5 K and becomes positive and after showing a maximum it tends to