



Multiple phase transitions and magnetoresistance of HoFe_4Ge_2



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ARTICLE INFO

Article history:

Received 4 November 2014

Received in revised form 12 December 2014

Accepted 13 December 2014

Available online 19 January 2015

Keywords:

Rare earth intermetallics

Magnetic properties

Heat capacity

Electrical resistivity

Magnetoresistance

ABSTRACT

A systematic study of the structural, magnetic, heat capacity, electrical resistivity and magnetoresistance properties of HoFe_4Ge_2 has been performed. The temperature dependencies of the magnetization and heat capacity show three magnetic transitions at $T_N = 51$ K, $T_{f1} = 42$ K, and $T_{f2} = 15$ K. The high temperature transition is antiferromagnetic ordering and the two low temperature phase transitions are due to rearrangements of the magnetic structure. A kinetically arrested phase is observed below a freezing point of ~ 11 K. Below 35 K, the behavior of the isothermal magnetization reflects a first-order metamagnetic phase transition. Multiple phase transitions are also manifested in the electrical resistivity behavior. For a field change of 30 kOe, a large magnetoresistance of $\sim 30\%$ is observed near T_{f2} (15 K).

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1. Introduction

Rare earth intermetallic compounds are of particular interest and importance in both fundamental research and various applications based on physical properties related to partially filled 4f shells. For example, heavy fermion behavior [1], Kondo effect [2], quantum criticality [3], magnetocaloric effect [4], magnetoresistance [5] and magnetostriction [6] have been reported to occur in rare earth intermetallics. Therefore, investigation of lanthanide containing compounds is an important pathway towards discovery of new and interesting physical properties. Nearly 25 years ago, rare-earth ternary intermetallic compounds with general composition $\text{R:Fe:Ge} = 1:4:2$ ($\text{R} = \text{Y, Ho, Dy, Er and Lu}$) have been discovered and reported to crystallize with the tetragonal ZrFe_4Si_2 type structure at room temperature [7]. Low temperature neutron and X-ray diffraction investigations of ErFe_4Ge_2 , DyFe_4Ge_2 and HoFe_4Ge_2 [8–13], reveal that these compounds order magnetically at low temperature and are paramagnetic at room temperature despite a relatively large concentration of Fe (57 at.%). A study employing ^{57}Fe Mössbauer spectroscopy and magnetic measurements in high magnetic fields [14,15] also confirmed the paramagnetic state at room temperature of these RFe_4Ge_2 intermetallics.

Among RFe_4Ge_2 compounds, DyFe_4Ge_2 [16] and ErFe_4Ge_2 [17] have been systematically investigated by magnetic measurements; both compounds exhibit interesting and complex magnetic

behaviors. Both DyFe_4Ge_2 and ErFe_4Ge_2 were found to show three magnetic phase transitions, with two of them being associated with the magnetic structure changes and one being an antiferromagnetic (AFM) – paramagnetic (PM) transition. Besides, a re-entrant magnetic glassy state was observed in DyFe_4Ge_2 below the freezing point of 15 K, which is related to the geometrical frustration in the Fe sublattice, making the low temperature antiferromagnetic state metastable [16].

For the title compound HoFe_4Ge_2 , X-ray and neutron diffraction experiments had been performed and reported in the past [11,13] yet a detailed investigation of the physical properties, including magnetic, thermal and electrical, has not been carried out. Based on the results of the neutron diffraction study, HoFe_4Ge_2 exhibits three magnetic transitions, two of them being coupled magneto-elastic transitions at $T_N = 52$ K (from tetragonal $P4_2/mnm$ paramagnetic to orthorhombic $Cmmm$ antiferromagnetic) and $T_{ic1} = 15$ K (from orthorhombic $Cmmm$ back to tetragonal $P4_2/mnm$), the third being a purely magnetic structure change at $T_{c2} = 40$ K. The peculiar re-entrant magneto-elastic transition at T_{ic1} of HoFe_4Ge_2 at low temperature is unique among other RFe_4Ge_2 compounds. This interesting behavior of HoFe_4Ge_2 warrants further investigation of its low temperature magnetic state to uncover the nature of the complex behavior. It is also of interest to study its magnetocaloric and magnetoresistance behaviors. Therefore, in this paper, we present a detailed investigation of the low temperature antiferromagnetic state by using static and dynamic magnetic measurements coupled with heat capacity, and electrical resistivity, and magnetoresistance measurements of HoFe_4Ge_2 .

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2. Experimental details

The Ho and Fe metals used to prepare the HoFe_4Ge_2 compound were obtained from the Materials Preparation Center of the Ames Laboratory [18] and were 99.8+ at.% and 99.98 wt.% pure respectively with respect to all other elements in the Periodic Table including the interstitial impurity elements H, C, O and N. The Ge with a 99.999 wt.% purity was purchased from Alfa Aesar, Inc. A polycrystalline button of HoFe_4Ge_2 was prepared by arc melting of the stoichiometric mixture of constituent elements under an argon atmosphere. The alloy was flipped and re-melted four times to ensure the compositional homogeneity. The crystal structure and phase purity of the sample were studied by X-ray powder diffraction (XRD) using PANalytical X'Pert diffractometer with $\text{Cu K}\alpha_1$ radiation at room temperature. Rietica LHPM software [19] was employed to perform a Rietveld refinement. The alloy adopts the ZrFe_4Si_2 -type tetragonal structure with the lattice parameters $a = 7.2863(5) \text{ \AA}$, $c = 3.8685(3) \text{ \AA}$ at room temperature. Minor amounts of Fe ($\sim 5 \text{ wt.}\%$) and HoFe_2Ge_2 ($\sim 4.5 \text{ wt.}\%$) have been detected as impurity phases. The ac and dc magnetic measurements were carried out using a superconducting quantum interference device (SQUID) magnetometer MPMS XL-7. Isothermal magnetization up to 140 kOe magnetic field was measured using Physical Property Measurement System (PPMS) from Quantum Design Inc. Heat capacity data were collected on heating in an automatic semi-adiabatic heat pulse calorimeter [20] in the temperature range between 2 and 350 K at various magnetic fields after cooling the sample in a zero magnetic field from room temperature.

The sample for the electrical resistivity measurements with dimensions $9.3 \times 1.2 \times 1.5 \text{ mm}^3$ was cut from the polycrystalline button. The electrical resistivity was measured using a standard four-probe method. The temperature (T) and magnetic field (H) dependencies of the electrical resistivity (ρ) were measured with a constant ac excitation electrical current of 25 mA and frequency 35 Hz in the temperature range between 2 and 300 K and in magnetic fields between 0 and 70 kOe. These experiments were performed using PPMS apparatus. The temperature dependencies of the electrical resistivity, $\rho(T)$ were measured in the zero-field-cooled heating (ZFC) and field-cooled cooling (FCC) modes. The isothermal $\rho(H)$ measurements were made after thermal demagnetization by heating to 150 K and then cooling down to the measurement temperature in a zero magnetic field. The magnetoresistance ratio was calculated as $\text{MR} = (\rho_H - \rho_0)/\rho_0 \times 100\%$, where ρ_H and ρ_0 is the electrical resistivity measured in a magnetic field H and zero magnetic field, respectively.

3. Results and discussion

3.1. Magnetic properties

Fig. 1 shows the dc magnetization of HoFe_4Ge_2 measured as a function of temperature in a magnetic field of 1 kOe. In order to clearly depict the phase transitions, the figure only shows the magnetization over the temperature range from 2 to 100 K. The zero-field-cooled heating (ZFC) magnetization shows five anomalies: a kink at 51 K (T_N), a cusp at 42 K (T_{f1}) (also see inset in Fig. 1), a sharp peak at 15 K (T_{f2}), a deep minimum at 11 K (T_{f3}) and a small peak at 4 K (T_{f4}). In agreement with the neutron diffraction measurements of HoFe_4Ge_2 at low temperature [13], the transition at

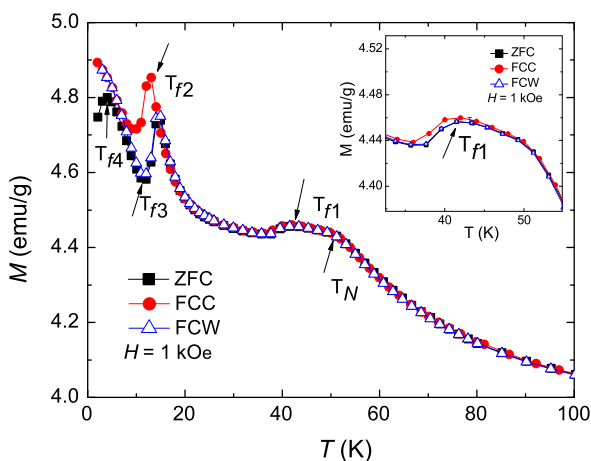


Fig. 1. Temperature dependencies of the magnetization of HoFe_4Ge_2 measured in a 1 kOe applied magnetic field upon ZFC, FCC and FCW conditions. The inset is the expanded view of the details of the regions around T_N and T_{f1} .

51 K reflect bulk antiferromagnetic ordering. The low temperature XRD analysis [5,11] also indicated that this antiferromagnetic transition is coupled with a structural transition from the tetragonal ($P4_2/mnm$ symmetry) to a distorted orthorhombic structure that adopts $Cmmm$ symmetry. Transitions at T_{f1} and T_{f2} , are both related to a magnetic structure change within the antiferromagnetic state. However, the transition at 15 K (T_{f2}) is also related with a structural transition from the orthorhombic back to the original tetragonal (space group $P4_2/mnm$) structure [11]. With regard to the anomalies at T_{f3} and T_{f4} , nothing was observed in either the low temperature XRD or neutron diffraction measurements. It is known that HoFe_2Ge_2 exhibits a long range antiferromagnetic ordering below 1.6 K being paramagnetic above this temperature [21], and therefore, two minor impurity phases do not play a role in these magnetic anomalies. Further, T_{f3} is 9 K in the FCC protocol and the small peak at T_{f4} can only be observed in the ZFC curve. A similar phenomenon was also found in DyFe_4Ge_2 and explained as a freezing point signaling the reentrant glassy state [16]. Therefore, in the case of HoFe_4Ge_2 , a metastable state may also form in the low temperature regime due to geometrical frustration arising from the antiferromagnetic Fe–Fe interactions, as discussed in Ref. [16].

Below 15 K, a strong thermomagnetic irreversibility is observed between the field-cooled cooling (FCC) and field-cooled warming (FCW) plots. A noticeable thermal hysteresis of 2 K between FCW and FCC curves at T_{f2} suggests that this is a first-order phase transition [22], confirming the XRD results of Ref. [11]. It is worth noting that this structural phase transition is incomplete during cooling based on the analysis of XRD results [11]. From the inset of Fig. 1, the expanded range near the T_{f1} transition temperature, a small irreversibility between the FCW and FCC is also observed and from the low temperature XRD results [11], this transition is also first order.

Temperature dependent magnetization of HoFe_4Ge_2 was also measured under applied fields of 5, 10, 15, 25, 30 and 50 kOe, see Fig. 2. Magnetic phase transitions at T_N , T_{f1} , T_{f2} and T_{f3} can still be clearly observed at $H = 5, 10$ and 15 kOe. The anomaly at the lowest temperature $\sim 4 \text{ K}$ is not observed in fields higher than 1 kOe and the peak at T_{f2} in $H = 15 \text{ kOe}$ is smoothed out compared to that at $H = 1 \text{ kOe}$ (Fig. 1). The temperature of the transition at T_{f2} decreases from 15 K in 1 kOe to 11 K in 25 kOe. Below $H = 15 \text{ kOe}$, substantial thermomagnetic irreversibility among ZFC, FCC, and FCW curves below T_{f2} remains.

The location of the peak at T_{f1} (ZFC data) decreases from 42 K with $H = 1 \text{ kOe}$ to 39 K with $H = 15 \text{ kOe}$. The thermomagnetic irreversibility between FCW and FCC curves associated with T_{f1} becomes more obvious with the increasing applied field and is most evident when H is 15 kOe, where M_{FCC} is higher than M_{FCW} from 39 to 28 K (see inset in Fig. 2c). Below $H = 15 \text{ kOe}$, the transition at T_{f1} exhibits thermal hysteresis of 1 K between the heating and warming curves. With a further increase of the magnetic field, the transition at T_{f1} shifts downward to 34 K, smears out, and $M(T)$ displays only a small thermal irreversibility at $H = 25 \text{ kOe}$; it finally disappears when magnetic field reaches and exceeds 30 kOe.

In 30 and 50 kOe magnetic fields, a bifurcation between ZFC and FCC is clearly observed starting from 14 and 12 K, respectively. Apart from the bifurcation, the ZFC curves of 30 kOe magnetic fields also show a small peak around 8 K, which is related to the transition at T_{f2} . The inset of Fig. 2e shows the field dependence of the temperature of the peak at T_{f2} . Further, it appears that the anomaly at T_N may become a ferrimagnetic to paramagnetic transition at $H = 30$ and 50 kOe due to the high magnetic fields which can change the magnetic state of the compound from antiferromagnetic to ferrimagnetic.

To further ascertain the magnetic states of HoFe_4Ge_2 , Fig. 3a shows the real (χ') component and Fig. 3b shows the imaginary

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