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Electrical resistivity and thermodynamic properties of the ferromagnet Nd_2Pt_2In

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ABSTRACT

The Nd_2Pt_2In compound was investigated by means of electrical resistivity $\rho(T)$, heat capacity $C_p(T)$, magnetic susceptibility $\chi(T)$, magnetization $M(\mu_0 H)$ and magnetocaloric effect (MCE) measurements. The material orders ferromagnetically at $T_C = 16$ K with a second - order phase transition. In the ordered state, $\rho(T)$ can be represented in terms of ferromagnetic (FM) spin - wave dispersion with an energy gap $\Delta_R = 13(1)$ K in zero field. In concert, the $C_p(T)$ data in this region can be well described by the same model getting $\Delta_C = 8(1)$ K in zero field. Above T_C, the $\rho(T)$ variation is characteristic of electron - phonon interaction in the presence of $s - d$ scattering and crystalline-electric field (CEF). The $4f$ - electron specific heat shows a Schottky - type anomaly around 60 K associated with CEF. On the other hand, $C_p(T)$ data of the non-magnetic homologue La_2Pt_2In can be described by the Debye - Einstein model, giving a Debye and Einstein temperature values of 190.3(5) K and 69.8(7) K respectively. At high temperatures, the $\chi(T)$ data follows the Curie - Weiss relation with an effective magnetic moment $\mu_{eff} = 3.61(2)\ \mu_B$ and a Weiss temperature $\theta_p = 17(1)$ K. The magnitude of MCE was estimated from the isothermal magnetization data to be 6.25 J/(kg.K), 5.01 J/(kg.K), 3.18 J/(kg.K) and 0.47 J/(kg.K) for a field change of 7 T, 5 T, 3 T and 1 T, respectively. The characteristic behaviour of the isothermal magnetic entropy change points to a second - order character of the FM phase transition.

1. Introduction

Magnetic refrigeration technology based on the exploration of magnetocaloric effect (MCE) shows superior application prospects over conventional gas compression/expansion refrigeration due to its higher energy efficiency and environmental friendliness [1–3]. MCE is an intrinsic thermal response for the application or removal of a magnetic field to a magnetic material, which brings about isothermal magnetic entropy change (ΔS_M) and/or the adiabatic temperature change (ΔT_{ad}). In recent years, magnetic materials with giant MCE have been the subject of intensive experimental investigations due to their potential application in magneto refrigerant and eco - friendly cooling industry [4–6]. Among many possible magnetocalorics, rare - earth based alloys and oxides, which exhibit large reversible MCE and refrigeration capacity with negligible hysteresis have attracted much attention [6–17].

Numerous efforts have been devoted to the group of ternary intermetallic compounds RE_2T_2M (RE = f - electron element, T = d - electron metal and M = III group p - electron metalloid). The ternaries RE_2T_2M crystallize in the primitive tetragonal structure of the Mo_2FeB_2 - type that is an ordered derivative of the U_3Si_2 - type [18,19]. In Ce_2T_2In with T = Ni and Rh, an intermediate valence behaviour was observed, while the compounds with T = Cu, Pd and Au were found to order magnetically at low temperatures [18]. The Pd - containing phase is the only known ferromagnet within this series and one of very few ferromagnets amidst rare - earth based 2:2:1 materials [19,20]. In turn, no evidence of any magnetic ordering was observed for Ce_2Pt_2In down to 1.7 K [18].

In the series of rare - earth compounds RE_2Pt_2In, antiferromagnets and ferromagnets with ordering temperatures between 8

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and 40 K were reported [21]. Among them, Pr_2Pt_2In and Nd_2Pt_2In order ferromagnetically at $T_C = 9.5$ K and 17 K, respectively [21], while Tb_2Pt_2In and Ho_2Pt_2In order antiferromagnetically below $T_N = 40$ K and 8 K, respectively [21].

Recently, a series of the RE_2Cu_2M (M = Mg, Cd, Sn or In) compounds was intensively studies due to their unique physical properties [22–26]. In particular, large MCE was found for a few of them which order ferromagnetically at Curie temperature lower than 50 K [22,23,25].

In this work, we investigated in details the electrical transport, thermodynamic and magnetocaloric properties of ferromagnetic Nd_2Pt_2In.

2. Experimental details

Polycrystalline samples of Nd_2Pt_2In and La_2Pt_2In were synthesized in an arc - furnace by melting the appropriate amounts of constituents (Nd, La, Pt and In metals with the purity 99.9, 99.9, 99.95 and 99.999 wt.% respectively) under high purity argon atmosphere. The ingots were flipped over and remelted several times to promote good homogeneity. The weight loss after final melting was less than 1%. No further heat treatment was applied. Room temperature X-ray diffraction (XRD) experiments were performed on the powder samples using a Bruker D8 Advance diffractometer with CuKα1 radiation (λ = 1.540598Å). The XRD patterns were analyzed using the Cell And Intensity Least (CAIL) square refinement method from TOPAS ACADEMIC programme [27]. Both samples were free of any secondary phases or unreacted elements. Temperature dependent electrical resistivity measurements were performed from 1.9 to 300 K in external magnetic fields up to 5 T using a four - probe dc method implemented in a physical property measurement system (PPMS, Quantum Design). Heat capacity measurements were performed in the temperature interval 1.9–300 K and in magnetic fields up to 5 T using the same PPMS platform. The magnetic properties were studied from 1.72 to 400 K using a SQUID magnetometer (MPMS, Quantum Design). The isothermal magnetization was measured in the temperature interval 4–30 K in step of 2 K and in magnetic fields up to 7 T.

3. Results and discussion

3.1. X - ray diffraction

The XRD pattern refinements confirmed for both the compounds studied the tetragonal Mo_2FeB_2 - type structure with space group $P4/nmm - D_{4h}^5$ (No. 127). The obtained lattice parameters (a and c) and the unit cell volume (V) were $a = 7.7576(9)$ Å, $c = 3.8383(5)$ Å and $V = 230.99(8)$ Å^3 for Nd_2Pt_2In and $a = 7.8524(5)$ Å, $c = 3.9365(3)$ Å and $V = 242.72(4)$ Å^3 for La_2Pt_2In, in good agreement with the literature data [21]. The observed relation $V_{La_2Pt_2In} > V_{Ce_2Pt_2In} > V_{Pr_2Pt_2In} > V_{Nd_2Pt_2In}$ [18,21,28], conforms the lanthanide contraction between La and Nd.

3.2. Electrical resistivity

The temperature dependence of the electrical resistivity, $\rho(T)$, of Nd_2Pt_2In is presented in Fig. 1a. The compound shows metallic conductivity with some bending of the $\rho(T)$ curve characteristic of $s - d$ interband scattering and/or crystal-electric-field (CEF) effect. This behaviour is similar to $\rho(T)$ of the homologue compound Pr_2Pt_2In [28] and several other rare - earth intermetallics such as La_3NiGe_2 [29]. Generally for the Nd - bearing compounds, CEF is predominant in the electrical resistivity [30,31], and a simple analysis in terms of the Bloch - Grüneissen - Mott (BGM) relationship [32,33] is not possible. The bottom inset of Fig. 1a displays an expanded view of the low - temperature $\rho(T)$ data, which shows a sudden drop near $T_C = 16$ K, signaling an onset of ferromagnetically (FM) ordered state, as confirmed by the $C_p(T)$ and $\chi(T)$ results (see sections 3.3 and 3.4). The observed decrease of the electrical resistivity with decreasing temperature in the magnetically ordered state results from gradual reduction of scattering conduction electrons on magnetic moments. The magnetic phase transition in Nd_2Pt_2In manifests itself as a pronounced anomaly in the temperature derivative of the resistivity $d\rho/dT$ (see the upper inset of Fig. 1a). The value of $T_C = 16$ K was estimated according to the Sato criterion [34], which is at the midpoint of the anomaly in the $d\rho/dT(T)$ curve (note the arrow in the upper inset of Fig. 1a).

The low temperature $\rho(T)$ data of Nd_2Pt_2In measured in an external magnetic field of 0.5 and 5 T are shown in Fig. 1b and c, respectively. It is observed that T_C increases with increasing field as expected for ferromagnets. Below T_C, $\rho(T)$ is governed by scattering conduction electrons on FM spin - wave excitations with an energy gap (Δ_R) in the magnon spectrum, and can be described by formula [35]:

$$\rho(T) = \rho_0 + A\Delta_R T\left[1 + 2\frac{T}{\Delta_R}\right]\cdot\exp\left(-\frac{\Delta_R}{T}\right), \quad (1)$$

where ρ_0 represents the residual resistivity due to static imperfections and A is a prefactor. The values of the parameters resulting from the LSQ fits of Eq. (1) to the experimental data below T_C are listed in Table 1. The fits of $\rho(T)$ are shown by the solid lines in Fig. 1b and c and in the bottom inset of Fig. 1a.

3.3. Heat capacity

The temperature dependencies of the specific heat, $C_p(T)$, of Nd_2Pt_2In and the nonmagnetic reference compound La_2Pt_2In are displayed in Fig. 2. While the $C_p(T)$ curve of La_2Pt_2In is featureless down to 1.8 K, that of the Nd-based phase shows a λ-shaped anomaly at $T_C = 16$ K that corresponds to the phase transition from the paramagnetic state to the FM state one. This value of T_C is an agreement with that derived from the $\rho(T)$ data. At room temperature, the specific heat of La_2Pt_2In reaches the Dulong - Petit limit, $3NR = 124.7$ J/mol K ($N = 5$ is the number of atoms per formula unit, R stands for the gas constant). In the whole temperature range measured, the $C_p(T)$ data of La_2Pt_2In can be described by the Debye - Einstein model [36]:

$$C_p(T) = \gamma T + 9NR(1-d)\left(\frac{T}{\theta_D}\right)^3\int_0^{\theta_D/T}\frac{x^4e^x}{(e^x-1)^2}dx, + 3NRd\left(\frac{\Theta_E}{T}\right)^2 e^{\Theta_E/T}\left(e^{\Theta_E/T}-1\right)^2 \quad (2)$$

where d is a number of Einstein modes, the first term represents the electronic contribution with γ being the Sommerfeld coefficient and the second and third terms are the standard Debye and Einstein expressions for the phonon contribution with θ_D and Θ_E being the Debye and Einstein temperatures, respectively. The LSQ fit of Eq. (2) to the experimental data (note the red line in Fig. 2) yielded the parameters: $\gamma = 9(1)$ mJ/(mol.K^2), $\theta_D = 190.3(5)$ K, $\Theta_E = 69.8(7)$ K and $d = 0.2$. The value of θ_D is similar to those reported for isostructural members of the RE_2Pt_2In series [18,28]. The inset (b) of Fig. 2 displays the plot of Cp/T^3 versus temperature for La_2Pt_2In that exhibits a local maximum near T_{max} of about 14 K. From the