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Systematically study on the magnetism and critical behaviour of layered NdMn1.4Cu0.6Si2

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Abstract

The strong dependence of the field-induced on the body-centered tetragonal ThCr2Si2-type and the critical behaviour of magnetic phase transition in NdMn2-xCuxSi2based compounds guide us to study the substitution Mn (atomic radius = 1.35 Å) by Cu (atomic radius = 1.28 Å) in layered NdMn1.4Cu0.6Si2compound. Room temperature x-ray diffraction study indicates clearly that most of the reflections can be identified with ThCr2Si2-type structure with space group I4/mmm. It found lattice parameters a slightly increases and lattice parameter c decrease compare to NdMn2Si2as indicated the decreasing of volume structure. The Neel temperature TNis found at 340 K while Curie temperature TCfound at 75 K respectively. The increasing concentration of Cu in replacement of Mn changes the magnetic phase transition from first order type for NdMn2Si2to second order type for layered NdMn1.4Cu0.6Si2compound as been determined by particular the S-shaped nature of the Arrott plot near TC. Our results indicate that the magnetic-field-induced magnetic phase transition plays a critical role on producing large magnetocaloric effect in these systems especially on second order type as the key for further investigation. The critical behaviour analysis in the vicinity of TCdemonstrates that the magnetism of the layered NdMn1.4Cu0.6Si2compound is governed by long range interactions.

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Systematically Study on the Magnetism and Critical Behaviour of Layered NdMn$_{1.4}$Cu$_{0.6}$Si$_2$

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Abstract. The strong dependence of the field-induced on the body-centered tetragonal ThCr$_2$Si$_2$-type and the critical behaviour of magnetic phase transition in NdMn$_{2-x}$Cu$_x$Si$_2$ based compounds guide us to study the substitution Mn (atomic radius=1.35 Å) by Cu (atomic radius=1.28 Å) in layered NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound. Room temperature x-ray diffraction study indicates clearly that most of the reflections can be identified with ThCr$_2$Si$_2$-type structure with space group I4/mmm. It found lattice parameters a slightly increases and lattice parameter c decrease compare to NdMn$_2$Si$_2$ as indicated the decreasing of volume structure. The Neel temperature T$_N$ is found at 340 K while Curie temperature T$_C$ found at 75 K respectively. The increasing concentration of Cu in replacement of Mn changes the magnetic phase transition from first order type for NdMn$_2$Si$_2$ to second order type for layered NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound as been determined by particular the S-shaped nature of the Arrott plot near T$_C$. Our results indicate that the magnetic-field-induced magnetic phase transition plays a critical role on producing large magnetocaloric effect in these systems especially on second order type as the key for further investigation. The critical behaviour analysis in the vicinity of T$_C$ demonstrates that the magnetism of the layered NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound is governed by long range interactions.

INTRODUCTION

Research on near-room-temperature magnetic cooling which related to magnetocaloric effect (MCE) phenomena has been a favorable increase nowadays [1,2]. This MCE process, will supersede the conventional refrigeration technology based on gas operation which its special interest because of considerable socio-economic benefits and high efficiency on performance [3,4]. Material systems which use as refrigerant such as LaFeSi [5], NiMnGa [6], MnAsSb [7], CeMn$_2$Ge$_{2-x}$Si$_x$ [8] and GdSiGe [9] undergo first-order magnetic transitions and have been found to possess a giant MCE. However, issue which include large thermal and magnetic hysteresis that is detrimental to the refrigerant capacity, drive them undesirable for real applications. From this issue, layered NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound become to be significant interest, which indicated a second-order phase transition as well as no thermal and magnetic hysteresis behavior.

An investigation on substituting other transition metal (Cu) for Mn in NdMn$_2$Si$_2$ compound (NdMn$_{1.4}$Cu$_{0.6}$Si$_2$) has been implemented in this case study, and it was found that an appropriate Cu concentration successfully changed the magnetic phase transition type from first order to second order and Curie temperature into the temperature range of interest, leading to the attainment of a high contribution in order to produce giant magnetocaloric effect (GMCE). From this point of view, the replacement of Mn by Cu is expected to significantly modify the magnetic state of both

the Nd and the Mn sublattices due to the difference of magnetic moment and atomic radius of Mn and Cu. Thus, in an effort to understand the nature of the magnetic transition in $NdMn_{1.4}Cu_{0.6}Si_2$, critical exponent analysis in the vicinity of the ferromagnetic (FM) – paramagnetic (PM) region [10-12] has been performed. The outcomes revealed that this compound undergoes a second-order ferromagnetic (FM) - paramagnetic (PM) transition at ~ 75 K. Based on this ability to design the overall $NdMn_{2-x}Cu_xSi_2$ compounds, we present a detailed investigation of the crystal structure, magnetic phase transitions and critical behaviour in the layered $NdMn_{1.4}Cu_{0.6}Si_2$ compound.

EXPERIMENTAL AND PROCEDURES

The layered $NdMn_{1.4}Cu_{0.6}Si_2$ compound were prepared by arc melting the appropriate amounts of Nd (99.9%), Mn (99.999%), Cu (99.9%) and Si (99.9%) chips in an argon atmosphere. During arc melting, a 3% excess of Mn over the stoichiometric amount was added to compensate for the weight loss of Mn. The compounds were melted several times to achieve good homogeneity. The compound were then wrapped in tantalum foil, sealed in a quartz ampoule, and subsequently annealed at 900 °C for 120 h and then quenched in water at room temperature. The crystal structure of the samples were checked by room temperature powder x-ray diffraction (XRD) measurements of λ = 1.54059 Å using Cu Kα1 radiation with the diffraction patterns refined using the Fullprof software package [13]. The magnetization measurements were carried out using the vibrating sample magnetometer option of a Quantum Design 14 T physical properties measurement system (PPMS) and a Quantum Design magnetic properties measurement system (MPMS) in the temperature range of 100–340 K at applied fields of up to 5 T. For critical exponent study, magnetization isotherms were measured in the temperature range of 65–105 K with an interval of 3 K at applied fields of up to 5 T.

RESULT AND DISCUSSION

Crystal Structure

Confirmation that the layered $NdMn_{1.4}Cu_{0.6}Si_2$ compound crystallize in the expected $NdMn_{1.4}Cu_{0.6}Si_2$ type structure with space group I4/mmm was provided by analysis of the x-ray powder diffraction patterns as indicated in Fig. 1. Layered $NdMn_{1.4}Cu_{0.6}Si_2$ compound have Nd atoms in the 2a site (0, 0, 0), while Cu and Mn share the position at the 4 d site (0, 1/2, 1/4), and Si atoms occupy the 4e site (0, 0, z). The measured data from the diffraction patterns were analysed using the Rietveld refinement technique, and the distances between neighbouring atoms have been obtained with the BLOKJE program, using the structural and positional parameters. It found lattice parameters a slightly increases from 4.004 (5) Å to 4.036 (7) Å and lattice parameter c decrease from 10.527 (4) Å to 10.358(5) Å compare to $NdMn_2Si_2$ as indicated the decreasing of volume structure from 168.76 Å to 168.59 Å.
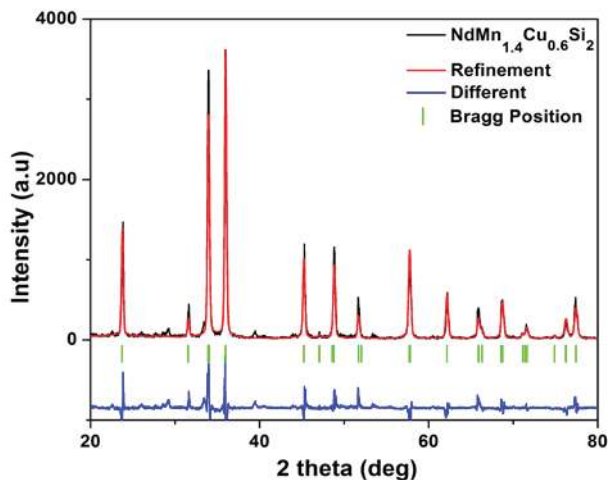

FIGURE 1. Room temperature x-ray diffraction   patterns of $NdMn_{1.4}Cu_{0.6}Si_2$

Magnetic Properties

The temperature dependences of the magnetization and the differential scanning calorimetry curve obtained at higher temperatures of $NdMn_{1.4}Cu_{0.6}Si_2$ is shown in Fig. 2. As is also evident from the analyses of the neutron diffraction data presented below, $NdMn_{1.4}Cu_{0.6}Si_2$ indicated two magnetic transitions over the temperature range of 10–450 K. The $T_N \sim 340$ K antiferromagnetic transition temperatures were determined from a graph of the DSC data plotted against inverse temperature with the $T_C \sim 75$ K ferromagnetic transition temperature determined from the maximum of the dM/dT versus T curve from the zero-field cooling magnetization data. Comparison of the cooling and warming magnetisation results in shows that the no thermal hysteresis at T_C.
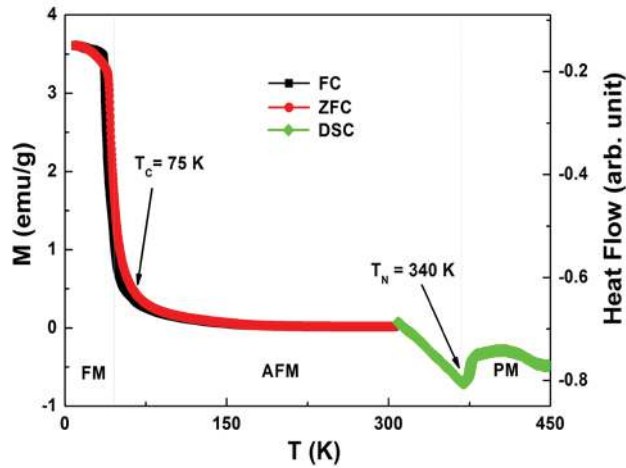
FIGURE 2. Temperature dependence of the magnetization as measured in a field of 0.01 T (left axis: zero field cooling (ZFC) and field cooling (FC)); right axis: DSC results over the range T=300–450 K)

Critical Behaviour

In an effort to further clarify the nature of the FM – PM phase transition, an analysis of the critical behaviour near T_C for $NdMn_{1.4}Cu_{0.6}Si_2$ compound was performed. The isothermal magnetization versus the applied field around T_C has been measured using intervals of 5 K and 3 K, as shown in Fig. 3.

FIGURE 3. Isothermal magnetization curves for $NdMn_{1.4}Cu_{0.6}Si_2$ compound in the vicinity of T_C

A plot of B/M versus M^2, known as the standard Arrott plot, is shown in Fig. 4 for the temperatures in the vicinity of T_C. According to the criterion proposed by Banerjee [14], the order of magnetic transition can be determined from the slope of the isotherm plot. If the B/M versus M^2 curves show a negative slope, the transition is first order, whi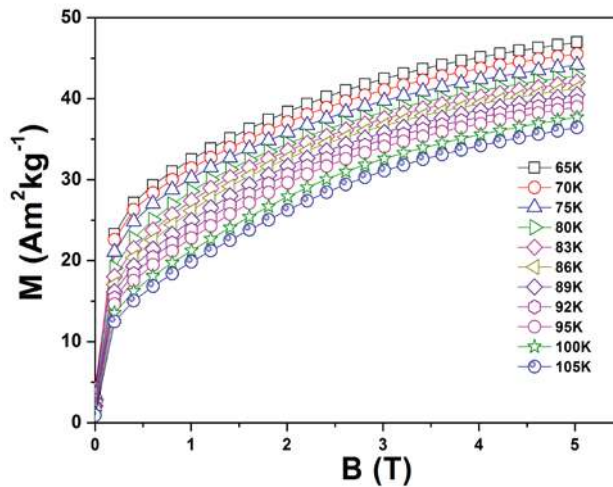le a positive slope corresponds to a second order transition. For the NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound, the always positive slope of the B/M, versus M^2 curves throughout the transition temperature range indicates that the phase transition is second order. This result is consistent with the absence of thermal and magnetic hysteresis in the vicinity of the FM – PM transition as discussed, thereby confirming the second order nature of this transition. The scaling hypothesis postulates that a second order magnetic phase transition near T_C is characterized by a set of critical exponents, namely, β, γ, and δ [15]. In this work, different methods have been used to investigate the critical behaviour of NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound, namely, modified Arrott plots (MAPs), the Kouvel-Fisher method, critical isotherm analysis, and the the Widom scaling relation. The first method used to calculate the critical exponents is the MAPs method, which is based on the Arrott-Noakes equation of state [15]. Quantitative fits are made to the Arrott plots using the following equations [16].

$$M_S(T) = \lim_{H \to 0}(M) = M_0\,(-\varepsilon)^\beta, \varepsilon < 0 \tag{1}$$

$$\chi_0^{-1}(T) = \lim_{H \to 0}(H/M) = (h_0/M_0)\varepsilon^\gamma, \varepsilon > 0 \tag{2}$$

where M_0 and h_0 are constants and ε (= $(T-T_C)/T_C$) is the reduced temperature. Initial values of β and γ are selected, and then a plot of $M^{1/\beta}$ versus $(B/M)^{1/\gamma}$ is obtained. The spontaneous magnetisation, M_S, is then determined from the intersection of the linearly extrapolated curve with the $M^{1/\beta}$ axis.
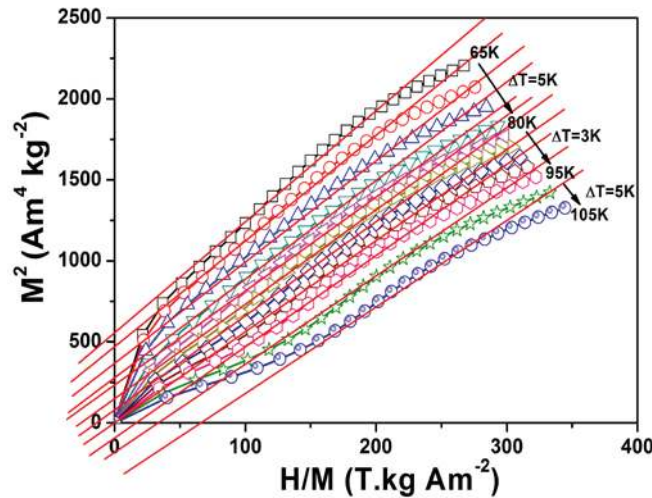

FIGURE 4. Arrott plots (M^2 vs. B/M) at temperatures in the vicinity of
T_C for NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound

It is imperative to note that only the high field linear region is used for the analysis because MAPs tend to deviate from linearity at low field due to the mutually misaligned magnetic domains [17]. Next, M_S is plotted as a function of temperature. To determine the inverse initial magnetic susceptibility, $\chi_0^{-1}(T)$, a similar procedure is used in conjunction with the $(B/M)^{1/\gamma}$ axis. The values of $\chi_0^{-1}(T)$ and M_S(T) are plotted as a function of temperature in Fig. 5 and these plots are then fitted with Equations (1) and (2), thus obtaining values of β and γ. These new critical exponent values are then used to construct new MAPs. These steps are repeated until the iterations converge to the optimum β, γ, and T_C values. Using Equations (1) and (2), the MAPs shown in Fig. 5 yielded critical parameters β = 0.42 and γ = 0.71.

The Kouvel-Fisher (KF) method, which makes use of Equations (3) and (4) shown below, is a more accurate way of determining the critical exponents β and γ [17].

$$\frac{M_S(T)}{\frac{dM_S(T)}{dT}} = \frac{T-T_C}{\beta} \tag{3}$$

$$\frac{\chi_0^{-1}(T)}{d\chi_0^{-1}(T)dT} = \frac{T-T_C}{\gamma} \tag{4}$$

According to Equations (3) and (4), plotting $M_S(T)[dM_S/dT]^{-1}$ and $\chi_0^{-1}(T)[d\chi_0^{-1}/dT]^{-1}$ versus temperature yields straight lines with slopes of $1/\beta$ and $1/\gamma$, respectively, as shown in Fig. 5. The value of T_C is obtained from the intercepts on the x-axis. The critical exponents β, γ, and δ, as well as T_C, obtained using the Kouvel-Fisher method are $\beta = 0.42$, $\gamma = 0.71$, $\delta = 2.70$, and $T_C = 75$ K. A comparison of the critical exponents' β and γ obtained using the MAPs and those obtained using the KF method reveals that these values match extremely well. The value of the critical component δ can be determined directly from the critical isotherm $M(T_C, H)$ according to Equation (5) below.

$$M_{T_C} = DH^{1/\delta}, \varepsilon = 0, T = T_C \tag{5}$$

Another way of obtaining the critical exponent δ is by using the Widom scaling relation shown in Equation (6):

$$\delta = 1 + \frac{\gamma}{\beta} \tag{6}$$

Using Equation (5) and the critical parameters β and γ obtained using the MAPs and those obtained using the Kouvel-Fisher method, the deduced δ values are 2.92 and 2.89, respectively. Thus, the Widom scaling relation has confirmed the reliability of the critical exponents deduced from the experimental data. The reliability of the calculated exponents β and γ can be confirmed by using the scaling theory.
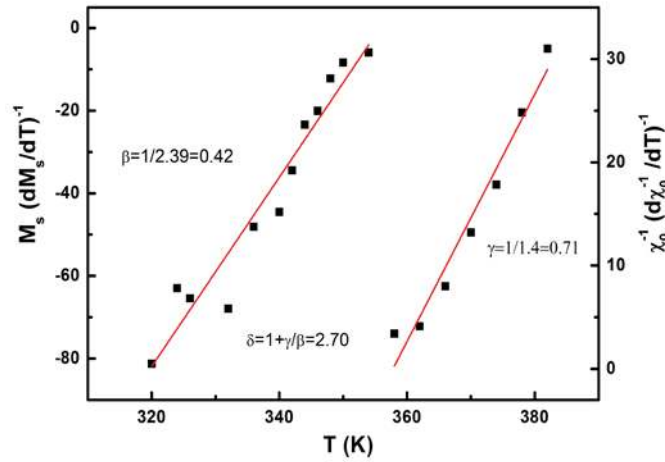

FIGURE 5. Kouvel-Fisher plot for the spontaneous magnetization M$_S$ and the inverse initial susceptibility $\chi_0^{-1}(T)$. (Solid lines are fitted to Equations (3) and (4))

In the critical region, according to the scaling theory, the magnetic equation of state can be written as:

$$M (H, \varepsilon) = \varepsilon^\beta f_\pm (H / \varepsilon^{\beta+\gamma}) \tag{7}$$

Where ε is the reduced temperature, $(T-T_C)/T_C$, and f_+ and f_- are regular analytical functions above and below T_C, respectively. Using β and γ obtained from the Kouvel-Fisher method, the plots of M/ε^β versus $H/\varepsilon^{(\beta+\gamma)}$ (shown in Fig. 6) yield two universal curves, one for temperatures above T_C and the other one for temperatures below T_C, in agreement with the scaling theory. This therefore confirms that the obtained values of the critical exponents as well as the T_C are reliable and in agreement with the scaling hypothesis.
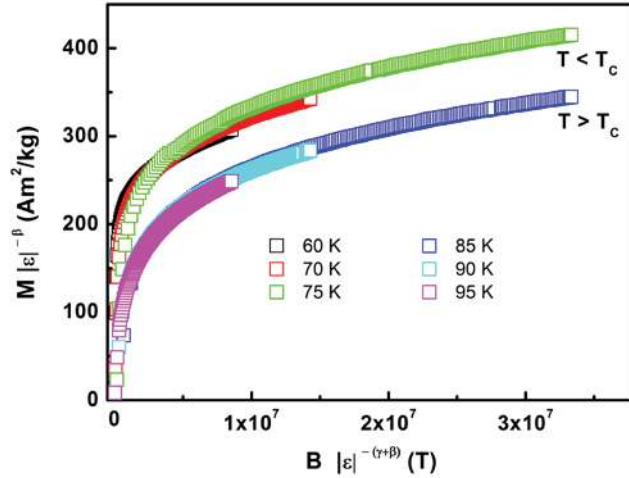
FIGURE 6. Scaling plots indicating universal curves below and above T_C for the NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound

It is well accepted that the order parameter of the phase transition around the magnetic transition temperature is fluctuating on all available length scales, and those fluctuations smear out the microscopic details of the interactions in the continuous phase transition system [18]. The mean-field interaction model for long range ordering has theoretical critical exponents of $\beta = 0.5$, $\gamma = 1.0$, and $\delta = 3.0$ [19], while the theoretical values based on the three dimensional Heisenberg model corresponding to short range interactions are $\beta = 0.365$, $\gamma = 1.386$, and $\delta = 4.80$ [20].

The δ, β, and γ values derived for the NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound are close to the mean-field values, thus indicating that long range interactions dominate the critical behaviour around T$_C$ in this compound. Thus, the critical behaviour analysis in the vicinity of T$_C$ demonstrates that the magnetism of the NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound is governed by long range interactions, which is in agreement with the linear fit to the data, which clearly also demonstrates that the relationship $\Delta S_M \propto (\mu_0 H/T_C)^{2/3}$ is valid around T_C.

CONCLUSION

An excellent agreement of the critical exponents of the NdMn$_{1.4}$Cu$_{0.6}$Si$_2$ compound was determined using the isothermal magnetization in the vicinity of T$_C$, based on various techniques such as the Kouvel-Fisher method, modified Arrott plots, and critical isotherm analysis. Moreover, all critical exponents fulfil the Widom scaling law. The validity of the calculated critical exponents was confirmed by the scaling equation with the obtained magnetization, field, and temperature data below and above T$_C$, showing a collapse into two different curves. Thus, the scaling of the magnetization data above and below T_C obtained using the respective critical exponents and the consistency in the values of the critical exponents determined by different methods confirm that our calculated exponents are unambiguous and intrinsic. The determined critical exponents are close to those predicted by the mean-field theory, with long range interactions.

ACKNOWLEDGEMENTS

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