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Critical Behavior Studies in the Vicinity of the Curie Point in the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ Alloy

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The main goal of this work is to study the critical behavior in the annealed $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy in the vicinity of the critical temperature T_C. The second order phase transition from the ferro- to paramagnetic state was confirmed by the positive slope of the Arrott plots. Critical exponents (β, γ, and δ) have been revealed using the Kouvel–Fisher method. Moreover, the Kouvel–Fisher analysis revealed the detailed Curie temperatures for all investigated samples.

topics: critical behavior, Curie temperature, critical exponents

1. Introduction

At this time, the magnetocaloric effect (MCE) is the cooling technique with the highest efficiency. Moreover, such technique is environmentally friendly, which is extremely important nowadays. MCE is observed as heating or cooling of magnetic material placed in an external magnetic field [1]. Relatively cheap magnetocaloric materials with relatively good magnetic properties are alloys of the $La(Fe,Si)_{13}$ type. This group of alloys bases on the face centered cubic $NaZn_{13}$type structure [2]. For many years, the $La(Fe,Si)_{13}$ type alloys have been modified by Co [3], Ni [4], Al [5], Ga [6] in order to tune the Curie temperature. The partial substitution of La by Ce [7], Pr [8], Nd [9], Er [10], Ho [11] or Dy [12] had an important role as it contributed to a change of the order of phase transition and change the magnetic entropy change (ΔS_M). For the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy, the magnetic entropy change was 14.6 J/(kg K) with the Curie point at 267 K and under the change of the external magnetic field up to $\sim$ 5 T. In the previous paper [12], the carried out Arrott plots and scaling analysis showed the presence of second order phase transition.

2. Experimental section

A detailed description of the sample preparation is provided in [12]. The thermomagnetic properties (T_C and ΔS_M) were investigated using a Quantum Design MPMS XL-5 system working in the temperature range (2–400 K) and in a magnetic field of up to 5 T. Critical exponents were investigated using the Kouvel–Fisher technique [13].

3. Results and discussion

The nature of the phase transition was preliminarily checked by constructing the Arrott plots shown in the previous paper [12]. The Arrott plots revealed a series of parallel lines. The construction was done using the critical exponents corresponding to the mean-field theory ($\beta = 0.5$, $\gamma = 1$). The positive slope of the $M^2 = f(H/M)$ isotherms confirmed the second order nature of the phase transition, according to the Banerjee criterion [14]. It was clearly visible that the results were nearly linear and linear extrapolation was done to known the values of spontaneous magnetization and inverse susceptibility.

The second order phase transition can be mathematically described by the system of critical exponents and the relations between them. These exponents β, γ and δ are strongly correlated to such parameters as: spontaneous magnetization M_S, initial susceptibility χ_0 or critical magnetization isotherm at the Curie temperature, respectively. The definitions of the exponents are given by following mathematical equations [13]

$$M_S(T) = M_0\,(-\varepsilon)^{\beta}, \quad \varepsilon < 0,\, T < T_C, \tag{1}$$

$$\chi_0(T)^{-1} = \left(\frac{H_0}{M_0}\right)\varepsilon^{\gamma}, \quad \varepsilon > 0,\, T > T_C, \tag{2}$$

$$M = D\,H^{1/\delta}, \quad \varepsilon = 0,\, T = T_C, \tag{3}$$

where $\varepsilon = (T - T_C)/T_C$ means the reduced temperature, M_0, H_0 and D are the critical amplitudes, H is the applied magnetic field, and M is the magnetization.

Linear extrapolation of high field regions of the M^2 vs (H/M) isotherms allowed to determine the spontaneous magnetization M_S and the inverse

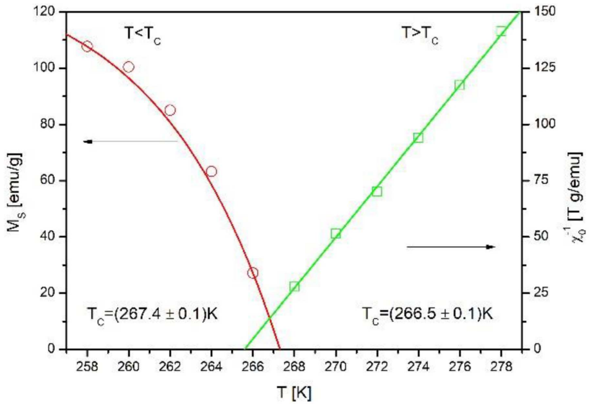

Fig. 1. The temperature dependence of spontaneous magnetization M_S and inverse initial susceptibility χ_0^{-1} of the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy.

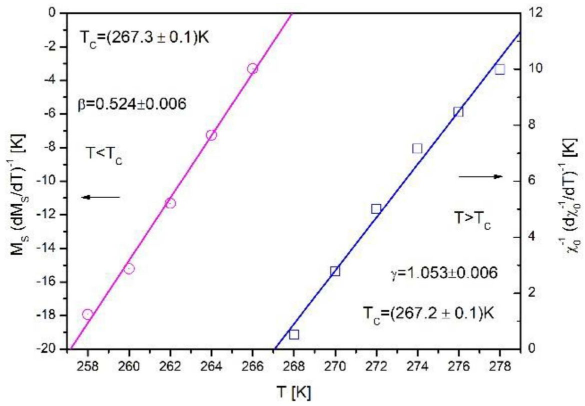

Fig. 2. Kouvel–Fisher plots for calculating the critical exponents β and γ for the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy.

initial susceptibility χ_0^{-1} from the intersections of the linear extrapolation with M^2 and (H/M) axes, respectively. The dependence M_S vs T and the dependence χ_0^{-1} vs T are depicted in Fig. 1.

The analysis of the temperature dependence of M_S and χ_0^{-1} allowed us to determine more detailed values of the Curie point. We considered two different approaches through the critical point: (i) from the ferromagnetic state (FM) one has $T_C = 267.4$ K (ii) and from the paramagnetic (PM) region T_C is 266.5 K. The revealed values are in agreement with value reported in [12].

A relatively simple way to determine the critical exponents was proposed by Kouvel and Fisher in [13], and today it is called the Kouvel–Fisher method. This method guarantees relatively high accuracy and is based on the relation (1) and (2), modified to the following form, respectively

$$\frac{M_S(T)}{\left[\frac{dM_S(T)}{dT}\right]} = \frac{T - T_C}{\beta}, \quad (4)$$

and

$$\frac{\chi_0^{-1}(T)}{\left[\frac{d\chi_0^{-1}(T)}{dT}\right]} = \frac{T - T_C}{\gamma}. \quad (5)$$

Taking into account (4) and (5), the critical exponents β and γ can be revealed directly from the slopes of the linear fitting for $M_S(T)/[dM_S(T)/dT]$ and $\chi_0^{-1}(T)/[d\chi_0^{-1}(T)/dT]$, respectively.

Now, (4) and (5) allowed to construct the Kouvel–Fisher plots depicted in Fig. 2. On the grounds of these plots, the Curie temperature was found based on the intersections of fitted lines with the temperature axis.

The dependency analysis in Fig. 2 revealed the values of β and γ, which were 0.524 and 1.053, respectively. Calculation of the critical exponent δ was proposed by Widom [14] using the scaling relation

$$\delta = 1 + \frac{\gamma}{\beta}. \quad (6)$$

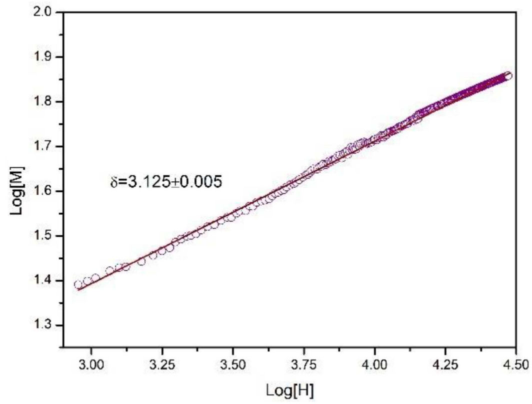

Fig. 3. The field dependence H of magnetization Mon a log–log scale collected at 266 K for the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy. The red line is the best linear fit according to (7).

The value of δ calculated using (6) is 3.01. Additionally, taking into account (3), it is possible to determine the δ exponent by the logarithm of this relation. After such operation, (3) can be written as

$$\ln(M) = \ln(D) + \frac{1}{\delta}\ln(H). \quad (7)$$

In the vicinity of the critical point (the Curie temperature), the exponent δ is hardly related to the M vs H curve. As mentioned above, the value of the Curie point, determined based on Kouvel–Fisher plots, was 267.3 K. The M vs H isotherms were collected in the range of temperatures from 258 to 278 K with temperature step 2 K. The M vs H curve collected at 266 K is relatively close to the Curie temperature. This curve on a log–log scale was shown in Fig. 3. The exponent δ was calculated as the inverse of the slope of the linear fit of $\ln(M) = f\big(\ln(H)\big)$. The value of δ is 3.125, which is slightly higher than this delivered by the Widom scaling relation.

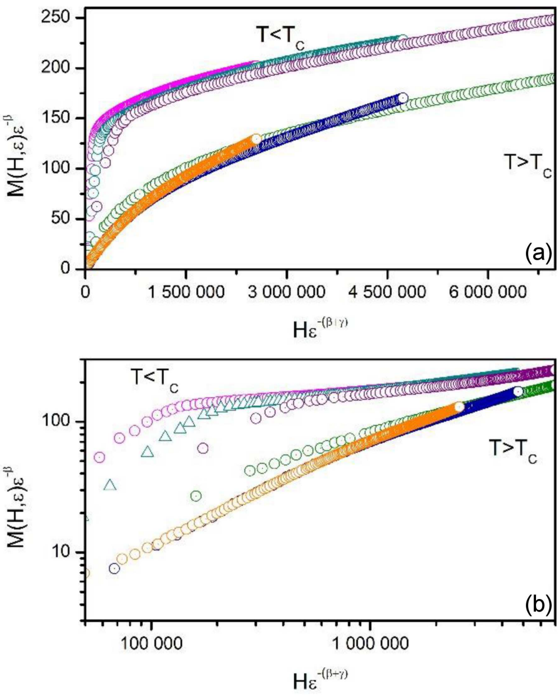

Fig. 4. Scaling plots revealed for the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy (a) and the same data on a log–log scale (b).

The validation of the calculated exponents was carried out using the following test based on the magnetic equation of state [23], i.e.,

$$M(H,\varepsilon) = \varepsilon^{\beta} f_{\pm}\left(\frac{H}{\varepsilon^{\beta+\gamma}}\right), \quad (8)$$

where $f_{\pm}$ are the regular functions; f_{+} for the paramagnetic ($T > T_C$) region and f_{-} for the ferromagnetic ($T < T_C$) region. Note that (8) describes the dependence $M(H,\varepsilon)\varepsilon^{-\beta}$ vs $H\varepsilon^{-(\beta+\gamma)}$, and two independent universal curves are generated. The first one is constructed for temperatures higher than T_C, and the second one for temperatures lower than T_C. The M vs H isotherms measured at temperatures higher or lower than the Curie point collapse into these two universal curves. The determined critical exponents were used to construct universal curves in the vicinity of the critical point T_C and plotted in Fig. 4a. For better visibility, the same curves were shown in Fig. 4b on a log–log scale.

Figure 4 shows that all data collapses onto two independent curves. The first universal scaling curve is constructed for temperatures lower than the Curie point, and the second for temperatures higher than T_C. Such behavior confirms validity of the determined values of the critical exponents and the Curie point.

The values of the revealed critical exponents are comparable to those reported in [15]. Our results indicate the presence of long-range ferromagnetic interactions, and the mean-field theory describes well produced material.

As it was shown in the previous paper [12], the magnetic entropy change increases with an increase of external magnetic field. Franco et al. [16] proposed the phenomenological formula describing the field dependence of ΔS_M according to

$$\Delta S_M = C\left(B_{\max}\right)^n, \quad (9)$$

where C is a constant depending on temperature and n is the exponent related to the magnetic state of specimen. The analysis carried out for the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy revealed the value of n equals 0.67. This value is the same as theoretical value. Taking into account the critical exponents calculated in this paper, the value of the exponent n was equal to 0.64 and 0.65 for the critical exponents, which were revealed using the Widom scaling relation and the linear fitting from (7), respectively. Importantly, the determined values are close to the theoretical value.

4. Conclusions

In the present paper, the critical behavior of the $LaFe_{11.0}Co_{0.7}Si_{1.3}$ alloy was studied in the vicinity of the Curie point. The Kouvel–Fisher method was used in order to determine the values of the critical exponents and the more precise value of the Curie point. The calculated values of the β, γ, δ exponents are reliable and reasonable, which was proved by the construction of the scaling plots based on the M–H isotherms. Such modified isotherms collapsed into two independent universal curves higher and lower than the Curie temperature. The calculated critical exponent values indicate long-range ferromagnetic interactions described by the mean-field theory, which is typical for this type of alloys. The calculated values of the exponent n correspond well with theoretical value.

References

[1] A.M. Tishin, Y.I. Spichkin, *The Magnetocaloric Effect and Its Applications*, Institute of Physics Series in Condensed Matter Physics, CRC Press, Boca Raton 2003.

[2] P.I. Kripyakevich, O.S. Zarechnyuk, E.I. Gladyshevsky, O.I. Bodak, *Z. Anorg. Chem.* **358**, 90 (1968).

[3] P. Gębara, P. Pawlik, *J. Magn. Magn. Mater.* **442**, 145 (2017).

[4] P. Gębara, P. Pawlik, M. Hasiak, *J. Magn. Magn. Mater.* **422**, 61 (2017).

[5] P. Gębara, P. Pawlik, B. Michalski, J.J. Wysłocki, *Acta Phys. Pol. A* **127**, 576 (2015).

[6] P. Gębara, P. Pawlik, B. Michalski, J.J. Wysłocki, K. Kotynia, *Acta Phys. Pol. A* **128**, 87 (2015).

[7] P. Gębara, J. Marcin, I. Skorvanek, *J. Electron. Mater.* **46(11)**, 6518 (2017).

[8] J. Shen, Q.Y. Dong, Y.X. Li, J.R. Sun, *J. Alloys Compd.* **458**, 115 (2008).

[9] D.T. Kim Anh, N.P. Thuy, N.H. Duc, T.T. Nhien, N.V. Nong, *J. Magn. Magn. Mater.* **262**, 427 (2003).

[10] M. Balli, D. Fruchart, G. Gignoux, M. Rosca, S. Miraglia, *J. Magn. Magn. Mater.* **313**, 43 (2007).

[11] P. Gębara, J. Kovac, *Mater. Des.* **129**, 111 (2017).

[12] P. Gębara, J. Kovac, *J. Magn. Magn. Mater.* **454**, 298 (2018).

[13] J.S. Kouvel, M.E. Fisher, *Phys. Rev.* **136**, A1626 (1964).

[14] S.K. Banerjee, *Phys. Lett.* **12**, 16 (1964).

[15] Z. Shuotong, L. Yi, G. Qi, Z. Kewei, C. Fenghua, H. Jifan, *J. Magn. Magn. Mater.* **547**, 168932 (2022).

[16] V. Franco, J.S. Blasquez, A. Conde, *Appl. Phys. Lett.* **89**, 222512 (2006).