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Investigations of the Magnetic Phase Transition in the $\mathbf{LaFe_{11.14}Co_{0.66}Si_{1.1}M_{0.1}}$ (Where M = Al or Ga) Alloys

P. Gębara[a,b,*]

[a]Institute of Physics, Częstochowa University of Technology, Armii Krajowej 19, 42-200 Częstochowa, Poland

[b]Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47, 040 01 Košice, Slovakia

The aim of the present work was to study the phase transition in the $LaFe_{11.14}Co_{0.66}Si_{1.1}M_{0.1}$ (where M = Al or Ga) alloys. Research was carried out using field dependences of magnetization measured at a wide temperature range. Positive slope of the Arrott plots showed that magnetic phase transition in both investigated samples was of second order nature. The temperature dependences of the Landau coefficients also revealed second order phase transition in both specimens. The analysis carried out using universal curve confirmed second nature of phase transition in both samples.

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1. Introduction

An interesting alternative for conventional refrigeration is well known magnetocaloric effect (MCE), discovered by Warburg [1]. The MCE is defined as a cooling of magnetic material under external magnetic field. The efficiency of magnetic cooling process equals almost 60% and it is the highest value in the any refrigeration techniques [2]. The ideal magnetocaloric material (MCM) is characterized by high adiabatic change of temperature ΔT_{ad}, high isothermal magnetic entropy change ΔS_M, small hysteresis losses, low thermal hysteresis and low specific heat [3]. These conditions were formulated in the decade after the discovery of giant magnetocaloric effect (GMCE) in the $Gd_5Si_2Ge_2$ alloy by Pecharsky and Gschneidner Jr. in 1997 [4]. The GMCE, observed in this compound, is the result of two effects: second order phase transition from ferro-to-paramagnetic state and structural transformation in vicinity of the Curie temperature. However, high Gd content in the alloy composition generates its high price. According to that, in many laboratories all over the world, explorations of cheaper MCMs have been started. Interesting results have been observed in shape memory Ni–Mn–(In,Ga) based Heusler alloys [5].

Besides the listed compounds, Mn- [6], Co- [7], Gd- [8, 9] and Fe-based [10–12] alloys have been intensively studied. The Fe-based alloys are relatively cheap compared to the above mentioned materials and are characterized by excellent magnetic properties [13–15]. In this group of alloys, GMCE was observed in the $La(Fe,Si)_{13}$ compounds [16–18]. Large magnetic entropy change is caused by metamagnetic first order phase transition in the vicinity of the Curie temperature [16]. The magnetic phase transition is accompanied by a negative expansion of the unit cell of the $La(Fe,Si)_{13}$-type phase in the vicinity of the Curie point [16]. The $La(Fe,Si)_{13}$-type phase crystallizes in the fcc $NaZn_{13}$-type unit cell (space group $Fm3c$). As it was shown in [19–21] elements such as Gd [19], Co [20] or Al [21] cause weakening of itinerant electron metamagnetic transition. It results in decrease of the magnetic entropy change $|\Delta S_M|$ in the $La(Fe,Si)_{13}$-type alloys. Kumar et al. in [19] have shown the analysis of the phase transition nature using the Landau theory. In my previous works [22, 23] the magnetocaloric effect of the $LaFe_{11.14}Co_{0.66}Si_{1.1}M_{0.1}$ (M = Al, Ga) was studied. For sample doped by Ga, the calculated value of $|\Delta S_M|$ is about –6 J/(kg K) under the change of external magnetic field $\mu_0 \Delta H = 2$ T. In the $LaFe_{11.0}Co_{0.8}Si_{1.2}$ alloy [24] a similar value of $|\Delta S_M|$ was a result of first-order phase transition. According to that, the aim of present paper was to study phase transition in the $LaFe_{11.14}Co_{0.66}Si_{1.1}M_{0.1}$ (M = Al, Ga) alloys.

2. Experimental method

A detailed description of preparation, structural and magnetic measurements of the samples are given in [22, 23]. Phase transitions were investigated using Landau theory and based on field dependences of magnetization collected at different temperatures. In order to confirm the analysis of the Landau coefficients, the scaling method based on the temperature dependences of the magnetic entropy change was used.

3. Results and discussion

The field dependences of magnetization (M vs. $\mu_0 H$) measured at a wide temperature range for both samples were used to construct the Arrott plots. The Arrott plots constructed in the vicinity of the phase transition ($M^2 = f(\mu_0 H/M)$) are shown in Fig. 1. The positive slope of the Arrott plots suggests occurrence of second-order phase transition near the Curie temperature, according to the Banerjee criterion [25].

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*corresponding author; e-mail: pgebara@wip.pcz.pl

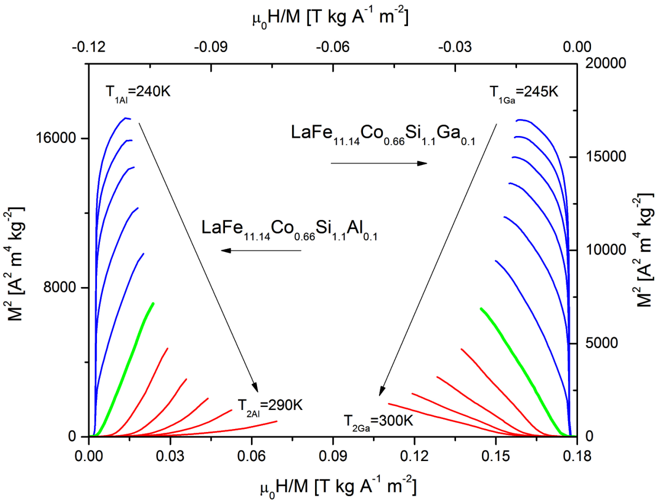

Fig. 1. Arrott plots constructed for sample doped by Al (left side) and Ga (right side).

As it was shown in [23, 26], the Arrott plots, in some cases, do not deliver correct information about the order of the phase transition. According to that, more detailed study of the phase transition was carried out using the Landau theory.

The fundamentals of the Landau theory of phase transition are based on the free energy $F(M,T)$ expanded in powers of magnetization M:

$$F(M,T) = \frac{c_1(T)}{2}M^2 + \frac{c_2(T)}{4}M^4 + \frac{c_3(T)}{6}M^6 + \ldots - \mu_0 HM, \qquad (1)$$

where $F(M,T)$ — free energy, c_1, c_2, c_3 — the Landau cofficients, M — magnetization, μ_0 — magnetic permability, H — magnetic field.

The order of the phase transition is determined by the sign of the c_2 coefficient determined at the Curie temperature. The negative sign of $c_2(T_C)$ corresponds to first-order phase transition, otherwise to second-order phase transition. Basing this on the condition of equlibrium $\delta F/\delta M = 0$, Eq. (1) has been rewritten as

$$\mu_0 H = c_1(T)M^1 + c_2(T)M^3 + c_3(T)M^5. \qquad (2)$$

This equation has been used as a model of function and experimental data have been approximated as such. The temperature dependences of the Landau coefficients ($c_1(T)$ and $c_2(T)$) are shown in Fig. 2a and b for sample doped by Al and Ga, respectively. The c_1 vs. T dependences have minimum at the Curie temperature for both studied specimens, which is typical for this coefficient. In case of the sign of $c_2(T_C)$, it is positive for both specimens, which confirmed second-order nature of the phase transition observed in investigated alloys.

An interesting scaling method was proposed by Franco et al. in [27] as a simple technique to identification of phase transition nature. It bases on the ΔS_M vs. T curves determined for the different magnetic fields. Each curve is rescaled in the following way: $\Delta S_M \to \Delta S_M/\Delta S_M^{pk}$ and $T \to \Theta_1$.

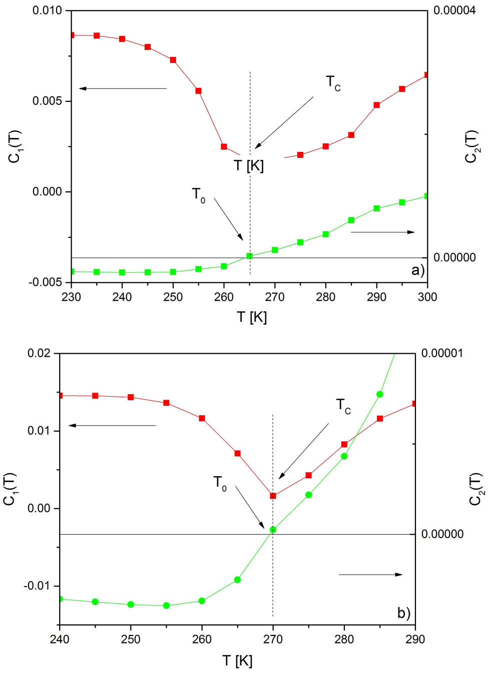

Fig. 2. Temperature dependence of the Landau coefficients calculated for sample doped by Al (a) and Ga (b).

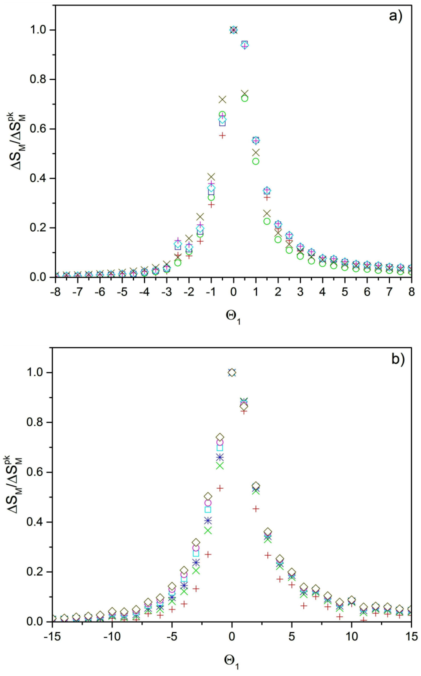

Fig. 3. Phenomenological universal curve determined for sample doped by Al (a) and Ga (b).

Temperature scaling was carried out using relation [27]:

$$\Theta_1 = (T - T_C)/(T_r - T_C), \qquad (3)$$

where T_C — the Curie point or peak temperature, T_r — reference temperature.

In present work, T_C value corresponds to ΔS_M^{pk} and T_r was selected for $\Delta S_M(T_r) = 0.5\Delta S_M^{pk}$.

As can be seen in Fig. 3, the phenomenological universal curves corresponding to the experimental data for both samples are shown.

The collapse of the universal curve for both sample is observed. This behavior suggests the occurrence of second order phase transition in both investigated samples. It confirms the results delivered by analysis of the Landau coefficients.

These results are in good agreement with previous predictions contained in [22, 23], which were based on the symmetrical shapes of the temperature dependences of magnetic entropy change.

4. Conclusions

In the present paper, the phase transition nature in the $\mathrm{LaFe_{11.14}Co_{0.66}Si_{1.1}M_{0.1}}$ alloy doped by Al or Ga was studied. Preliminary analysis based on the Arrott plots suggested second-order phase transition due to their positive slope. More detailed studies of phase transition order were carried out using the Landau theory. Studies of the temperature dependences of the c_2 Landau coefficient revealed its positive value for both samples and confirmed the occurrence of the second-order phase transition in both investigated samples. Results delivered by the Landau theory are consistent with those provided by construction of universal scaling curve based on temperature dependence of the magnetic entropy changes. In both cases rescaling of the $|\Delta S_M|$ vs. T curves revealed collapse in one universal curve and it is typical behavior for second order phase transition. Second order nature of phase transition in investigated samples is caused by weakening of itinerant electron metamagnetism observed in this type of alloys.

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