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First order phase transitions may occur in several magnetic systems, with two structural phases having different magnetic properties each and a structural transition between them. Here, a novel physics based phenomenological model of such systems is proposed, in which magnetization is represented by the volumetric amounts of ferromagnetism (described by extended Jiles-Atherton theory) and paramagnetism (described by the Curie-Weiss law) in respective phases. An identification procedure to extract material parameters from experimental data is proposed. The proposed phenomenological approach was successfully applied to magnetocaloric $Gd_5(Si_xGe_{1-x})_4$ system and also has the potential to describe the behavior of Griffiths phase magnetic systems. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4875678]

I. INTRODUCTION

Magnetic systems having more than one magnetic phases or materials have received a lot of attention recently. Their magnetic response to external influence, such as magnetic field, temperature, stress, etc., is often complex and, in some cases, accompanied with other interesting physical properties which in turn can be utilized in various devices.

Multilayered structures consisting of layers having different magnetic properties are relatively general example of such a magnetic system. The layers in such structures can have various magnetic properties, either being ferromagnetic, paramagnetic, or nonmagnetic at all. The specially chosen physical dimensions can add extra complexity, such as antiferromagnetic coupling between the layers, leading to unusual magnetic response. The situation can be complicated further if one of the layers has ability to change its magnetic state/nature while being subject to an external influence. In majority cases, phenomenological description of such systems does not exist.

A simple magnetic system for which the phenomenological description does exist is a material with two-magnetic-phase behavior, i.e., a material that exhibits two magnetic phases (soft and hard) in one hysteresis cycle. Such behavior can be induced in certain materials by special thermal treatment[1–3] or by stress.[4,5] In the latter case, for example, electrical steels under the influence of compressive stress are known to exhibit two distinct magnetic phases appearing in one hysteresis cycle: magnetic phases evolve due to 90 domain wall motion at lower magnetic fields and 180 domain wall motion at higher fields, with the transition occurring at a critical field over a transition region. An extended Jiles-Atherton model was proposed to successfully describe such behavior.[5,6]

A more complex example is Griffiths phase in the material, i.e., in the state where coexistence of ferromagnetic and paramagnetic phases occur. Above some specific temperature, the material is in fully paramagnetic state. If the temperature decreases below this temperature, the material will split into volumes, some of which are in ferromagnetic and the others are in paramagnetic states. Further decrease of the temperature will convert all the volume of the material into ferromagnetic state. The transition from fully paramagnetic to fully ferromagnetic phases is gradual and is a function of temperature only.[7,8]

Finally, one of the most complex examples is the system with magnetic-structural (MS) first order phase transition (FOPT). In the absence of applied magnetic field, the material exhibits a transition from one structural phase to another structural phase as a first order phase transition. Due to the fact that these two different *structural* phases have different *magnetic* phases as well (ferromagnetic vs. paramagnetic), the transition happens at different temperatures whether the temperature is increasing or decreasing, therefore, exhibiting hysteresis. The opposite case happens as well, magnetic field can induce the MS transition due to presence of the *magnetic* phases. Magnetocaloric material $Gd_5(Si_xGe_{1-x})_4$ in the region ($x \leq 0.503$) is an example of such a system. Upon temperature variation, an MS FOPT is induced from high temperature monoclinic paramagnetic phase to low temperature orthorhombic ferromagnetic phase.[9,10] At higher temperatures above the transition temperature this FOPT can be induced by application of magnetic field exceeding a specific value which is a function of temperature T. The transition from one structural phase to the other is gradual but is a function of the magnetic state in this case.

In order to describe the magnetic response for systems with MS FOPT, we extend the widely applicable phenomenological Jiles-Atherton model[11] with temperature dependence, where Curie temperature was added as an extra model parameter.[12,13] More specifically, we will refer mainly to the magnetocaloric material $Gd_5(Si_xGe_{1-x})_4$ in the region ($0.4 \leq x \leq 0.503$); however, one can easily see how the

proposed approach can easily be applicable to other special systems such as systems with the Griffiths phase or other simpler systems.

II. EXTENDED JA MODEL TO DESCRIBE MS FOPT SYSTEMS

A. Overview of the physical processes in the system

Before discussing the model equations and the terms in it, let us discuss in more detail what physical processes and what critical temperatures exist in the system.

First of all, there is a transition temperature, $T_{1stORDER}$, at which an MS FOPT from monoclinic phase to orthorhombic phase occurs in the absence of magnetic field.

For temperatures below the transition temperature $T < T_{1stORDER}$, orthorhombic structural phase is ferromagnetic and, therefore, it must have its own magnetic second order phase transition temperature $T_{2ndORTHO}$, and $T_{1stORDER} < T_{2ndORTHO}$.

For temperatures above the transition temperature $T > T_{1stORDER}$, monoclinic structural phase is paramagnetic. It is reasonable to assume that at some lower temperature, $T_{2ndMONO}$, magnetic second order phase transition occurs, and $T_{2ndMONO} < T_{1stORDER}$.

The situation is, of course, complicated as neither $T_{2ndMONO}$ nor $T_{2ndORTHO}$ can be observed experimentally as the structural phase transition occurs at $T_{1stORDER}$ which is measurable. However, in order to describe correctly the magnetic response of the system we assume for now that we know $T_{2ndMONO}$, $T_{2ndORTHO}$, and $T_{1stORDER}$ temperatures. We will discuss this further in Sec. II F where identification procedures for all model parameters will be suggested.

An important property of such systems that is utilized in magnetocaloric devices is that at higher temperatures $T > T_{1stORDER}$, when the material is in its monoclinic phase, the structural transition is induced if sufficiently high magnetic field is applied. The physical reason for this induced transition to occur can be found from energy considerations: the orthorhombic structural phase, which is ferromagnetic at that temperature, is energetically favorable than the monoclinic structural phase which is paramagnetic already.[10] The higher the temperature, the higher magnetic field one needs to apply.[14]

Of course, it is reasonable to assume that as the temperature is increased above $T > T_{2ndORTHO}$, there will be not enough magnetic energy to facilitate the structural transition as orthorhombic structural phase will be in its paramagnetic state already. Therefore, the system will remain in its monoclinic structural phase.

Now the necessary features that the model needs to be able to describe can be laid down with respect to temperature as shown here:

$T < T_{1stORDER}$—Ferromagnetic behavior of the orthorhombic phase with $T_{2ndORTHO}$;

$T_{1stORDER} < T < T_{2ndORTHO}$—Mixture of structural/magnetic phases;

$T > T_{2ndORTHO}$—Paramagnetic behavior of the monoclinic phase with $T_{2ndMONO}$.

B. Model equation

In order to describe dependence of magnetization on magnetic field and temperature for the MS FOPT systems, we assume that the total magnetization $M(H, T)$ can be represented as a sum of magnetizations coming from monoclinic and orthorhombic phases weighted by the corresponding volumetric amounts of the respective phases

$$M(H, T) = N_{MONO} \cdot M_{MONO}(H, T) + (1 - N_{MONO}) \cdot M_{ORTHO}(H, T), \quad (1)$$

where $M_{MONO}(H, T)$ is magnetization of the monoclinic phase, $M_{ORTHO}(H, T)$ is magnetization of the orthorhombic phase and N_{MONO} is volumetric amount of the monoclinic phase. In this model we assume that the magnetic interaction between volumes is either neglected or is already expressed in a nonlinear function of N_{MONO}.

C. Ferromagnetic phase

Ferromagnetic behavior of the orthorhombic phase can be described by the extended Jiles-Atherton model.[12,13] According to the classical Jiles-Atherton model, the magnetization is split into irreversible and reversible components and energetic considerations will lead to the following differential equation:[11]

$$M_{ORTHO} = M_{an} - k \; \delta \left(\frac{\partial M_{ORTHO}}{\partial H_e} - c \; \frac{\partial M_{an}}{\partial H_e} \right), \quad (2)$$

where M_{an} is anhysteretic function of the effective field $H_e = H + \alpha M_{ORTHO}$ and can take various forms based on the anisotropy of the material,[15] M_S is the spontaneous magnetization, k is pinning, a is domain density, c is reversibility, α is domain coupling, and $\delta = \pm 1$ depending whether magnetic field H is increasing or decreasing. Thermal effects can be incorporated into the classical Jiles-Atherton model by expressing these hysteresis parameters as functions of temperature with the Curie temperature, $T_{2ndORTHO}$ and two critical exponents as extra parameters.[12,13]

D. Paramagnetic phase

Paramagnetic behavior of the monoclinic phase can be described by the standard linear relationship between magnetization and magnetic field, and the Curie-Weiss law can be used to introduce temperature dependence[16,17]

$$M_{MONO}(H, T) = \frac{C}{T - T_{2ndMONO}} H, \quad (3)$$

where the parameter C is proportional to the Curie constant and $T_{2ndMONO}$ is the Curie temperature of the monoclinic phase.

E. Amount of the monoclinic phase

In order for an MS FOPT from monoclinic phase to orthorhombic phase to happen above the transition temperature, $T_{1stORDER}$ the system requires external magnetic energy which should be higher than the thermal energy at a given

temperature.[10,14,18] The comparison of the thermal energy and magnetic energy defines the critical magnetization, M_{CrPM}, and the critical applied magnetic field, H_{CrPM}, at which the transition starts to happen

$$\frac{\mu_0 M_{CrPM} H_{CrPM}}{\frac{3}{2} k_B (T - T_{1stORDER})} = 1, \tag{4}$$

where $\mu_0 = 4\pi \cdot 10^{-7}$ (H·m^{-1}) is permeability of vacuum and $k_B = 1.3806488 \cdot 10^{-23}$ (m^2 kg s^{-2} K^{-1}) is Boltzmann constant. At a first approximation, the critical magnetization can be found from the paramagnetic phase magnetization in Eq. (3)

$$M_{CrPM} = \frac{C}{T - T_{2ndMONO}} H_{CrPM}. \tag{5}$$

Equations (4) and (5) determine the start of the transition under any experimental conditions, such as fixing one control input (magnetic field or temperature) and varying the other one (temperature or magnetic field, respectively).

At a first approximation, it is assumed that the end of the transition occurs when the magnetic energy is bigger that the thermal energy at a given temperature by a certain constant value, R

$$\frac{\mu_0 M_{CrFM} H_{CrFM}}{\frac{3}{2} k_B (T - T_{1stORDER})} = R. \tag{6}$$

The second equation, which relates M_{CrFM}, H_{CrFM}, and T, can be taken in a form of anhysteretic function, for example,

$$M_{CrFM} = M_S \left[\coth\left(\frac{H_{CrFM} + \alpha M_{CrFM}}{a} \right) - \frac{a}{H_{CrFM} + \alpha M_{CrFM}} \right], \tag{7}$$

where M_S, a, and α are functions of temperature with $T_{2ndORTHO}$ and two critical exponents as extra parameters.[12,13] Similarly to Eqs. (4) and (5), Eqs. (6) and (7) determine the end of the transition under any experimental conditions and it is possible to see that if temperature exceeds $T_{2ndORTHO}$, there will be no transition at all.

Finally, knowing the magnetization values at the start and at the end of the transition the amount of the monoclinic phase N_{MONO} is a function of current magnetization and varies between 1, i.e., the system is fully in monoclinic phase (paramagnetic phase), and 0, i.e., the system is still fully in orthorhombic phase (ferromagnetic phase). It can be expressed as, for example,

$$N_{MONO} = \left(\frac{|M| - M_{CrFM}}{M_{CrPM} - M_{CrFM}} \right)^{\Delta M}, \tag{8}$$

where ΔM is the parameter responsible for the transition region which could be sharp or broad depending on the quality of the material and its composition.

F. General identification procedure

General Identification Procedure starts with estimation of the transition temperature, $T_{1stORDER}$, at which an MS FOPT from monoclinic phase to orthorhombic phase occurs in the absence of magnetic field. This is estimated from the corresponding experiment.

The way to estimate the "non-existent" second order magnetic phase transition temperature of the orthorhombic phase, $T_{2ndORTHO}$, was proposed recently,[8] where the Arrott Plot technique was applied to the ferromagnetic portions of the magnetic moment vs. magnetic field isotherms.

The same isotherms can be used to estimate the "non-existent" second order magnetic phase transition temperature of the monoclinic phase, $T_{2ndMONO}$, using the same modified Arrott plot[19] or by the Curie-Weiss law[20] as was applied to the paramagnetic response of the isotherms and the parameter C and $T_{2ndMONO}$ were identified in this paper.

One of these isotherms can also be used to identify the parameters ΔM and R numerically, assuming that they are not functions of temperature. The magnetization values at the start and at the end of the transition the amount of the monoclinic phase can easily be found from Eqs. (4)–(7).

Finally, the procedure to identify the parameters of the Jiles-Atherton model (M_S, k, a, α, c) and the corresponding parameters describing their temperature dependence was established earlier[12,13] and is applicable for the measurements made at temperatures below the first order phase transition temperature, i.e., $T < T_{1stORDER}$.

The proposed procedure completes the model expressed by Eqs. (1)–(8) to describe the magnetic response as a function of magnetic field and temperature for systems with MS FOPT.

III. EXPERIMENTAL VERIFICATION AND DISCUSSION

As an example, we applied our model to $Gd_5(Si_xGe_{1-x})_4$ system, which exhibits MS FOPT. Single crystal samples of $Gd_5Si_{1.95}Ge_{2.05}$ (x = 0.49) were prepared by Tri-arc pulling method using ultra pure 99.996% pure gadolinium (weight basis), 99.9999% pure silicon (weight basis), and 99.999% germanium (weight basis). The samples were then annealed at 2000 °C for 1 h to remove the residual secondary phases in the material. Single crystal samples were indexed by Laue backscattered diffraction method. Sample preparation is as discussed in depth previously.[18] Magnetic measurements were carried out in superconducting quantum interference device (SQUID) Quantum Design magnetometer with magnetic field strengths up to 5 T.

The measured magnetization dependences under different protocols of varying magnetic field and temperature are shown in Figs. 1(a) and 1(c). These data were used to extract parameters of the model as discussed in the Sec. II F, with the main parameters being $T_{2ndMONO} = 233$ K (estimated[20]), $T_{2ndORTHO} = 296$ K (estimated[10]), and $T_{1stORDER} = 263$ K (measured). The modeled curves in Figs. 1(b) and 1(d) show good agreement with the experimental data in the wide range of fields and temperatures $T_{1stORDER} < T < T_{2ndORTHO}$, where mixture of structural/magnetic phases exists.

The presented model is also able to describe correctly the magnetization dependences under different protocols of varying magnetic field and temperature, including change of hysteresis in magnetization curves occurring with cycling for
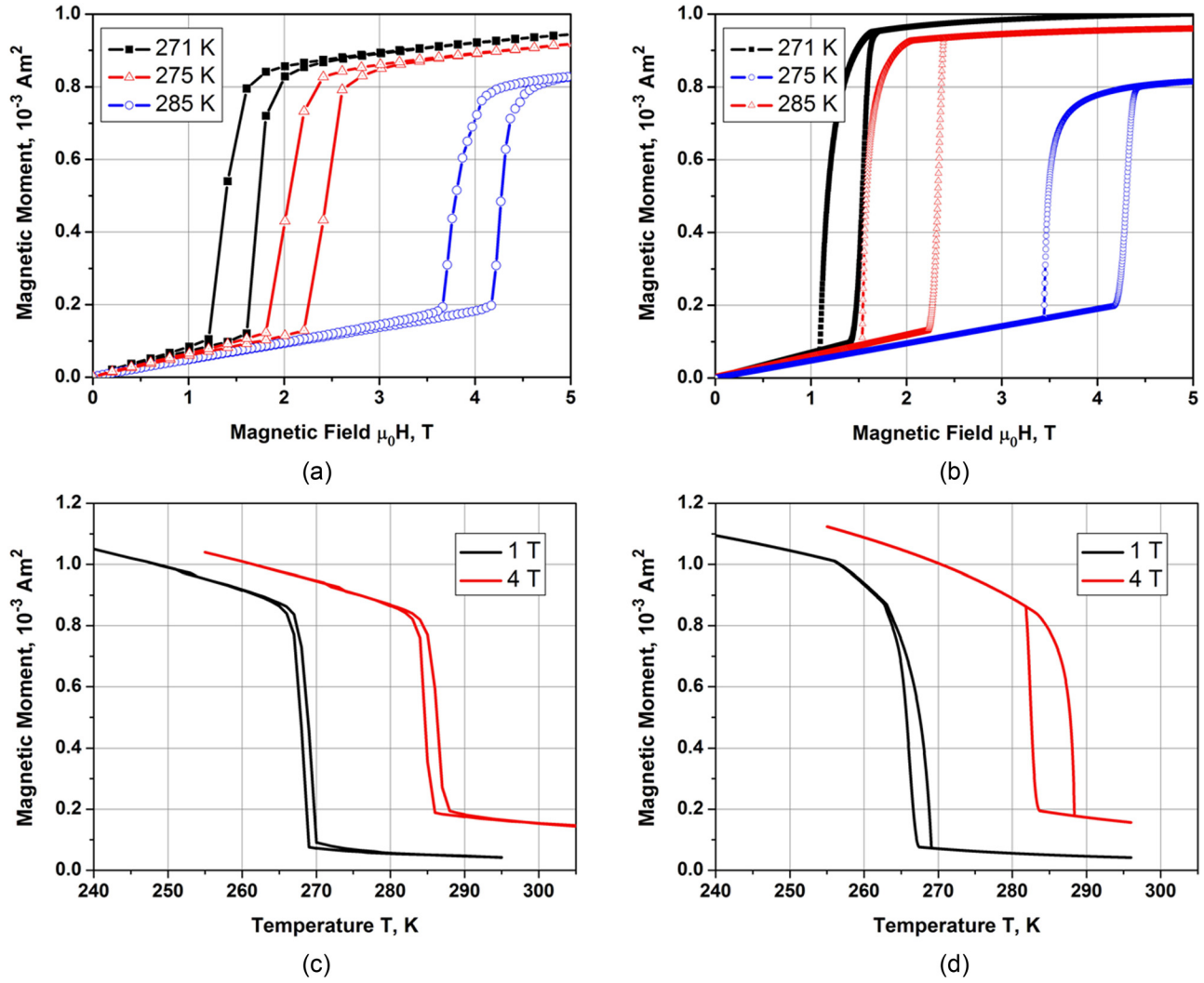
FIG. 1. (a) Measured (adapted from Ref. 10) and (b) modeled magnetization curves showing the induced MS FOPT at different temperatures; and (c) measured (adapted from Ref. 14) and (d) modeled magnetization curves showing the induced MS FOPT at different magnetic fields for $Gd_5(Si_{0.49}Ge_{0.51})_4$.

the MS FOPT systems.[21] In such a case the curve representing the first cycle differs significantly from the curves of the following cycles, which is a clear sign of presence of the residual phase during the transition cycling from the other phase. The proposed model can describe this case if the amount of the monoclinic phase N_{MONO} will not take exact values of 0/1 within the experimental range.

IV. CONCLUSION

To conclude, a phenomenological approach to modelling of first order phase transition in magnetic systems was proposed. In this approach, the ferromagnetic and paramagnetic responses of the system were described using the extended Jiles-Atherton theory of hysteresis and the Curie-Weiss law, respectively. The magnetization of the system is represented by the volumetric amounts of ferromagnetism and paramagnetism in respective phases. An identification procedure to extract material parameters from experimental data was given. The proposed approach was successfully applied to magnetocaloric $Gd_5(Si_xGe_{1-x})_4$ system. The experimental and modeled curves were in good agreement and the approach was shown to describe correctly the

magnetization dependencies under scenarios of varying magnetic field and temperature, including changes in magnetic hysteresis occurring with thermal cycling for the MS FOPT systems. This approach also has the potential to describe the behavior of Griffiths phase magnetic systems.

ACKNOWLEDGMENTS

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