Field dependence of the adiabatic temperature change in second order phase transition materials: Application to Gd

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(Received 13 August 2009; accepted 10 October 2009; published online 19 November 2009)

The field dependence of the adiabatic temperature change ΔT_{ad} of second order phase transition materials is studied, both theoretically and experimentally. Using scaling laws, it is demonstrated that, at the Curie temperature, the field dependence of ΔT_{ad} is characterized by $H^{1/\Delta}$. Therefore, as the magnetic entropy change ΔS_M follows a $H^{(1-\alpha)/\Delta}$ power law, these two dependencies coincide only in the case of a mean field model. A phenomenological construction of a universal curve for ΔT_{ad} is presented, and its theoretical justification is also given. This universal curve can be used to predict the response of materials in different conditions not available in the laboratory (extrapolations in field or temperature), for enhancing the resolution of the data and as a simple screening procedure for the characterization of materials. © *2009 American Institute of Physics*. [doi:10.1063/1.3261843]

I. INTRODUCTION

The optimization of magnetic materials to be applied in magnetic refrigerator prototypes goes through the understanding of the magnetic field dependence of their magnetocaloric effect, in order to fine tune the response of the material under the applied field available in the actual device. Recently, there has been an increasing interest in describing the field dependence of the magnetic entropy change, ΔS_M, but much less efforts have been made to analyze the evolution of the adiabatic temperature change, ΔT_{ad}, as the field is increased. This paper studies in detail the similarities and differences between the field dependencies of these magnitudes, in order to be able to use this knowledge for practical applications.

It has been recently shown that there exists a universal curve for the magnetic entropy change, ΔS_M, of second order phase transition materials. It can be constructed using a phenomenological procedure[1] which does not require the knowledge of either the equation of state or the critical exponents of the material. This construction facilitates the application of the universal curve for practical purposes for the characterization of materials, like predicting the response of the material under experimental conditions not available in the laboratory (extrapolations in temperature or field), enhancing the resolution of the experimental data, etc. It has been demonstrated that it can be successfully applied to different families of soft magnetic amorphous alloys[1,2] (for which it was initially developed) and subsequently extended to rare earth based crystalline materials.[3–5] The theoretical background for this universal curve construction lies on scaling laws.[6] More recently, it has been shown[7,8] that the same phenomenological procedure is suitable for scaling the adiabatic temperature change, ΔT_{ad}, of second order phase transition materials. However, the justification for that finding relied only on a strong simplification: either a weak dependence of the specific heat with magnetic field, which allows to reduce the description of the field dependence of ΔT_{ad} to that of ΔS_M, or the assumption that as the specific heat scales with field close to the Curie transition, the adiabatic temperature change should also scale.

The aim of this work is to show that it is possible to use scaling laws to predict the existence of the universal curve for ΔT_{ad} and that, in contrast with the previous literature which restrict the approach to a mean field model, the field dependence of ΔT_{ad} is not necessarily the same as that of ΔS_M (which is the prediction of the mean field model). Polycrystalline Gd has been used to evidence the accuracy of these claims.

II. EXPERIMENTAL

Polycrystalline Gd (99.5 at. % pure) was obtained from a commercial source. The method of the direct $\Delta T_{ad}(H)$ measurements was described in detail by Tishin *et al.*[9] and Spichkin *et al.*[10] The measurements were made on the Magnetocaloric Measuring Setup "MagEq MMS 801" manufactured by Advanced Magnetic Technologies and Consulting, Ltd., Moscow, Russia. The magnetic field was created by permanent magnet Halbach magnetic field source with changeable magnetic field in its working bore. ΔT values were measured by a differential thermocouple with the measuring junction clamped between two pieces of the material under investigation and a reference junction placed on the nonmagnetic me-

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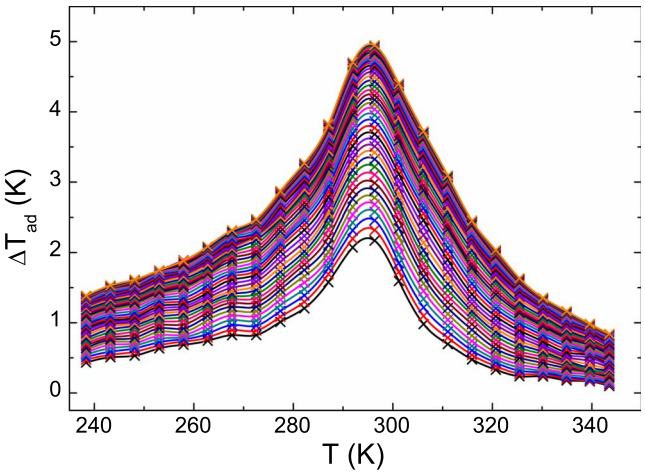

FIG. 1. (Color online) Temperature dependence of the adiabatic temperature change measured in polycrystalline Gd for maximum applied fields ranging from 0.38×10^6 up to 1.50×10^6 A/m. Lines are spline fits to the data.

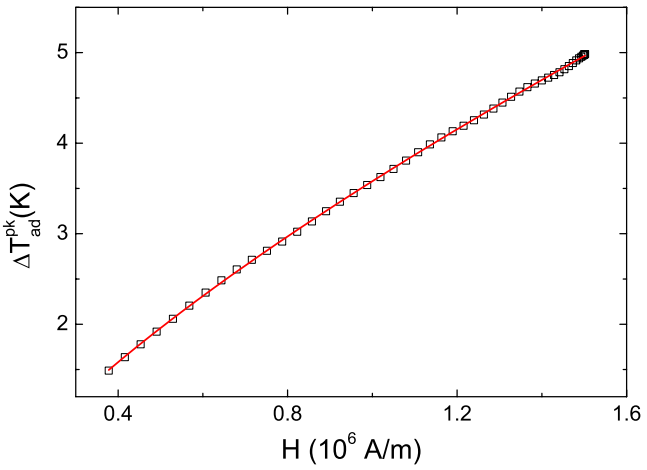

FIG. 2. (Color online) Field dependence of the maximum adiabatic temperature change determined from the spline fits of Fig. 1. Line is the fit to a power law.

tallic sample holder near the sample. ΔT and H values were recorded simultaneously and continuously over the whole cycle of the magnetic field change, which makes it possible to construct the $\Delta T_{\mathrm{ad}}(H)$ curves and on the basis of this data get $\Delta T_{\mathrm{ad}}(T)$ at necessary magnetic field change (ΔH) value. The measurements were made near the corresponding Curie temperatures T_C at the magnetic field change rate of 1 T/s. The maximum magnetic field (H) was 1.87 T.

III. RESULTS AND DISCUSSION

Figure 1 shows the different $\Delta T_{\mathrm{ad}}(T)$ curves for different values of the maximum applied field. In order to find the position and magnitude of the peak, a spline interpolation has been performed and is indicated as continuous lines in the figure. At the peak, the field dependence of ΔT_{ad} can be assumed to be a power law, with an exponent p

$$\Delta T_{\mathrm{ad}}^{\mathrm{pk}} \propto H^p \tag{1}$$

in an analogous way to that of the power law for ΔS_M. From the interpolated curves of Fig. 1, the exponent p can be extracted by performing a nonlinear fit to the data, as shown in Fig. 2. The obtained value is $p=0.70\pm0.01$. This result is in good agreement with previous literature results.[11] Two conclusions can be extracted from this value: the exponent is different from that of ΔS_M for Gd,[4] and it is different from the mean field prediction of 2/3 of the mean field model.[12]

The adiabatic temperature change can be expressed as

$$\Delta T_{\mathrm{ad}} = -\mu_0 \int_0^H \frac{T}{c_p}\left(\frac{\partial M}{\partial T}\right)_H dH. \tag{2}$$

According to Krasnow and Stanley,[13] the specific heat scales with field as

$$\frac{c_p(t,H)}{H^{-\alpha/\Delta}} = c\left(\frac{H}{|t|^{\Delta}}\right), \tag{3}$$

where c is a scaling function. The scaling form of the magnetic equation of state can be expressed as

$$\frac{M}{|t|^{\beta}} = m_{\pm}\left(\frac{H}{|t|^{\Delta}}\right), \tag{4}$$

where $t=(T-T_C)/T_C$ is the reduced temperature, T_C is the Curie temperature, $\Delta=\beta\delta$ is the gap exponent and the plus (minus) sign corresponds to $t>0$ ($t<0$), respectively. Therefore, Eq. (2) transforms to

$$\begin{aligned}\Delta T_{\mathrm{ad}} &= -\mu_0 \int_0^{H/|t|^{\Delta}} dx \frac{T}{H^{-\alpha/\Delta}c(x)}|t|^{1-\alpha}[\beta m_{\pm}(x) - \Delta x m'_{\pm}(x)] \\ &= -\mu_0|t|\int_0^{H/|t|^{\Delta}} dx \frac{T}{c(x)}x^{\alpha/\Delta}[\beta m_{\pm}(x) - \Delta x m'_{\pm}(x)] \\ &= |t|\tilde{f}(t/H^{1/\Delta}) \\ &= H^{1/\Delta} f(t/H^{1/\Delta}).\end{aligned} \tag{5}$$

This equation shows that if the reduced temperature t is rescaled by a factor proportional to $H^{1/\Delta}$, and the adiabatic temperature change also by $H^{1/\Delta}$, the experimental data should collapse onto the same curve. As ΔT_{ad} is a temperature magnitude, it is reasonable that it scales with field in the same way as the temperature axis. It is worth noting the difference with the scaling for the magnetic entropy change, which is $H^{(1-\alpha)/\Delta}$ (where $1-\alpha=\beta+\Delta-1$).[6] In the case of the mean field model, $\alpha=0$ and the field dependences for both magnitudes coincide, as predicted in Ref. 12. However, for any other value set of the critical exponents, the field dependences of ΔT_{ad} and ΔS_M are different, in agreement with our experimental results.

Taking into account the small values of the critical exponent α, a fact which introduces an additional complication for its experimental determination, the comparison between the two different field dependencies of ΔT_{ad} and ΔS_M could be a method for determining the value of exponent α. However, as the experimental setups for measuring the magnetization curves, from which ΔS_M is calculated, and the setup for measuring ΔT_{ad} are intrinsically different, the finding of minor differences in both field dependencies relies extensively on the calibration in field and temperature of both

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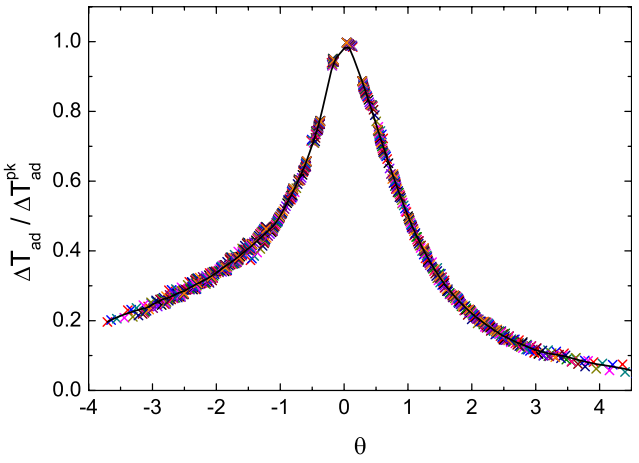

FIG. 3. (Color online) Universal curve for the adiabatic temperature change curves of Fig. 1. Line is the average of the different curves corresponding to the different fields.

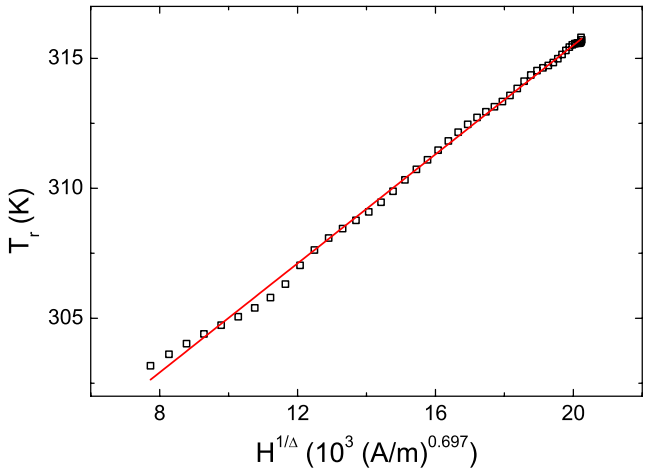

FIG. 4. (Color online) Field dependence of the reference temperature used for constructing the universal curve. The exponent Δ was obtained from the nonlinear fit of Fig. 2. Line corresponds to a linear fit to the data.

setups. Moreover, magnetometers usually do not place the temperature sensor in contact with the sample or sample holder, but measure the temperature of the sample chamber. Therefore, depending on measuring conditions, small oscillations of sample temperature could be expected, imposing a limitation on the accurate determination of α by using this comparison. More detailed studies on these experimental constraints are being undertaken.

In order to construct the phenomenological universal curve for ΔT_{ad}, an analogous procedure to that described in Refs. 1 and 6 for ΔS_M can be used. It consists in normalizing the ΔT_{ad} curves with respect to their maximum and rescaling the temperature axis as

$$\theta = (T - T_C)/(T_r - T_C), \tag{6}$$

where T_r is the temperature of the point of each curve which fulfils $\Delta T_{\mathrm{ad}}/\Delta T_{\mathrm{ad}}^{\mathrm{pk}} = K$ for $T > T_C$, and K is a constant smaller than one which can be arbitrarily chosen for each study (for the present work $K=0.5$) and whose purpose is to set the equivalent points of the different curves at the same reduced temperature $\theta=1$. Figure 3 shows the universal curve constructed for the experimental $\Delta T_{\mathrm{ad}}(H,T)$ curves of Fig. 1. The agreement between this phenomenological construction and the prediction of Eq. (5) can be checked by testing the field dependence of the reference temperature T_r. According to the previously presented theoretical predictions, T_r should scale with field in the same way as ΔT_{ad} does. Therefore, once the field dependence of ΔT_{ad} has been extracted from the nonlinear fit of Fig. 2, it has been used to construct the abscissa axis of Fig. 4, where the straight line is a linear fit to the data, showing a good agreement between theory and phenomenological procedure. A further test would be to check that the temperature at which the peak of ΔT_{ad} takes place, T_{pk}, also scales with field in the same way. In this case, the procedure is trickier, as for determining T_{pk} with enough resolution the previously mentioned spline interpolation of the curves has to be performed. By imposing a resolution of 0.1 K in the interpolate temperature axis, Fig. 5 shows the evolution of the peak position with field, together with a linear fit to the data. Regardless of the discretization of the data points imposed by the resolution, the predicted field dependence is fulfilled. For the maximum applied field of 1.87 T, the increase of the peak temperature with applied magnetic field remains around 1K. The intercept of the linear fits of Figs. 4 and 5 should give the Curie temperature of the material and should be coincident in both cases. The values of the intercepts are 294.5 ± 0.1 and 294.3 ± 0.1, respectively, confirming the good agreement between the experimental results and the theoretical predictions. The accurate determination of the Curie temperature of the sample could be done using standard methods as that of Belov–Goryaga or the one proposed by Kouvel–Fisher. Less standard techniques should be tested on raw samples.

There would be another possible test of the accuracy of the claims, alternative to the self consistent test presented above. It would consist in using the literature values of the critical exponents of Gd for checking the field dependencies of the peak adiabatic temperature change and reference temperatures. However, there is a large dispersion in the literature data for Gd, ascribed to different sample preparations,

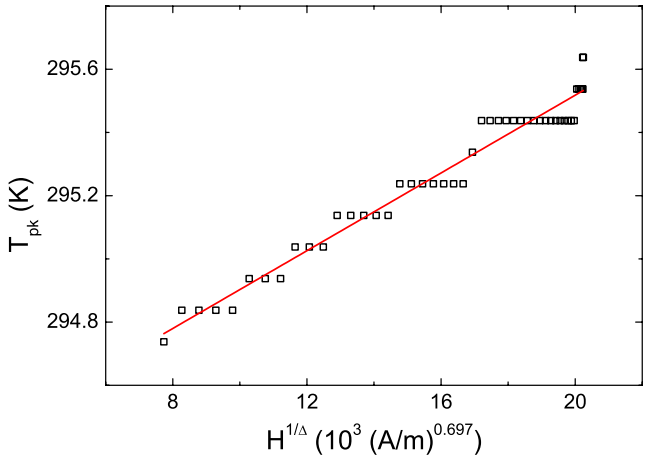

FIG. 5. (Color online) Temperature of the maximum adiabatic temperature change, determined from the spline fits of Fig. 1, as a function of field. The exponent Δ was obtained from the nonlinear fit of Fig. 2. Line corresponds to a linear fit to the data.

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different purity, etc. and results would not be conclusive. Therefore, we consider that checking that all the magnitudes scale with field in the proper way, for the same sample and the same set of measurements, is a stronger test of the consistency of the results.

IV. CONCLUSIONS

A physical basis for the phenomenological construction of the universal curve for ΔT_{ad} has been given by using scaling laws. It is shown that the field dependence of ΔT_{ad} and ΔS_M at the temperature of the peak are not the same, except in the case of a mean field model. This difference in the scaling exponent for both magnitudes can be used to determine the critical exponent α. The phenomenologically constructed universal curve for ΔT_{ad} and the presented field dependencies can be used for predicting the response of the material in different conditions not available in the laboratory (extrapolations in field or temperature), for enhancing the resolution of the data and as a simple screening procedure for the characterization of materials. Theoretical predictions have been checked using experimental result for pure Gd.

ACKNOWLEDGMENTS

This work was supported by the Spanish Ministry of Science and Innovation and EU FEDER (Grant No. MAT 2007-65227), and the PAI of the Regional Government of Andalucía (Grant No. P06-FQM-01823). Y. I. Spichkin and V. I. Zverev thank Advanced Magnetic Technologies and Consulting Ltd. for financial support. Authors are thankful to I. Zubkov for the assistance with experimental measurements.

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