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Scaling laws for the magnetocaloric effect in second order phase transitions: From physics to applications for the characterization of materials

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ABSTRACT

The detailed procedure for constructing the recently proposed phenomenological universal curve for the magnetic entropy change is presented, together with the exponents which control the field dependence of the different magnetocaloric-related magnitudes. Practical applications of the universal curve are also outlined: as a simple screening procedure of the performance of materials, as a method for making extrapolations to temperatures or fields not available in the laboratory, for the reduction of the experimental noise, for correcting the influence of non-saturating conditions, or as a way to eliminate the contribution of minority magnetic phases, among others.

Lois d'échelle gouvernant l'effet magnétocalorique des transitions de phase de seconde ordre: de la physique aux applications permettant de caractériser les matériaux

Mots clés : Réfrigérateur magnétique ; Propriété magnétique ; Alliage ; Matériau

1. Introduction

The search for energy efficient technologies for developing new refrigerator appliances has made the magnetocaloric effect a field of current scientific interest (Tishin, 1999, 2007; Gschneidner and Pecharsky, 2000; Tishin and Spichkin, 2003; Brück, 2005, 2008). Together with their increased performance, it is expected that magnetic refrigerators will have a more reduced environmental impact when compared with those based on the gas compression–expansion cycle, as they

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Nomenclature

ΔS_M	magnetic entropy change ($J\,kg^{-1}\,K^{-1}$)
$\Delta S'_M$	normalized magnetic entropy change
ΔS_M^{pk}	peak magnetic entropy change ($J\,kg^{-1}\,K^{-1}$)
T	temperature (K)
T_r	reference temperature (K)
T_C	Curie temperature (K)
T_{pk}	temperature of the peak entropy change (K)
θ	reduced temperature
H	magnetic field (T)
M	magnetization ($emu\,g^{-1}$)
n	exponent of the field dependence of ΔS_M
δ	critical exponent
β	critical exponent
Δ	critical exponent
α	critical exponent
γ	critical exponent
a	parameter of the Arrott–Noakes equation of state ($g^{1/\gamma}cm^{-3/\gamma}K^{-1}$)
b	parameter of the Arrott–Noakes equation of state ($g^{1/\gamma+1/\beta}cm^{-3/\gamma}emu^{-1/\beta}$)
a_M	scaling factor of ΔS_M ($J\,kg^{-1}\,K^{-1}$)
M_s	spontaneous magnetization ($emu\,g^{-1}$)
M_{max}	maximum magnetization ($emu\,g^{-1}$)
s	scaling function of ΔS_M
RC	refrigerant capacity ($J\,kg^{-1}$)
RC_{FWHM}	refrigerant capacity calculated as ΔS_M^{pk} times the full width at half maximum ($J\,kg^{-1}$)

do not involve ozone-depleting or green house effect related gases.

The magnetocaloric effect is associated to a large change in magnetization close to working temperature of the refrigerant material. Therefore, magnetic refrigeration at low temperatures relied on paramagnetic salts, as their magnetization increase remarkably when approaching 0 K. However, for applications at temperatures close to room temperature, a different approach had to be found: the existence of a phase transition in the material close to the working temperature would produce the required abrupt change in magnetization. From the physical point of view, magnetic refrigerant materials can be classified by the type of phase transition that they undergo. It can be a second order magnetic phase transition (like the ferro-paramagnetic transition of a ferromagnetic material at its Curie temperature), which is characterized by the lack of thermal and magnetic hysteresis, and in which the magnetization decreases continuously to zero (a second order phase transition implies a continuous change in the first derivative of the Gibbs free energy and a discontinuous change in the second derivative of the same) (Zemansky and Dittman, 1996). Pure Gd is a paradigmatic example of a magnetic refrigerant undergoing a phase transition of this kind. But phase transitions can also be of the first order type, in which the first derivative of the Gibbs free energy is discontinuous. Therefore, magnetization shows an abrupt change at the transition temperature, usually associated to a magneto-structural phase transition, giving rise to the giant magnetocaloric effect (GMCE), being GdSiGe the typical case of this kind of magnetic refrigerant materials. However, although the large abrupt change in magnetization causes a correspondingly giant magnetic entropy change, this appears at the cost of thermal and magnetic hysteresis, which should be avoided in order to be able to apply these materials in refrigerators appliances.

The description of the second order magnetic phase transitions in the environment of the critical temperature can be done by using the critical exponents (Stanley, 1999), i.e. by expressing the magnitudes under study as a power law of the relevant magnitude. In a magnetic system, the most relevant critical exponents correspond to the temperature dependence of the spontaneous magnetization at zero field ($M \propto (T_C - T)^{\beta}$, where T_C is the Curie temperature and $T < T_C$), and to the field dependence of magnetization at the Curie temperature ($M \propto H^{1/\delta}$). There exist relationships which allow obtaining other critical exponents, like $\beta + \gamma = \beta\delta$, $\Delta = \beta\delta$ and $1 - \alpha = \beta + \Delta - 1$. Depending on the kind of material, these critical exponents would take certain values and all materials which have the same set of critical exponents belong to the same universality class, following therefore the same power laws in the environment of the phase transition.

The study of the field dependence of magnetocaloric response of the material is also a field of increasing research interest (Tishin et al., 2007; Arora et al., 2007). From a fundamental point of view, an understanding of this field dependence for different types of materials can give further clues on how to improve the performance of refrigerant materials for the magnetic field range employed in actual refrigerators (generally 10–20 kOe), which would be a major breakthrough for the design of eventually domestic appliances. From a practical point of view, the knowledge of the laws governing the field dependence of the magnetic entropy change (ΔS_M) can provide tools for making plausible extrapolations to experimental conditions not available in some laboratories. Reliable tools for extrapolating the measured curves in the magnetic field axis could be a help for simplifying the identification of potential magnetic refrigerant materials.

A deep understanding of the field dependence of ΔS_M goes through the modelization of the magnetocaloric response of the materials, which can be approached in different ways. Some authors develop first principles models for particular materials, which have a strong predictive power but have to be tailored for each particular alloy (Paudyal et al., 2007). Thermodynamical models, as they contain less specific information for the material under study, can be regarded as more general (in the sense that it can be extended to different types of materials in an easier way), but usually this approach has been limited to mean field models (Amaral and Amaral, 2004). However, when studying the field dependence of the magnetic entropy change in materials with a second order magnetic phase transition, serious discrepancies between the predictions of the mean field approach (Oesterreicher and

Parker, 1984) and the experimental data (Franco et al., 2006a, b) were found. Nevertheless, it has been recently shown that there exists a universal curve for the field dependence of the magnetic entropy change in materials with a second order phase transition, which can be constructed phenomenologically without knowing the critical exponents of the material or its equation of state and which is not restricted to the mean field case. This phenomenological procedure has been successfully applied to different families of soft magnetic amorphous alloys (Franco et al., 2006c, 2007a) (for which it was initially developed) and subsequently extended to rare earth based crystalline materials (Franco et al., 2007b, 2009a; Dong et al., 2008).

The purpose of this work is to give a detailed overview of the procedure to construct this phenomenological universal curve, showing also an alternative construction which does not require a detailed knowledge of the experimental data at the peak entropy change. A theoretical justification for the existence of the universal curve and of the validity of this phenomenological procedure will also be overviewed. We will focus our attention on practical applications of this universal curve: (1) to make extrapolations up to fields or temperatures which are not available in the laboratory, (2) to detect the existence of overlapping magnetic phenomena, and (3) to enhance the resolution of the measurements close to the temperature of the peak, being a procedure to reduce the noise in the measurements without distorting the shape of the peak. Nevertheless, as real samples depart from the ideal cases contemplated in the theory, this originates some limit cases to the applicability of this universal curve. By studying materials with some magnetic impurity phases in them, or situations in which the sample does not reach technical saturation, a modified universal curve can be constructed, adding new practical functionality to the method, like the extraction of the information arising from the main magnetic phase, eliminating that coming from the minority impurity phase, or the ability to compare the results of samples with different (or even unknown) shapes.

2. Phenomenological universal curve

The virtue of the phenomenological universal curve is that either if a single magnetic material is measured up to different maximum applied fields, or if different materials with similar values of their critical exponents (essentially, but not limited to, alloys of the same compositional series), their $\Delta S_M(H,T)$ curves can be rescaled onto a single curve which does not depend on field and in which the different temperature dependencies of the different alloys are unified (Fig. 1).

The phenomenological procedure for the construction of this curve is shown in Fig. 2 with the help of simulated data for a mean field model calculated for different maximum applied fields. The basic assumption of the method is that if there is a universal curve, equivalent points of the different experimental curves should collapse onto the same point of the universal curve. Therefore, the key is to identify which are the equivalent points of the different curves for different fields. As there is no doubt that the peaks (which in the mean field case coincide at the Curie temperature, T_C) should be in equivalent conditions, we can assume that points which are at a certain level with respect to the peak are also in equivalent magnetic states. The temperatures of these points, which are marked by crosses in Fig. 2, will be denoted as reference temperatures T_r, and their identification constitutes the first step of the procedure. The value of the factor selected for this identification is arbitrary, not affecting the procedure (in the case of Fig. 2, the choice has been $\Delta S_M(T_r) = 0.7\Delta S_M^{pk}$). The second step

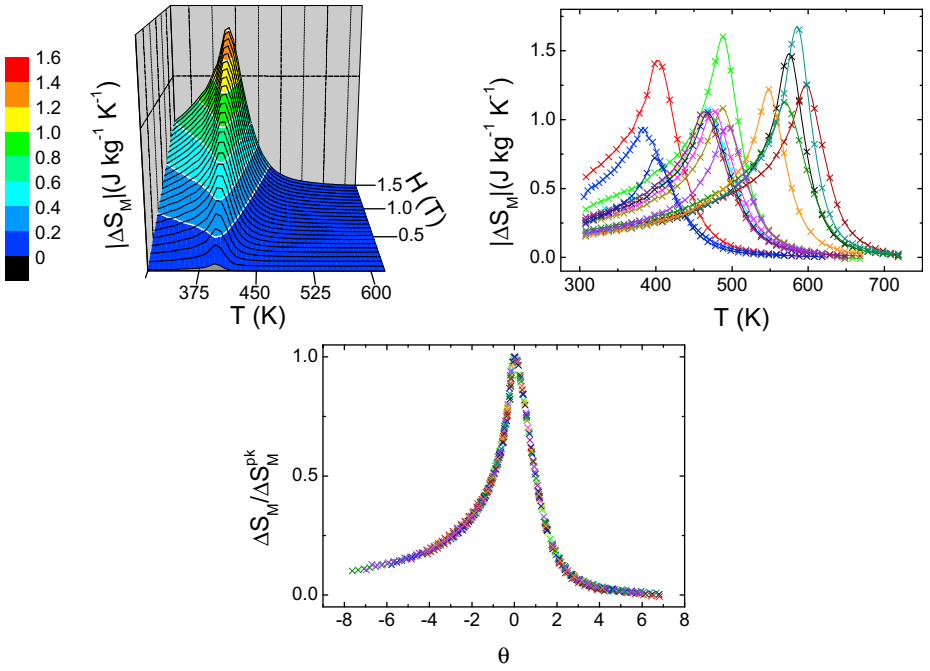

Fig. 1 – Upper-left: $\Delta S_M(H,T)$ curves for a typical Fe-based amorphous alloy; upper-right: results for 16 different alloys with similar values of the critical exponents; lower: phenomenological universal curve corresponding to these experimental data.

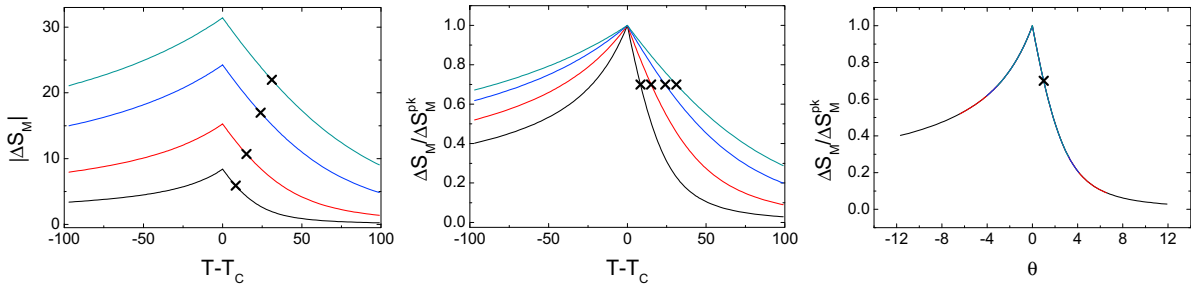

Fig. 2 – The three different steps in the phenomenological construction of the universal curve: identification of the reference temperatures (crosses); normalization; rescaling the temperature axis to place T_r at $\theta=1$.

is the normalization of the curves with respect to their maximum. Finally, the temperature axis is rescaled in such a way that the reference temperatures are all at $\theta = 1$ by using

$$\theta = (T - T_C)/(T_r - T_C) \tag{1}$$

It is therefore shown that by imposing the position of two points of each of the curves (those at T_C and at T_r), which implies three free parameters, the whole curve collapses in a single universal curve. In the case that the critical exponents of the material are different, i.e. the material is from a different universality class, the shape of the curve will be altered. Most notably, the position of the peak will be displaced with respect to T_C. However, it has been shown that using either T_C or T_{pk} in Eq. (1) does not alter the construction of the universal curve (Franco et al., 2009b).

3. Field dependence of ΔS_M

It can be assumed that the field dependence of the magnetic entropy follows a power law of the field:

$$\Delta S_M \propto H^n \tag{2}$$

with an exponent which depends on temperature and field which can be locally calculated as:

$$n = \frac{d\ln|\Delta S_M|}{d\ln H} \tag{3}$$

Fig. 3 shows, for a mean field model, the typical evolution of the $n(T)$ curves calculated for different values of the maximum applied field. At low temperatures, well below T_C, n has a value which tends to 1, which indicates that although the magnetization curves depend on temperature at these temperatures, this dependence is essentially field independent. At temperatures well above T_C, n tends to 2 as a consequence of the Curie–Weiss law. At $T = T_C$, n has a minimum. For the mean field case $n(T_C) = 2/3$, as predicted by Oesterreicher and Parker (1984). However, for any other case, the value of the minimum is different from that and is related to the critical exponents of the material (Franco et al., 2006c):

$$n(T = T_C) = 1 + 1/\delta(1 - 1/\beta) = (1 - \alpha)/\Delta \tag{4}$$

There were experimental evidences that the values of n also collapse when plotted against the same rescaled temperature axis for which the normalized values of ΔS_M collapse (Franco et al., 2007c, d). This allows for an alternative construction of the universal curve, which does not rely on the magnitude of ΔS_M^{pk} and therefore is useful when data close to the peak are not dense enough. The procedure consists in identifying the reference temperatures as those which have a certain value of n. In Fig. 3 this value has been arbitrarily selected as $n(T_r) = 1.5$ and is marked as a dashed line in the $n(T)$ curves. Once the reference temperatures are selected, the rescaled temperature axis is constructed using Eq. (1) and the normalization of the ΔS_M curves can either be done using ΔS_M^{pk}, as indicated in the previous section, or as $\Delta S'_M = \Delta S_M/\Delta S_M(T_r)$. This latter method has been used in Fig. 3.

It is worth mentioning that when the critical exponents of the material are not of the mean field case (i.e. different from $\beta = 0.5;\ \gamma = 1;\ \delta = 3$), the temperature at which n has its

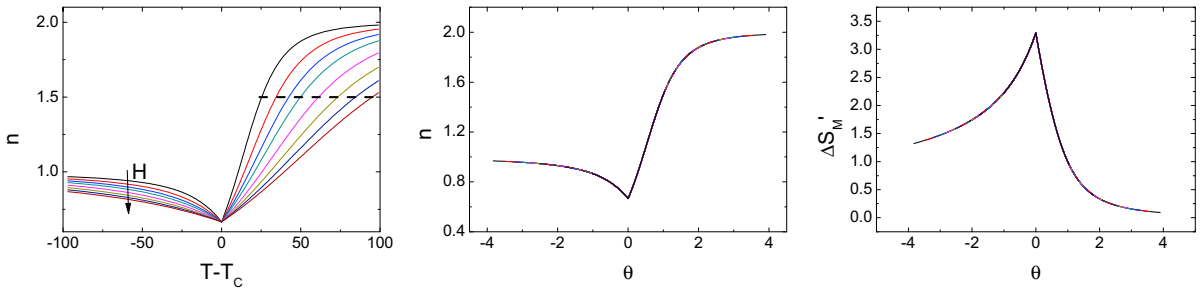

Fig. 3 – Temperature dependence of n calculated for a mean field model for different values of the maximum applied field (left). Center: rescaled curves when Eq. (1) is used with reference temperatures selected as $n(T_r) = 1.5$. Right, corresponding universal curve after normalizing with $\Delta S_M(T_r)$.

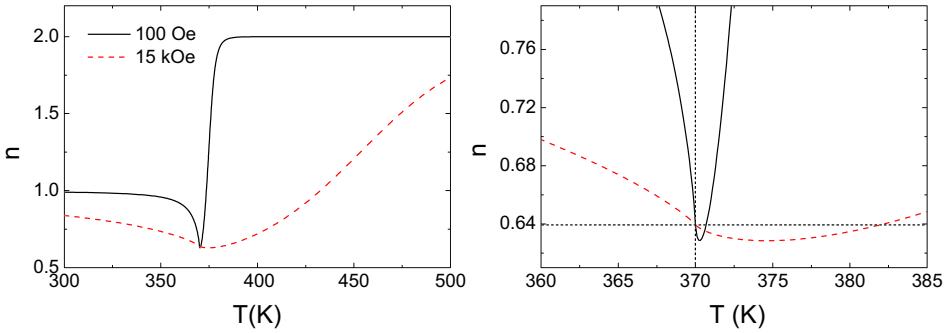

Fig. 4 – Temperature dependence of the exponent *n* for the Heisenberg model for two maximum applied fields. On the right is the magnification of the region close to the minimum. The vertical dotted line corresponds to T_C.

minimum does not correspond to T_C. As an example Fig. 4 shows the $n(T)$ curves for a Heisenberg model. It is observed that there are two temperatures which have the same value of n: T_C and the temperature of the peak entropy change. In between these two temperatures the minimum of n takes place. However, while the former temperature is field independent, the latter is displaced by field to higher temperatures.

4. Theoretical justification and further field dependencies

The first attempt to make a formal check of the suitability of this phenomenological procedure and find the scaling laws for the different magnetocaloric-related magnitudes may consist in using a particular magnetic equation of state, which relates magnetization, temperature and field. As the magnetic entropy change can be calculated from the derivatives of the $M(H,T)$ curves

$$\Delta S_M = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH \tag{5}$$

the manipulation of the equation of state may lead to the desired results. In the case of Oesterreicher and Parker (1984), they chose the description of the system in the paramagnetic region, assuming a mean field case; in our case (Franco et al., 2006c), the Arrott–Noakes (1967) equation of state was chosen,

$$H^{1/\gamma} = a(T - T_C)M^{1/\gamma} + bM^{(1/\beta)+(1/\gamma)} \tag{6}$$

which is valid in the environment of the Curie temperature of the material. In this case, most of the magnitudes can be calculated from the magnetization curves without the need of numerical derivatives (which can be a source of errors close to the transition temperature):

$$\Delta S_M = -\int_{M_s}^{M_{max}} a\gamma M\left(a(T - T_C) + bM^{1/\beta}\right)^{\gamma-1} dM \tag{7}$$

$$n = \frac{H}{\Delta S_M}\frac{d}{dH}\int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH = \frac{H}{\Delta S_M}\left(\frac{\partial M}{\partial T}\right)_H \tag{8}$$

$$\frac{\partial M}{\partial T} = \frac{-aM}{a(T - T_C)(1/\gamma) + b((1/\beta) + (1/\gamma))M^{1/\beta}} \tag{9}$$

For the mean field case, these expressions are further simplified to:

$$\Delta S_M \ (\text{mean field}) = -\frac{1}{2}a\left(M^2 - M_s^2\right) \tag{10}$$

$$n(\text{mean field}) = 2\frac{MH}{\left(M^2 - M_s^2\right)\left(\frac{H}{M} + 2bM^2\right)} \tag{11}$$

However, it is not necessary to impose any particular equation of state to justify that there should be a universal curve for the magnetic entropy change. It is well known that second order phase transitions scale with field (in the magnetic case, if the magnetization curves are conveniently plotted, all the $M(H,T)$ curves should collapse onto a universal curve). Therefore, as the magnetic entropy change is obtained from the magnetization data though eq. (5), it is logical that the $\Delta S_M(H,T)$ curves should also collapse. By assuming a general scaling equation of state (Franco et al., 2008a), the magnetic entropy change can be expressed as

$$\Delta S_M/a_M = H^{(1-\alpha)/\Delta} s\left(t/H^{1/\Delta}\right) \tag{12}$$

where $s(x)$ is a scaling function. Therefore, the check of the agreement between the phenomenological procedure and this theoretical development can be made by checking the scaling of the peak entropy change, and that the reference temperature should scale with field as $1/\Delta$. There are numerous experimental evidences of the agreement with theory for both field dependencies (Franco et al., 2006c, 2008a, 2008b, 2009b).

Of particular interest can be the scaling law for the refrigerant capacity (RC). There are several different definitions of this magnitude in the literature. One is the product of the peak entropy change times the full width at half maximum of the peak (RC_{FWHM}); another the area under the $\Delta S_M(T)$ curve using the temperatures at half maximum of the peak as the integration limits (RC_{Area}). Taking into account the scaling of the peak entropy change and of the reduced temperature axis, any of these two definitions of RC should scale with field as $1/\Delta + (1-\alpha)/\Delta = 1 + 1/\delta$. A good agreement between the experimental RC data and this scaling law has also been evidenced (Franco et al., 2008a). Table 1 summarizes the different scaling laws for the relevant parameters of the magnetocaloric curves.

Table 1 – Exponents controlling the field dependence of different magnitudes related to the magnetocaloric effect (magnitude $\propto H^{exponent}$).

Magnitude	Exponent
T_r	$1/\Delta$
$T_{pk} - T_C$ (not mean field)	$1/\Delta$
$T_{pk} - T_C$ (mean field)	0
$\Delta S_M(T = T_c)$	$1 + 1/\delta(1 - 1/\beta) = (1 - \alpha)/\Delta$
ΔS_M^{pk}	$1 + 1/\delta(1 - 1/\beta) = (1 - \alpha)/\Delta$
RC_{Area} or RC_{FWHM}	$1 + 1/\delta$

The critical exponents employed have their usual meanings.

5. Applications of the universal curve

5.1. *Extrapolations in temperature/field*

From the different possible applications of the universal curve, probably the most straightforward is the extrapolation of the data to conditions not available in the laboratory. Fig. 5 shows that the low field $\Delta S_M(T)$ curve covers the whole rescaled temperature axis, while for the same number of points, the larger field data are constrained to a much narrower rescaled temperature range. Therefore, a more detailed information of the region close to the peak is provided by the high field curve, while the low field one can be used to provide extrapolations to higher or lower temperatures for the high field data. When this is applied to series of alloys, in the case that some composition falls outside the available temperature span, undoing the transformation of the universal curve to the real (not rescaled) axes can be a method to extrapolate the data for that composition (Franco et al., 2007d). In this way, a simple screening of the performance of a material can be made to assess its suitability for the desired application.

5.2. *Enhancement of the resolution of the data*

Apart from the more detailed information close to the peak which can be incorporated into the low field data by using the universal curve, as indicated in the previous subsection, a procedure for eliminating excessive noise in the measurements can be developed, without the need of smoothing each curve (that would be a source of distortions of the shape of the peak). Fig. 6 shows the addition of 15% peak to peak noise to the results of a mean field model. By averaging the different rescaled noisy curves, a less noisy shape is obtained. Once the transformation to experimental axes is made, it is seen that the noise level can be strongly reduced and the shape of the corrected curves resemble those of the theoretical starting point.

5.3. *Multiphase materials*

The validity of this phenomenological procedure, i.e. the use of the scaling equation of state for a particular material, has the restriction that it should be a single magnetic phase. In the

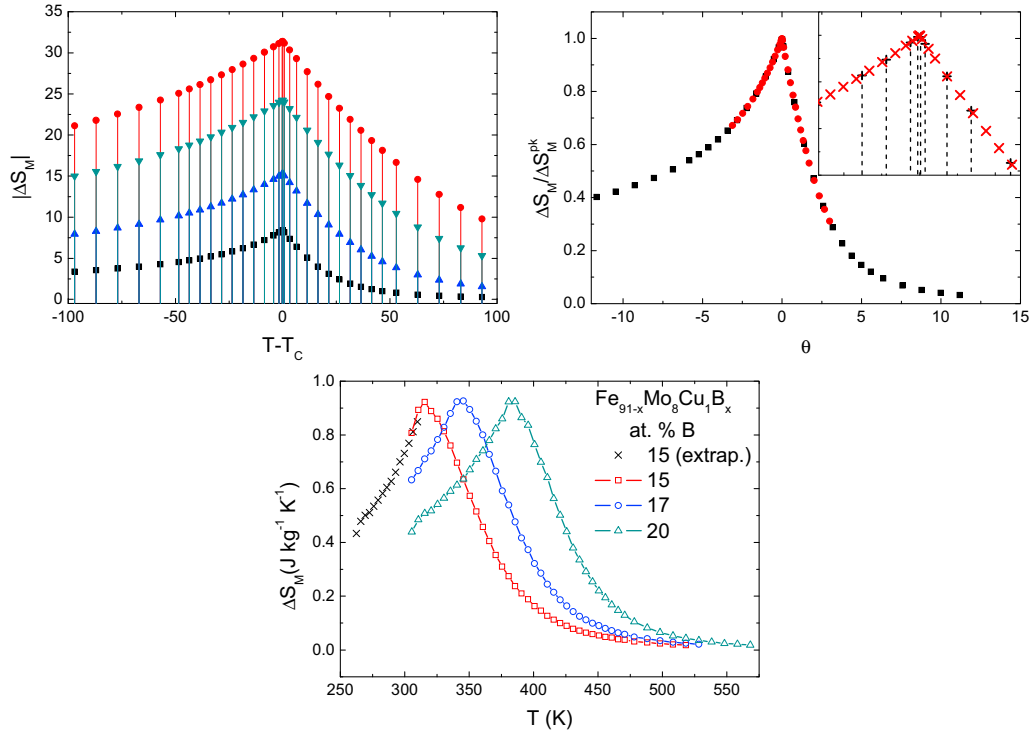

Fig. 5 – Construction of the universal curve for the mean field model, showing that the same number of points cover a different span for the low and high field data. Lower: extrapolation of the magnetic entropy change curve for a composition outside the available experimental range.

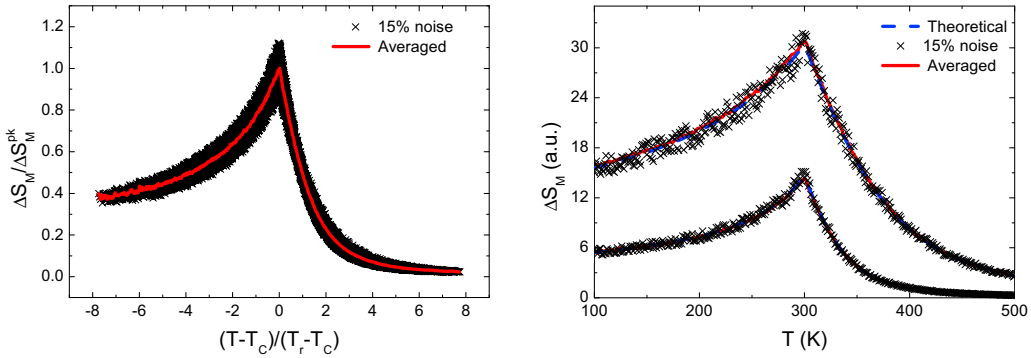

Fig. 6 – Addition of 15% peak to peak noise to a mean field model. The averaging of the different curves for the different fields in the rescaled axes provides a smooth version of the universal curve which, upon transformation to the experimental axes, does not differ from the theoretical noiseless curves. On the right, two values of the field are presented as an example.

case that the sample consists of different magnetic phases, the construction of the universal curve as exposed in the previous sections is no longer valid (Franco et al., 2009a), as can be observed in the deviations of the rescaled curves in the low temperature range of Fig. 7 for the case of a second order phase transition LaFeSi sample. However, by using two reference temperatures for each of the curves, one below and another above T_C, selected as those corresponding to a certain fraction of the peak (as explained in the original procedure), and rescaling the temperature axis as:

$$\theta = \begin{cases} -(T - T_C)/(T_{r1} - T_C); T \leq T_C \\ (T - T_C)/(T_{r2} - T_C); T > T_C \end{cases} \quad (13)$$

the temperature environment close to the peak collapses into a single curve again. Therefore, the existence of a second magnetic phase, with a transition temperature even well above the experimental range, can be detected by this non-collapsing character of the magnetocaloric curves.

The anomalous field dependence of the exponent n can be qualitatively described by assuming that the sample is composed of two independent magnetic phases (Franco et al., 2009a). However, the limitations of this simple approach is being currently analyzed by using a simpler sample (Caballero-Flores et al., 2009a).

It has also been shown that the existence of overlapped magnetic transitions can be more easily detected by analyzing

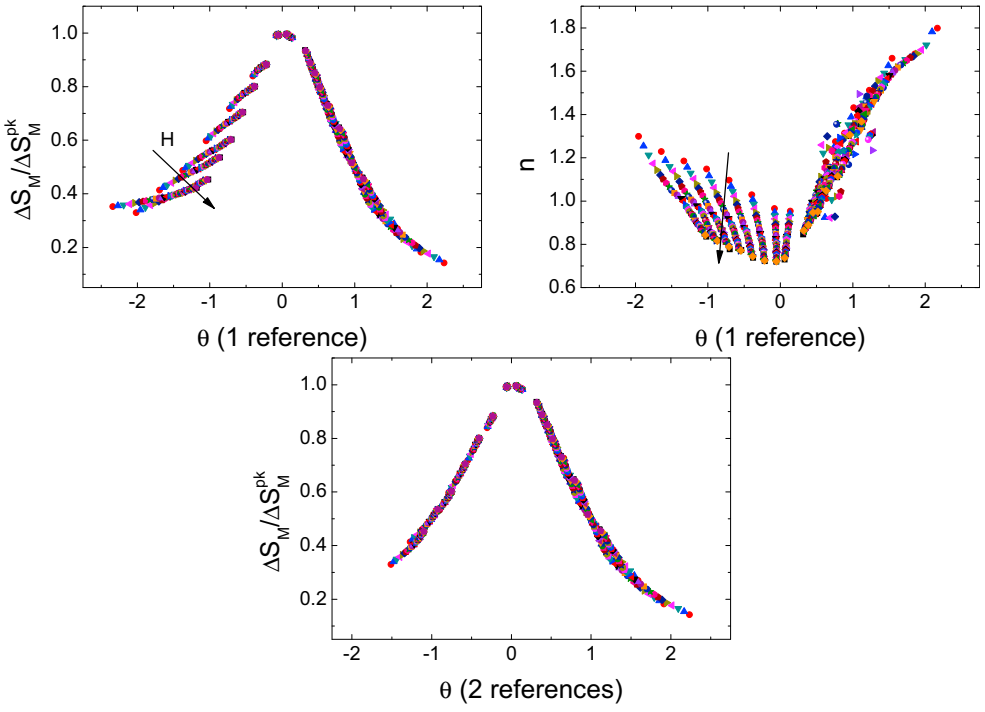

Fig. 7 – Attempt of constructing the universal curve for the $LaFe_{10.8}Si_{2.2}$ sample. Upper-left, failure of constructing the universal curve with a single reference temperature; upper-right: lack of collapse of the values of the exponent n; lower: collapse of the data using two reference temperatures. The arrows indicate the evolution of the curves with increasing magnetic field.

either the field and temperature dependence of n, or the lack of overlapping of the magnetic entropy change curves (Franco et al., 2007b). It is expected that by separating the collapsing and non-collapsing components of the $\Delta S_M(H,T)$ curves a procedure for performing deconvolutions will be developed.

5.4. *Non-saturating conditions*

Another limitation of the universal curve in the original form is the requirement that the samples should be single domain, i.e. they should be in a state of technical saturation. If the applied field is small, or if the demagnetizing factor of the sample under study is large, the construction of the universal curve with a single reference temperature is not possible. This can be seen as a distortion of the field dependence of the $\Delta S_M(H,T)$ curves with respect to the predictions of the equation of state. However, it has recently been shown (Caballero-Flores et al., 2009b) that the use of two reference temperatures also allows constructing a curve on which all the experimental data collapse in the environment of the magnetic entropy change peak.

6. Conclusions

The recently proposed phenomenological construction of the universal curve for the magnetocaloric effect has a theoretical background which relies on the scaling of second order phase transitions. However, although from a theoretical point of view obtaining the universal curve requires the knowledge of the critical exponents of the material and its equation of state (a situation which is rather unlikely when first going to the laboratory with a new material), the phenomenological procedure can be followed without this a priori knowledge. This opens the possibility of different practical applications of the universal curve in the characterization of new materials: as a simple screening procedure of the performance of materials, as a method for making extrapolations to temperatures or fields not available in the laboratory, for the reduction of the experimental noise, for correcting the influence of non-saturating conditions, or as a way to eliminate the contribution of minority magnetic phases.

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