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# Influence of the demagnetizing factor on the magnetocaloric effect: Critical scaling and numerical simulations

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In recent years, the magnetocaloric effect is studied not only for the search of potential magnetic refrigerant materials but also for the analysis of critical phenomena. In both cases, the demagnetizing field might have a notable influence on the results. In this work, we carry out a systematic study, based on theoretical simulations, of the influence of the demagnetizing factor on the magnetocaloric properties. On the one hand, we show that demagnetizing factor affects only slightly the magnetic entropy change ($\Delta S_M$), reducing its magnitude and shifting the peak to higher temperatures. On the other hand, it dramatically affects the exponent $n$ of field dependence ($\Delta S_M \propto H^n$) at temperatures below the peak. We demonstrate that scaling of the magnetocaloric curves can be used to remove the influence of the demagnetizing field and, to which extent, critical exponent determination can be affected. Results of numerical simulations are compared with experimental data from a ball milled powder alloy. © 2014 AIP Publishing LLC.
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The Magnetocaloric effect (MCE) has become a relevant topic in condensed matter magnetism and applied physics. This is due to its applications for near room temperature refrigeration,[1] as a useful tool in the study of magnetic phase transitions,[2] for more fundamental studies in apparently unrelated materials as graphene,[3] and even applications in medicine in treatments of malignant tumors by hyperthermia or in drugs delivery and release processes.[4] Since the discovery in 1997 (Ref. 5) of materials with a giant change of their magnetic entropy when removing the applied field, the interest of the scientific community on MCE grew enormously. After almost twenty years of intense research in this field, a lot of materials with excellent magnetothermal properties have been found (both with first and second order phase transitions), the physical foundations of this phenomenon have been clarified from the theoretical and experimental points of view and extensive review papers have been published.[6–8]

In order to perform reliable comparisons between experimental data measured under different conditions, gauge which is the influence of sample quality, discern the possible appearance of experimental artifacts, or fully analyze the results in order to make more fundamental studies of phase transitions, a number of usually neglected details have to be taken into account, like sample homogeneity, interactions between phases or, what will concern us in this work, the demagnetizing field. This is particularly important for powder shaped samples or when there are interphases between phases with different magnetic characters inside the sample. Most of the previous studies on this topic have followed a phenomenological experimental approach. It has already been shown a significant influence of demagnetizing field in amorphous ribbons[9,10] and more recently in powder samples.[11,12] Additionally, the field dependence of both maximum magnetic entropy change[13] and maximum adiabatic temperature change[14] are clearly affected by demagnetizing field so they must be determined and taken into account in order to get the same behavior regardless the shape of the sample or the direction of the applied field. Since it has been repeatedly proven that demagnetizing field is not negligible for the MCE determination, in some cases some corrections are being included in the standard procedures of integration of the experimental data in MCE measurements,[15] but these are rare cases. It is worth mentioning that most MCE publications do not even mention the shape of the samples which were studied.

In the present work, we carry out a systematical study of the influence of the demagnetizing field on MCE, with the aim of finding out the qualitative and quantitative behavior of samples for which the demagnetizing field is not negligible and propose a method to correct this influence. If we go deeper in the understanding of this phenomenon, we will be able to have a better characterization of MCE materials discriminating in the measurements the effects arising from the demagnetizing field. As we are performing numerical simulations in which we start with a "sample" without demagnetizing field and we gradually add the demagnetizing field into it, our results will not be affected by potential extraneous effects coming from other contributions from the sample (like inhomogeneities, experimental uncertainties, etc.). The MCE of a substance is characterized by its isothermal change in the magnetic entropy when an applied field $H$ is changed. This entropy increment can be calculated from the isothermal magnetization curves using Maxwell relation

$$\Delta S_M = \mu_0 \int_0^H \left( \frac{\partial M}{\partial T} \right)_{H'} dH'. \tag{1}$$

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It is convenient to define the following quantity:

$$n = \frac{d \ln|\Delta S_M|}{d \ln H}, \qquad (2)$$

which describes the variation of the magnetic entropy change with respect to the field at a given temperature following a power law $\Delta S_M \propto H^n$.[16] This exponent, which is a function of temperature and applied field, presents three field independent ranges: well below the $T_C$ ($n = 1$), at $T = T_C$ and well above the $T_C$ ($n = 2$).

Another important quantity to define in the MCE of materials is the refrigerant capacity ($RC$), which is a measure of the amount of heat that can be transferred between the cold and hot reservoirs and it is calculated from the magnetic entropy change curves as

$$RC = -\int_{T_1}^{T_2} \Delta S_M dT, \qquad (3)$$

where the integral is calculated in the interval $\delta T_{FWHM} = T_2 - T_1$, which corresponds to the full width at half maximum of the magnetic entropy change peak.

In our work, we will use the Arrott-Noakes (AN) equation of state[17] for generating the temperature and field dependent magnetization curves, which is given by the following expression:

$$\left(\frac{H}{M}\right)^{1/\gamma} = a(T - T_C) + bM^{1/\beta}. \qquad (4)$$

Here, $\beta$ and $\gamma$ are critical exponents, $T_C$ is the Curie temperature, and $a$ and $b$ are two fitting parameters of each material. We will use the data for a typical soft magnetic amorphous alloy (e.g., $Fe_{77}Cr_8B_{15}$), where $a = 1.228$ (Oe g/emu)$^{1/\gamma}$ K$^{-1}$, $b = 0.0077$ Oe$^{1/\gamma}$(g/emu)$^{1/\gamma+1/\beta}$, and critical exponents are $\beta = 0.4579$ and $\gamma = 1.5593$ and $T_C = 370$ K.[18] We choose these parameters set because of two reasons: first, as we have pointed out, in soft magnetic materials the demagnetizing field is more intense at low applied fields, and second, we

successfully showed that AN equation reproduces the experimental results with a high accuracy.[18] In order to model the MCE of a material, we first generate isothermal magnetization curves with equation 4 (zero demagnetizing factor case), and then we calculate the applied field from the internal field and magnetization using $H_{int} = H - N_D M$, where $N_D$ is the demagnetizing factor, which varies from 0 to 1 in SI units (0–4$\pi$ in CGS). Magnetization curves were generated for different $N_D$ values with close enough temperatures ($\Delta T = 0.2$ K in our case) in order to calculate numerically the integral of Eq. (1) and determine the magnetic entropy change.

In our investigation, we have carried out a series of simulations calculating the magnetic entropy change and the exponent $n$ of the cited model alloy in a wide temperature range (200–600 K) around $T_C$ for maximum applied fields of 0.5, 1.0, and 1.5 T (Fig. 1). As it would be expected, for $T \gg T_C$ the influence of demagnetizing field is practically negligible because it corresponds to the paramagnetic range where the magnetization is small and hence the demagnetizing field. However, we find a significant drop in the magnetic entropy change with increasing $N_D$ for $T < T_C$. The maximum entropy change $|\Delta S_M^{pk}|$ decreases linearly with increasing demagnetizing field. The slope of the linear decrement is independent of the applied field. The position of the maximum slightly moves to higher temperatures, as it was pointed out previously by Lyubina *et al.*[13] Interestingly, when we focus on the $n$ exponent behavior we observe a dramatic change in the general trends. Again for $T \gg T_C$, we do not find any detectable change and $n \to 2$. Using the Arrott-Noakes equation of state, we would expect at $T = T_C$ a value of $n(T_C) = 1 + (\beta - 1)/(\beta + \gamma)$,[16] and this is what we found when $N_D = 0$, but when we take into account the demagnetizing field we find a shift to higher $n$ values. For a given field value, $n$ increases with $N_D$, with a slope which decreases with increasing applied field as it is shown in the inset of Figure 1. For $T \ll T_C$, the field independent limit of $n \to 1$ is lost in the presence of demagnetizing field. This deviation from the ideal behavior must be understood as a proof of the
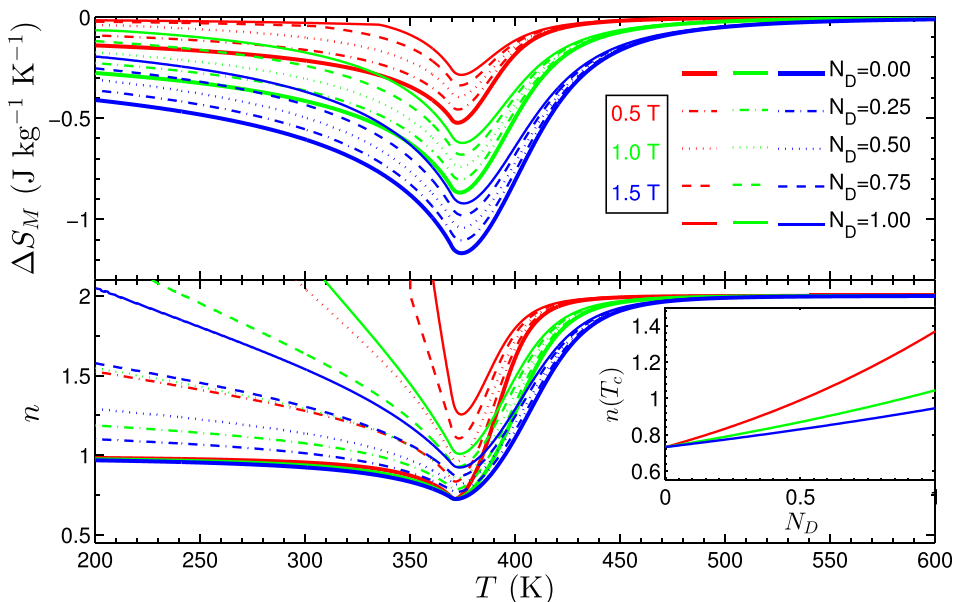


FIG. 1. Magnetic entropy change curves for applied fields of 0.5, 1.0, and 1.5 T (upper) and the corresponding curves of the $n$ exponent as a function of temperature (lower). Notice how $n$ exponent evaluated at the Curie temperature $n(T_C)$ varies with the applied magnetic field when demagnetizing effects are taken into account (inset).

existence of demagnetizing effect in the sample. Otherwise, the ideal limit would be reached for all applied fields.[19] Now we study if the effects of demagnetizing field can be corrected using the universal curve of the magnetic entropy change.[16,19] This universal curve allows us to get the same general behavior of $\Delta S_M$ as a function of reduced temperature for all applied fields. In order to construct this phenomenological curve, we first normalize the magnetic entropy change curves dividing by their maximum: $\Delta S_M/\Delta S_M^{pk}$. Then, we choose two reference temperatures which must fulfill the following conditions: $\Delta S_M(T_{r1} < T_C)/\Delta S_M^{pk} = \Delta S_M(T_{r2} > T_C)/\Delta S_M^{pk} = h$, where $h < 1$ is an arbitrary constant. Although in theory $h$ could be freely selected between 0 and 1, a too large value (reference temperatures chosen too close to the peak temperature) would produce large numerical errors due to the limited number of points—in experimental measurements—which lie in that region. Conversely, if $h$ is too small it implies selecting reference temperatures far away from the critical region, where other phenomena could take place. Therefore, in this case we will use $h = 0.6$. Once the two reference temperatures are found, we define a new variable $\theta$ for the temperature axis as

$$\theta = \begin{cases} -(T - T_C)/(T_{r1} - T_C) & \text{for } T \leq T_C \\ (T - T_C)/(T_{r2} - T_C) & \text{for } T > T_C. \end{cases} \quad (5)$$

The universal curve has been successfully proved in several materials with a second-order phase transition with very different compositions, ranging from rare earths to transition metal based amorphous alloys and manganites.[20–25] However, materials with first-order phase transitions do not collapse onto a universal curve and this fact can be used to determine the nature of the phase transition in a given material.[26] For $\theta > 0$, we expect a collapse for both first and second-order phase transition materials due to the paramagnetic behavior. It is only for $\theta < -1$, when we are well below $T_C$, when we would expect a vertical broadening $W$ in the curve. From this broadening, we can define a percentage dispersion $\sigma$ as follows:

$$\sigma = \frac{W(\theta = -5)}{\Delta S_M/\Delta S_M^{pk}(\theta = -5)} \times 100. \quad (6)$$

It has been shown[26] that for second-order phase transition materials this dispersion keeps under 30%, while for first-order phase transition materials is always bigger than 100%. In Figure 2, we have represented the universal entropy change curve of the alloy for different applied fields and demagnetizing factors and we can confirm how all effects can be included in the curve and $\sigma < 30\%$ as it corresponds to second order phase transition materials. Notice that for $\theta < -5$, the dispersion increases. However, this would not affect the practical correction of experimental data for several reasons: first, at those temperatures well below $T_C$ the magnetocaloric response is small and it is usually not measured due to the lack of technological interest and distance from the critical region; second, as the AN equation of state is only valid in the vicinity of the phase transition, we must not expect that it gives an accurate description of $\Delta S_M$ far away from $T_C$,[27] with some
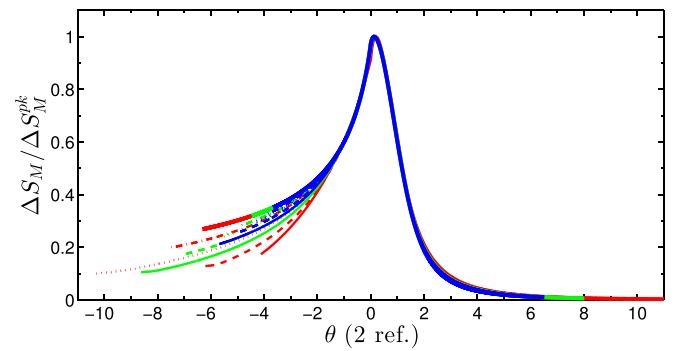


FIG. 2. All $\Delta S_M(T)$ curves from Figure 1 collapse onto the universal curve after the appropriate re-scaling.

additional phenomena not related to scaling taking place in that region.

Finally, we study how the demagnetizing factor affects the field dependence of $\Delta S_M^{pk}$ and $RC$. The field dependence of these quantities is very important because it allows us to have useful information of the magnetothermal properties of the studied materials when it is not possible to carry out experiments at high applied fields. For this reason, there have been several attempts to clarify the field dependence of the MCE properties.[13,14,16,19] Although it was pointed out that field dependence of $\Delta S_M^{pk}$ keeps unchanged in the presence of demagnetizing field,[11] if we study a wider range of applied fields we can distinguish three different regimes of influence as we show in Figure 3. For high enough applied fields with $H \gg N_D M$, we find that the field dependence is completely unaltered. However, if we compare the case without demagnetizing field ($N_D = 0$) to cases in which $N_D \neq 0$ we find that, for low applied fields ($H \ll N_D M$), the exponent of the field dependence which is found ($n = 2$) is clearly different from the ideal case ($n = 1 + (\beta - 1)/(\beta + \gamma)$). At low enough fields, this separation from the ideal behavior is found for all $N_D$ values but, as it would be expected, for higher $N_D$ the deviations are reached before. In the middle region where $H \sim N_D M$, we find a transition regime in which $n$ is not constant and it slowly varies from the ideal $n$ value to $n = 2$. This result is important in the sense that the transition region corresponds to applied fields which are in the usual work range ($\sim 1$ T). So it is necessary to take into account the effects of demagnetizing field if we
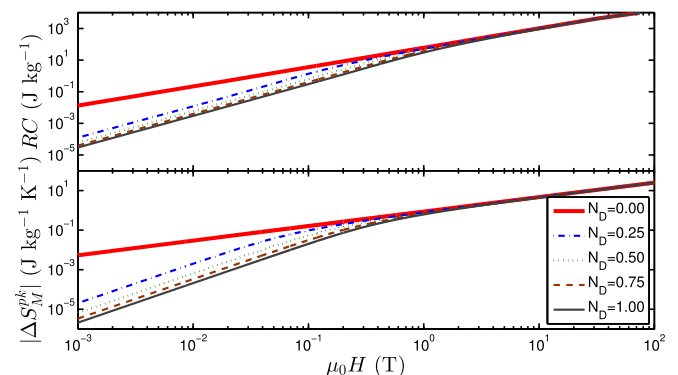


FIG. 3. Field dependence of $\Delta S_M^{pk}$ and $RC$ for different values of the demagnetizing factor.

are going to extrapolate results from the experiment using the habitual field dependence relations.[19]

However, to have some usefulness, these simulations should be checked against experimental data. For that purpose, an amorphous ribbon of composition $Fe_{75}Nb_{10}B_{15}$ has been ball milled in order to obtain powder particles with an average size of 20 $\mu$m and spheroidal form. Their isothermal magnetization curves have been measured in a Vibrating Sample Magnetometer and data processed using Maxwell relation (Eq. (1)). All experimental results are expressed with respect to the applied field used in the experiments. The low packing fraction of the powders and the external shape of the sample give an estimate of the demagnetizing factor of the order of 1/3. $\Delta S_M$ curves (Fig. 4) evidence the typical caret-like behavior of second order phase transition materials. Fig. 5 shows the construction of the universal curve of these experimental data using $h = 0.6$ and the field and temperature dependence of the exponent $n$. These experimental results show the following features: (a) even at the Curie temperature of the alloy, the demagnetizing field produces a field dependence of $n$, which decreases with increasing field, being this evolution smaller as the field increases; (b) at low temperatures, $n$ can have a value larger than 1, but with increasing field $n$ tends to 1; (c) the universal curve constructed using two reference temperatures allows for correction of the distortions produced by the demagnetizing factor in the environment of the Curie temperature; and (d) for low $\theta$ values, there is a deviation from the universal behavior, which decreases with increasing applied field. All these features are in full agreement with the results of our numerical simulations.

In summary, we have analyzed the MCE of a material with a second-order phase transition using the AN equation of state to reproduce its magnetothermal properties. We have seen that demagnetizing field clearly affects the $\Delta S_M$ curves, especially for $T < T_C$ where they slightly decrease ($\sim 5\%$). Although this variation is not too large, the exponent $n$ has a very different behavior in the same temperature region, when demagnetizing field effects appear. The limit of $n = 1$ for $T \ll T_C$ is not reached and the exponent becomes dependent of applied field and demagnetizing factor. We have shown how demagnetizing field effects can be corrected using the universal magnetic entropy change curve with two reference temperatures. Finally, we have determined a modification in the field dependence of the magnetic entropy
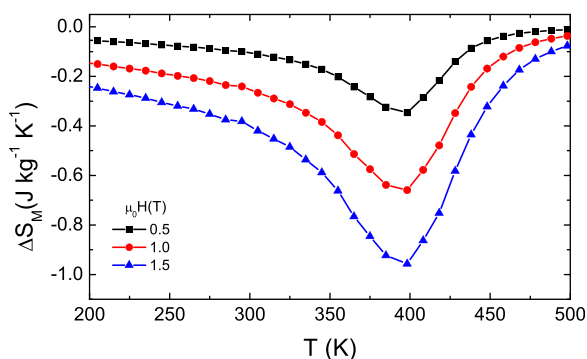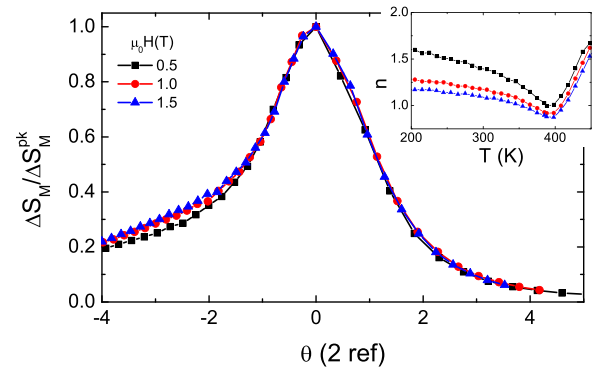


FIG. 5. Universal curve of $\Delta S_M$ of the data of Fig. 4. Inset shows the temperature and field dependence of exponent $n$.

change maximum and in the refrigerant capacity for applied fields of the same order of the demagnetizing field.

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FIG. 4. Temperature dependence of $\Delta S_M$ for an amorphous $Fe_{75}Nb_{10}B_{15}$ powdered alloy.