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3D-XY critical behavior of CsMnF$_3$ from static and dynamic thermal properties

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Received 5 December 2013, revised 3 January 2014
Accepted for publication 7 January 2014
Published 13 February 2014

Abstract

Static and dynamic critical behavior of the easy-plane antiferromagnet CsMnF$_3$ have been studied by means of a high-resolution ac photopyroelectric calorimeter. Thermal diffusivity, thermal conductivity and specific heat have been carefully measured in the near vicinity of the antiferromagnetic to paramagnetic transition (51.1 K). Specific heat and thermal diffusivity show singularities at the Néel temperature while thermal conductivity does not. Both the static and dynamic critical parameters agree with the standard 3D-XY universality class ($\alpha = -0.014$, $A^+/A^- = 1.06$); for specific heat $\alpha = -0.016$, $A^+/A^- = 1.09$ and for thermal diffusivity $b = -0.010$, $U^+/U^- = 1.09$. As the dynamic critical behavior of thermal diffusivity has not yet been theoretically established for the 3D-XY universality class, an approximate equation relating static and dynamic critical parameters has been obtained for it, leading to $b \approx \alpha$ and $A^+/A^- \approx U^+/U^-$ by studying the asymptotic behavior of the functions. This equation has also been experimentally verified for another XY antiferromagnet (SmMnO$_3$). As an easy-plane antiferromagnet with a hexagonal structure, CsMnF$_3$ could have been expected to comply with the 3D-XY chiral class ($\alpha = +0.34$, $A^+/A^- = 0.36$) as is the case of CsMnBr$_3$), but the experimental results rule out that possibility. This is attributed to the presence of a small in-plane anisotropy of the spins in CsMnF$_3$, which breaks the chiral degeneracy of the 120° spin structure.

Keywords: critical behavior, calorimetry, photopyroelectric, thermal diffusivity

(Some figures may appear in colour only in the online journal)

1. Introduction

Renormalization group theory has thoroughly studied static critical behavior in magnetic systems of different symmetry: isotropic, planar or uniaxial, stating that the critical behavior only depends on the dimensionality of the system ($d$) and on the degree of freedom of the order parameter ($n$). The corresponding universality classes in bulk magnets in three spatial dimensions ($d = 3$) together with their critical parameters are well established, as shown in table 1 for the three main classes: 3D-Heisenberg ($n = 3$), 3D-XY ($n = 2$), and 3D-Ising ($n = 1$) [1–3]. Experimental verification of this static critical behavior can be easily found in literature for both Heisenberg and Ising classes, confirming the predictions of the theory. Unlike them, experimental verification of the XY class is scarce.

<table>
<thead>
<tr>
<th>Universality class</th>
<th>$\alpha$</th>
<th>$\beta$</th>
<th>$\gamma$</th>
<th>$\nu$</th>
<th>$A^+/A^-$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3D-Heisenberg</td>
<td>$-0.115$, $-0.13$</td>
<td>0.36</td>
<td>1.39</td>
<td>0.71</td>
<td>1.52</td>
</tr>
<tr>
<td>3D-Ising</td>
<td>0.11</td>
<td>0.33</td>
<td>1.24</td>
<td>0.63</td>
<td>0.52</td>
</tr>
<tr>
<td>3D-XY</td>
<td>$-0.014$</td>
<td>0.34</td>
<td>1.30</td>
<td>0.66</td>
<td>1.06</td>
</tr>
<tr>
<td>Chiral XY</td>
<td>0.34</td>
<td>0.25</td>
<td>1.13</td>
<td>0.54</td>
<td>0.36</td>
</tr>
</tbody>
</table>

On the other hand, dynamic critical behavior has not attracted so much attention among the scientific community, either from the theoretical or from the experimental point of view. This is especially relevant when studying thermal transport properties such as thermal conductivity ($K$) and...
thermal diffusivity \((D)\). Theory is not well developed and there are not too many experimental evidences. Hohenberg and Halperin developed a comprehensive critical dynamics theory along the span of ten years by means of renormalization group theory [4–6], establishing a series of models (A–H) corresponding to different physical systems. Within the framework of that development, an Ising Hamiltonian corresponds to models A or C, a Heisenberg Hamiltonian to model G and an XY Hamiltonian to models E–F. Unfortunately, for the particular case of thermal diffusivity, its critical behavior is only theoretically developed in the case of model C but no conclusions can be derived from that comprehensive critical dynamics theory in the case of Heisenberg or XY systems.

More recently, some efforts have been made at predicting the values of the critical parameters for \(D\) and \(K\) and comparing them to experimental results, outside the renormalization group theory. Marinelli et al. studied both an Ising (FeF_3) [7] and a Heisenberg system (RbMnF_3) [8] and proposed a phenomenological rule for both systems where \(b = -|\alpha|, \ alpha\) being the critical exponent of \(D\) and \(\alpha\) the one for specific heat \(c_p\). In that work, the Ising case was theoretically supported on the basis of Hohenberg and Halpering theory. Pawlak studied the critical behavior of thermal diffusivity for the Ising case using a phenomenological–hydrodynamical approach [9], also concluding that \(b = -|\alpha|\) while Salazar et al. showed that this rule could be justified both for Heisenberg and Ising cases, studying the asymptotic behavior of the critical expressions for the inverse specific heat and its relation with thermal diffusivity [10]; the results were consistent with the critical behavior of KMnF_3 experimentally obtained fitting both specific heat and thermal diffusivity. No study of this kind has been published concerning the XY universality class yet.

The reason why there are very few experimental evidences concerning dynamic thermal properties lies in the conflict between the need for producing thermal gradients for thermal transport measurements and the need for keeping them as small as possible so as not to destroy critical information. The ac photopyroelectric calorimetry is specially suited to study dynamic critical behavior in the close vicinity of a phase transition since very small temperature gradients in the sample can be created, producing a very high signal to noise ratio, allowing thermal properties to be measured with great accuracy. The usefulness of this technique has been well demonstrated in the study of the critical behavior of second-order magnetic transitions in different materials, such as FeF_2 [7], RbMnF_3 [8], Cr_2O_3 [11], NiO [12], KMnF_3 [10], La_{1−\delta}Sr_{\delta}MnO_3 [13]; see the paper by Zammit et al. for an in-depth review of its applications [14].

In the case of \(d = 3, n = 2\) (3D-XY universality class), there are very few experimental evidences of any kind of phase transitions in materials or compounds definitely belonging to this class: superfluid transition of ^4He [6], superconductors such as RBa_2Cu_3O_7−\delta (R = Gd, Y, Nd, . . .) [15–17], and liquid crystals [18]. Concerning magnetic transitions, only SmMnO_3 [19] and some hexagonal ABX_3 compounds (B being a magnetic ion) have been experimentally shown to fit in this class [20, 21].

CsMnF_3 belongs to this last broad family of hexagonal ABX_3 compounds (where B stands for magnetic ions such as Ni, Cr, Mn, etc). In this class of materials, the magnetic ions form linear chains along the c axis which are coupled antiferromagnetically forming triangular layers in the basal a–b plane. Most of them undergo magnetic phase transitions into a three-dimensionally ordered state at low temperatures driven by the weak interchain coupling. The combination of the triangular-lattice structure and the antiferromagnetic nature of the interchain coupling can give rise to magnetic frustration, which is expected to play a major role in their magnetic structure [22]. Depending on the type of their magnetic anisotropy, the magnetic ordering can be roughly classified into two subclasses. If there is an easy-axis anisotropy (CsMnI_3, CsNiCl_3), the compound exhibits two successive transitions in zero applied field, marking the ordering of \(z\) and xy-components of the sublattice magnetization at \(T_{N1}\) and \(T_{N2}\) (\(T_{N1} > T_{N2}\)) [23, 24]. While if there is an easy-plane anisotropy (CsMnBr_3, CsMnF_3), the compound presents a single transition in zero applied field at \(T_N\) [25, 26]. Theoretical development done by Kawamura using Monte Carlo simulations and renormalization group analysis predicted that the chiral degeneracy associated with the helical spin ordering should give rise to a new universality class (chiral XY) with new parameters (see table 1) [27, 28]. In particular, \(\alpha = 0.34\) instead of the classical value of \(\alpha = −0.014\) for 3D-XY. Experimentally, this has been verified for CsMnBr_3 at zero field [22, 29] while the easy-axis anisotropy compounds CsMnI_3 and CsNiCl_3 fulfill this prediction only with an applied field which merges both transitions into one [30, 31]. At zero field they show a classical 3D-XY behavior [22].

CsMnF_3 has been less studied than CsMnBr_3, CsMnI_3, CsNiCl_3. As already explained, its magnetic structure (easy-plane magnetization with a very weak anisotropy in that plane) clearly suggests that it should belong to the 3D-XY universality class (chiral or not) or be very close to it [26]. Nevertheless, the only experimental evidence present in literature gives values of the critical parameters close to the Heisenberg class \(\alpha = −0.108, A/\alpha = 1.37\), fitting a reduced temperature range of only \(10^{-3} < |t| < 10^{-1}\) [32]. In the same work, performing a fitting using only points in the temperature range \(|t| < 1.5 \times 10^{-2}\) (but getting no closer than \(10^{-3}\)), it was claimed that a critical exponent \(\approx 0\) could be found. This was interpreted as evidence of a crossover from Heisenberg to XY universality class. But there was no evidence of the quality of that second fitting and, besides, the range fitted is not close enough to the critical temperature in order to extract sensible conclusions.

Another work on CsMnF_3 is the one by Shapira et al. where the prediction of an XY to Ising crossover with the application of a small magnetic field in the easy magnetic plane was studied, measuring the evolution of \(T_c\) with the applied field [33]. But in that work it was simply assumed that the critical behavior at \(H = 0\) is XY, it was not verified. The shape of the \(T_c (H)\) curves showed a particular shape which was interpreted as the natural break of the easy-plane and the favoring of a particular direction, leading to the Ising class. Both works were performed before the chiral XY classes were predicted for these materials. These results support the idea that the
critical behavior of CsMnF$_3$ is far from clear, and is still an open question.

Thus, the purpose of this work is twofold: firstly, to study the critical behavior of CsMnF$_3$ by means of high-resolution ac photopyroelectric calorimetry, extracting the critical exponent for specific heat, which is the best one to discriminate among the models as the values are much more different than in the case of the rest of the parameters (see table 1); besides, it is accompanied by the ratio of the critical parameters $A^+/A^-$ which is also very different among the classes. Secondly, to show that dynamic thermal properties such as thermal diffusivity can also be used to study the critical behavior in the case of the 3D-XY model.

2. Samples, experimental techniques and results

Single-crystal CsMnF$_3$ was prepared by the Czochralski method at the Kapitza Institute for Physical Problems at Moscow. A slice of thickness of about 0.73 mm was used for the present study. The sample was prepared so that it had well polished, plane-parallel surfaces.

As stated in section 1, high-resolution ac photopyroelectric calorimetry has been successfully used to extract thermal properties both from solid and liquid materials. In particular, using the standard back-detection configuration, simultaneous measurements of thermal diffusivity $D$ and specific heat $c_p$ can be carried out [34, 35]. As performed in the authors’ laboratory for the case of solid materials, in this configuration an acousto-optically modulated He–Ne laser beam of 5 mW illuminates the front surface of the sample under study. Its rear surface is in thermal contact with a 350 μm thick LiTaO$_3$ pyroelectric detector with Ni–Cr electrodes on both surfaces, by using an extremely thin layer of a highly heat-conductive silicone grease. The photopyroelectric signal is processed by a lock-in amplifier operating in the current mode. Both sample and detector are placed inside an adapted closed cycle He cryostat where measurements in the temperature range 15–320 K can take place. Cooling/heating rates can be performed from 100 mK min$^{-1}$ for measurements on a wide temperature range down to 10 mK min$^{-1}$ for high-resolution runs close to the phase transition.

For opaque and thermally thick samples (i.e. the thickness of the sample is higher than the thermal diffusion length $\mu = \sqrt{D/\pi f}$, where $f$ is the modulation frequency of the laser beam) the natural logarithm ($\ln V$) of the amplitude of the normalized photopyroelectric signal (which is obtained by dividing the measured signal by the signal provided by the bare detector) and its phase ($\Psi$) have a linear dependence on $\sqrt{f}$, with the same slope. From their slope $m$ and from the vertical separation between the two straight lines $d$, the thermal diffusivity and the thermal effusivity ($e = \sqrt{\rho c_p K} = K/\sqrt{D}$) of the sample can be obtained [36]

\[
D = \frac{\pi \ell^2}{m^2},
\]
\[
e = e_p \left( \frac{2}{\exp(d)} - 1 \right),
\]

where $\rho$ is the density, $\ell$ is the sample thickness and $e_p$ is the thermal effusivity of the pyroelectric detector.

Once thermal diffusivity and effusivity have been measured at a certain reference temperature ($T_{\text{ref}}$, $D_{\text{ref}}$, $c_{p\text{ref}}$), the temperature is changed while recording the amplitude and phase of the pyroelectric signal, at a fixed frequency. The temperature dependence of $D$ and $e$ are given by [36, 37]:

\[
D(T) = \left( \frac{1}{\sqrt{D_{\text{ref}}} - \frac{\Delta(T)}{\ell \sqrt{\pi f}}} \right)^2,
\]
\[
e(T) = e_p(T) \left( \frac{1 + \frac{c_{p\text{ref}} (T_{\text{ref}})}{\exp[\Delta'(T)]} - 1 \right),
\]

where $\Delta(T) = \Psi(T) - \Psi(T_{\text{ref}})$, $\Delta'(T) = \ln V(T) - \ln V(T_{\text{ref}})$ and $\Delta''(T) = \Delta'(T) - \Delta(T)$. It is worth noticing that, with this technique, thermal diffusivity is measured in the direction perpendicular to the sample surface.

Finally, the temperature dependence of specific heat and thermal conductivity are calculated from the following relations:

\[
e_p(T) = \frac{e(T)}{\rho \sqrt{D(T)}},
\]
\[
K(T) = e(T) \sqrt{D(T)}.
\]

The thermal diffusivity as a function of temperature in a long temperature range is presented in figure 1. The general evolution is quite typical, with high values at low temperature due to the longer length of the phonons’ mean free path, quickly diminishing as temperature increases. From 100 K to room temperature (not shown in the figure) there is still a soft but steady reduction till it reaches the room temperature value of 0.7 mm$^2$s$^{-1}$. The antiferromagnetic transition is manifested as a dip superimposed on the thermal diffusivity curve, as it commonly happens with second-order magnetic phase transitions. The Néel temperature is 51.1 K, in accordance with measurements by Shapira et al [33].

In order to study the critical behavior in detail, thermal diffusivity, specific heat and thermal conductivity curves in the near vicinity of the Néel temperature have been carefully measured with high resolution. These are shown in figure 2. Specific heat and thermal conductivity are always noisier than thermal diffusivity using this technique, as the latter
Figure 2. Thermal diffusivity, thermal conductivity and specific heat as a function of temperature in the vicinity of the magnetic transition for CsMnF$_3$.

is obtained only using the phase of the photopyroelectric signal while for the former two both amplitude and phase are needed. Specific heat shows the usual lambda-shape associated with second-order phase transitions. It is worth noting that thermal conductivity does not present a singularity at the Néel temperature.

3. Fitting procedure and discussion

In the case of specific heat, the experimental specific-heat data have been fitted to the well-known relation:

$$c_p = B + Ct + A^\pm |t|^{-\alpha} \left(1 + E^\pm |t|^{0.5}\right),$$  (7)

where $t = (T - T_N) / T_N$ is the reduced temperature, $T_N$ the critical temperature, and $\alpha$, $A^\pm$, $B$, $C$ and $E^\pm$ are adjustable parameters. Superscripts $+$ and $-$ stand for $T > T_N$ and $T < T_N$ respectively. The linear term represents the background contribution to the specific heat, while the last term is the anomalous contribution to the specific heat. The factor in parentheses is the correction to scaling that represents a singular contribution to the leading power as known from experiments and theory [11, 38].

The experimental data were simultaneously fitted for $T > T_N$ and $T < T_N$ with a Levenberg–Marquardt method. First of all, we selected a fitting range close to the transition while avoiding the rounding part, and kept fixed the value of $T_N$. We performed a first fitting without the correction to scaling term and obtained a set of adjusted parameters. Afterward, we tried to increase the number of data points included in the fitting, first fixing $t_{\text{min}}$ and increasing $t_{\text{max}}$, and then fixing $t_{\text{max}}$ and decreasing $t_{\text{min}}$. The next step was to introduce the correction to scaling term in order to improve the fitting. As a last checking, we let $T_N$ be a free parameter in order to confirm the fitting. In the whole process, we focused our attention on the root mean square value as well as on the deviation plot, which is the difference between the fitted values and the measured ones as a function of the reduced temperature.

The fitting curves are superimposed onto the experimental data points in figure 3(a), in which the dots correspond to the experimental data points (not all of them have been presented, for the sake of clarity) and the continuous lines of the best fitting to equation (6). As can be seen in figure 3(a), the fitting is very good. In order to better evaluate the quality of the fittings, figure 3(b) shows the deviation plots of the fitting with respect to the experimental curve. Again, not all points are presented, for the sake of clarity.

The fitting ranges are in all cases limited by the rounding in the curves; this rounding is inherent to the samples and not attributable to the technique, as shown elsewhere [19].

The values of the critical parameters, the fitting ranges and the quality of the fittings given by the root mean square value are presented in table 2. The values of the critical exponent $\alpha = -0.016 \pm 0.004$ and the ratio of the coefficients $A^+ / A^- = 1.09$ agree very well with the theoretical values for the 3D-XY universality class ($\alpha = -0.014$, $A^+ / A^- = 1.06$). It is worth mentioning that the correction to scaling factors $E^\pm$ are very small (0.16 and 0.13) supporting the good quality of the fitting.
Following relation can be obtained:

$$T = T_N = \frac{\alpha}{D}$$

which shows us that, if thermal conductivity $K$ has no singularity (as it is the case) the critical behavior of $c_p$ and $1/D$ must be the same. Thus, we have performed a second fitting of the experimental results of $1/D$ to the equation

$$\frac{1}{D} = B' + C'T + A^{\pm} |t|^{-\alpha} \left(1 + E^{\pm} |t|^{0.5}\right). \quad (9)$$

using an analogous procedure to the one followed for specific heat. Figures 3(c) and (d) show the fitting and the deviation plots to assess the quality of the fitting, which is better than the one for specific heat as the noise in the experimental curve is smaller.

The values of the critical parameters, the fitting ranges and the quality of the fittings given by the root mean square value are also presented in table 2. The values of the critical exponent $\alpha = -0.013 \pm 0.002$ and the ratio of the coefficients $A^+/A^- = 1.06$ agree even better with the theoretical values for the 3D-XY universality class ($\alpha = -0.014, A^+/A^- = 1.06$). Again, the correction to scaling factors $E^{\pm}$ are very small (0.15 and $-0.021$), expressing the good quality of the fitting.

Lastly, as pointed out in section 1, it is interesting to explore the possibility of using directly the thermal diffusivity curve to extract information about the critical behavior of the magnetic transition. A similar equation to equation (6), used for specific heat, can be written, with its own critical parameters

$$D = V + Wt + U^{\pm} |t|^{-b} \left(1 + F^{\pm} |t|^{0.5}\right). \quad (10)$$

Following the same fitting procedure as in the previous magnitudes, the obtained results are presented in figures 3(e) and (f). The values of the critical parameters, the fitting ranges and the quality of the fittings given by the root mean square value are presented in table 2. Once again, the correction factors $F^{\pm}$ are small enough so as to consider it a very good fitting (0.34 and 0.62). The values of the critical exponent $b = -0.010 \pm 0.004$ and the ratio of the coefficients $U^+/U^- = 1.09$ agree with the theoretical values for the 3D-XY universality class ($\alpha = -0.014, A^+/A^- = 1.06$). But the question is whether this has any kind of theoretical support for this class of universality.

In section 1 it has already been explained that there is no such theoretical support coming from renormalization group theory. As follows from equation (8), finding an analytical

### Table 2. Critical parameters, fitting ranges, quality of the fittings (given by the root mean square value). The values for the specific heat in SmMnO$_3$ are taken from [19].

<table>
<thead>
<tr>
<th>Material</th>
<th>Thermal property</th>
<th>$\alpha$</th>
<th>$A^+/A^-$</th>
<th>$t_{\text{min}}$ - $t_{\text{max}}$</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SmMnF$_3$</td>
<td>$c_p$ (J kg$^{-1}$ K$^{-1}$)</td>
<td>-0.016 ± 0.004</td>
<td>1.09</td>
<td>$9 \times 10^{-2}$ - $8.8 \times 10^{-4}$</td>
<td>0.997 20</td>
</tr>
<tr>
<td>SmMnF$_3$</td>
<td>$1/D$ (s mm$^{-2}$)</td>
<td>-0.013 ± 0.002</td>
<td>1.06</td>
<td>$9 \times 10^{-2}$ - $1.6 \times 10^{-3}$</td>
<td>0.999 78</td>
</tr>
<tr>
<td>SmMnO$_3$</td>
<td>$c_p$ (J kg$^{-1}$ K$^{-1}$)</td>
<td>-0.013 ± 0.001</td>
<td>1.03</td>
<td>$8.3 \times 10^{-2}$ - $1.9 \times 10^{-3}$</td>
<td>0.998 04</td>
</tr>
<tr>
<td></td>
<td>$D$ (mm$^2$ s$^{-1}$)</td>
<td>-0.010 ± 0.004</td>
<td>1.09</td>
<td>$9 \times 10^{-2}$ - $1.5 \times 10^{-3}$</td>
<td>0.999 39</td>
</tr>
<tr>
<td></td>
<td>$D$ (mm$^2$ s$^{-1}$)</td>
<td>-0.017 ± 0.001</td>
<td>1.07</td>
<td>$8.2 \times 10^{-2}$ - $1.9 \times 10^{-3}$</td>
<td>0.999 86</td>
</tr>
</tbody>
</table>
relationship between the critical parameters of the specific heat and the critical parameters of thermal diffusivity is not possible, only approximate relationships can be found if we combine (6), (8) and equation (10) to give
\[
D = \frac{K}{\rho c_p} = \frac{K}{\rho B} \left(1 + \frac{C}{B} t \pm \frac{A}{B} |t|^{-\alpha}\right)^{-1}
\]
\[
= V + W t + U^\pm |t|^{-b},
\]
where we have neglected the correction to scaling term, which is only introduced to increase the fitting temperature range.

The next obvious step is to develop the left hand side of the equation in a Taylor series, as has been previously performed for the case of the Heisenberg class by Salazar et al to show that \(b = \alpha\), having neglected terms of the second order and higher [10]. But in the XY class \(|t|^{-\alpha}\) tends to zero more slowly than in the Heisenberg case \((\alpha = -0.01\) against \(\alpha = -0.115\)). Taking into account that \(C/B\) and \(A^\pm/B\) are of the same order of magnitude and the small value of \(\beta\), \(t \ll \frac{A^\pm}{B} |t|^{-\alpha}\) so we can neglect that term when the reduced temperature approaches zero, in the range \(10^{-2} < |t| < 10^{-5}\), and write
\[
D = \frac{K}{\rho c_p} = \frac{K}{\rho B} \left(1 + \frac{C}{B} t \pm \frac{A}{B} |t|^{-\alpha}\right)^{-1}
\]
\[
\approx \frac{K}{\rho B} \left(1 + \frac{A^\pm}{B} |t|^{-\alpha}\right)^{-1}
\]
\[
= \frac{K}{\rho B} \left(1 - \frac{A^\pm}{B} |t|^{-\alpha} + \frac{(A^\pm)^2}{B^2} |t|^{-2\alpha} - \frac{(A^\pm)^3}{B^3} |t|^{-3\alpha} - \ldots\right).
\]
Evaluating the weight of the second and third order terms in the range \(10^{-2} < |t| < 10^{-5}\), where we are performing our fittings, they can only be neglected if we are very close to the critical temperature. This means that we can only write, as a very approximate equation, the following
\[
\frac{K}{\rho B} \left(-\frac{A^\pm}{B} |t|^{-\alpha}\right) \approx U^\pm |t|^{-b},
\]
which must be taken as a tendency more than as an exact value. Nevertheless, from equation (13), the following conclusions can be inferred: \(b \approx \alpha\) and \(A^+/A^- \approx U^+/U^-\) (with opposite signs for \(A^\pm\) and \(U^\pm\)). The closer we can get in our fittings to the critical temperature, the more precise these expressions will be. Our experimental results agree well with this theoretical approach, so we can conclude that critical behavior can be studied by means of thermal diffusivity not only in the Heisenberg and Ising universality classes, but in the XY as well. As this is a phenomenological result, it would be interesting to see if it can be applied to any other XY case. The only available experimental results of this kind having shown an XY universality class are those of the manganite SmMnO\(_3\) published by Oleaga et al where it was shown [19], by fitting the specific heat, that its antiferromagnetic to paramagnetic transition belongs to that universality class, with critical exponents \(\alpha = -0.013 \pm 0.001\) and \(A^+/A^- = 1.03\). We have retrieved the experimental curve for thermal diffusivity obtained at the same time as the specific heat and fitted it to equation (10). Figure 4 shows the experimental points, the fitting and the deviation curves. The values of the critical parameters, the fitting ranges and the quality of the fittings given by the root mean square value are presented in table 2. The obtained results for the critical parameters have been \(b = -0.017 \pm 0.001\) and \(U^+/U^- = 1.07\), which neatly satisfy the approximate relations between critical parameters, confirming the validity of our phenomenological approach. Of course, a complete theoretical development of dynamic critical behavior for thermal diffusivity in the 3D-XY universality case would be desirable in order to fully validate these results.

To sum up, the fittings performed on specific heat, inverse of thermal diffusivity and thermal diffusivity agree in assigning this magnetic transition to the 3D-XY universality class, in complete agreement with the magnetic structure of CsMnF\(_3\), as the spins lie on the basal plane with a very small in-plane anisotropy. Easy-plane anisotropic magnets correspond, theoretically, to the 3D-XY universality class. If we compare our results with those obtained by Ikeda [32], who suggested a crossover from the Heisenberg to the XY class, it can be said that the resolution in our measurements is of a higher quality and that we have been able to get closer to the critical temperature than in that work. Both features are fundamental in order to correctly describe the critical behavior of second-order phase transitions.

Besides, our results rule out the chiral XY class (where \(\alpha = +0.34\) and \(A^+/A^- = 0.36\)). In triangular antiferromagnets, the low temperature (ordered) state is the so-called ’120° spin structure’ in which the XY spins on three interpenetrating sublattices align within a plane forming 120° angles with neighboring spins on other sublattices. The ground state of such triangular XY spins is twofold degenerate according to whether the resulting non-collinear structure is right- or left-handed (chiral degeneracy) [28]. But the chiral symmetry of the spins has been shown to break in several cases, such as CsCuCl\(_3\); its lattice structure is distorted such that the anisotropic Dzyaloshinski–Moriya interaction arises between neighboring spins along the c-axis, reducing the spin symmetry.
from the perfect chiral one to a lower one \[39\]. Critical behavior of the hexagonal manganites RMnO\(_3\) (R = Ho, Er, Tm, Yb) also showed significant deviations from the chiral model \[19\].

In the particular case of CsMnF\(_3\), magnetic measurements show that the easy-plane ordering of the spins has a small in-plane anisotropy \[26\], which would be responsible for not achieving the perfect ‘120\(^{\circ}\) spin structure’, thus eliminating the possibility of the chiral degeneracy. The anisotropy is small enough so as not to become an easy-axis antiferromagnet such as CsMnI\(_3\), thus complying with the characteristics of the standard 3D-XY universality class.

\[4\]. Conclusions

Thermal diffusivity, specific heat and thermal conductivity have been measured for CsMnF\(_3\) (an easy-plane antiferromagnet) using a high-resolution ac photopyroelectric calorimeter. The antiferromagnetic to paramagnetic transition, which takes place at 51.1 K, has been studied in detail in order to retrieve its critical behavior. The static critical parameters obtained from specific heat (\(\alpha = -0.016, A^+/A^- = 1.09\)) and the inverse of thermal diffusivity (\(\alpha = -0.013, A^+/A^- = 1.06\)) agree very well with the standard 3D-XY universality class (\(\alpha = -0.014, A^+/A^- = 1.06\)). A chiral XY class is ruled out due to the small in-plane anisotropy of the spins, which would break the chiral degeneracy of the 120\(^{\circ}\) spin structure. It has also been shown that the dynamic critical behavior of thermal diffusivity experimentally agrees with the static critical behavior of specific heat (\(b = -0.010 \pm 0.004, U^+/U^- = 1.09\)). This is theoretically justified studying the asymptotic behavior of the critical equations, which leads to \(b \approx \alpha\) and \(A^+/A^- \approx U^+/U^-\) for the 3D-XY universality class; experimental results on another XY antiferromagnet (SmMnO\(_3\)) support this phenomenological approach. Further theoretical development of dynamic critical behavior for the 3D-XY universality class by means of renormalization group theory would be needed in order to fully solve this matter.

Acknowledgments

This work has been supported by the Ministerio de Ciencia e Innovación (MAT2011-23811), Gobierno Vasco (IT619-13), and UPV/EHU (UI111/55).

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