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DIFFUSE SECOND ORDER PHASE TRANSITION IN GADOLINIUM TRICHLORIDE (\(\text{(*)}\))

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Résumé. — Des mesures de grande résolution de la chaleur spécifique d’un monocristal de GdCl3 montre que la partie asymptotique de la chaleur spécifique est discontinue mais non singulière à \(T_c\) avec \(\alpha' = -0.45 \pm 0.12\) et \(\alpha = -0.18 \pm 0.08\). La courbure présente un maximum à environ 2 mK au-dessous de \(T_c\). Cet effet est attribué à l’existence de sous-systèmes présentant une distribution de température d’ordre.

Abstract. — High resolution measurements of the specific heat of a single crystal of GdCl3 indicate that the asymptotic form of the specific heat is discontinuous but non-singular at \(T_c\) with \(\alpha' = -0.45 \pm 0.12\) and \(\alpha = -0.18 \pm 0.08\). The observed rounding produces a maximum about 2 mK below \(T_c\); this effect is attributed to the existence of magnetic subsystems having a distribution of ordering temperatures.

During the past decade considerable interest has been aroused regarding the nature of the specific heat in the "critical region" of temperature near magnetic ordering points. With few exceptions experimental studies have yielded quite similar results: [1] although the specific heat is rounded quite near the ordering temperature \(T_c\).

(e. g. for \(\frac{c_p}{T} = |(T - T_c)/T_c| < 2 \times 10^{-3}\)), there is a region of temperature just outside \(\delta_6\) for which the specific heat actually appears to diverge. Initial measurements on GdCl3 hinted that an entirely different behavior might be found [2]. Instead of diverging the specific heat seemed to be the classical 2nd order type showing no singularity but merely being discontinuous at \(T_c\) and approaching \(T_c\) with finite gradients [3]. (Such transitions have often been observed in superconductors but not in magnetic systems.) Moreover, GdCl3 is a relatively simple system whose structure and interactions are well known. The Gd\(^{3+}\) ions are arrayed on a hexagonal close packed lattice, and EPR pair studies have shown that the magnetic interactions are dominated by isotropic nearest and next nearest neighbor exchange coupling although dipolar interactions are also significant [4].

We have made high resolution measurements of the specific heat of a large single crystal (\(\sim 40\) gm) GdCl3 in the critical region using the heat pulse technique with heating steps of about 100 \(\mu\)K near \(T_c\). The heat capacity of the sample holder was measured in a separate experiment, and the lattice contribution was extrapolated from the value for the isostructural, diamagnetic salt LaCl3. Our data for GdCl3 was corrected accordingly, the total correction being less than 0.3 \% over the entire range of measurement.

The experimental data showed a smooth, slightly rounded specific heat having a maximum at \(T_{max} = 2.2156\) K on our temperature scale. We then attempted to fit these results to the most general theoretical form [1]:

\[
C/R = A e^{-\alpha} + B
\]  

\((1)\)

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where the none of the constants were constrained to take on identical values below and above \(T_c\). Plots of \(\log (C/R - B)\) vs. \(\log \varepsilon\) should be linear with slopes equal to \(\alpha'\), \(\alpha\) for \(T < T_c\) \(T > T_c\) respectively. Unfortunately in this technique there are two parameters, \(T_c\) and \(B\), which may be varied; and since neither the entry into the critical region nor the onset of rounding are clearly defined, it is quite difficult to make a definitive analysis. From eq. (1), however, we can see that the number of variables can be reduced by examining the derivative of the specific heat rather than the specific heat itself: [5]

\[
\frac{\partial (C/R)}{\partial T} = \frac{A \alpha}{T_c} e^{-(\alpha + 1)}.
\]  

\((2)\)

Accordingly we have taken derivatives numerically and then plotted \(\log \left|\frac{\partial (C/R)}{\partial T}\right|\) vs. \(\log \varepsilon\) as shown in figure 1.

The value of \(T_c\) was varied on plots of \(T < T_c\) and \(T > T_c\), and we searched for that value of \(T_c\) that yielded the greatest range of linear behavior and the slope, which is \(-\alpha + 1\) indicates: \(\alpha' = -0.45, \alpha = 0.18\).
which produced the greatest range of linear behavior on both plots simultaneously. Note that $T_c$ is now the only variable at our disposal. Deviations from linearity for large values of $\varepsilon$, which are quite insensitive to changes in $T_c$, would indicate that these data were in the noncritical region. As figure 1 shows, our results lay totally within the critical region. The deviations for small $\varepsilon \sim 9 \times 10^{-4}$ are caused by rounding. The «best» behavior was obtained for a choice of $T_0 = 2.2178 \, ^\circ K$, i.e. 2.2 mK above $T_{\text{max}}$. The rounded region appeared to be relatively symmetric with respect to $T_c$, and it was impossible to reduce this region by changing $T_c$. From the slopes of these curves we find that $\alpha' = -0.45$ below $T_c$ and $\alpha = -0.18$ above $T_c$. This means that the asymptotic form for the specific heat is not singular, although the derivatives of the specific heat are infinite at $T_c$. The transition is therefore type 2b [3] or «diffuse». $T_c, \alpha', \alpha, A', \text{ and } A$ have all been determined directly from figure 1, and with the aid of eq. (1) and the data we calculated $B, B'$. Using these values we have plotted $\log (C/R - B)$ vs. $\log \varepsilon$ for $T < T_c, T > T_c$ in figure 2. The agreement between the asymptotic form and the data is excellent except in the «region of rounding» where the data points diverge from the calculated values.

Our results for $\alpha, \alpha'$ might not be the correct ones for an ideal GdCl$_3$ sample, for they could, in fact, simply be renormalized values due to any number of hidden variables [6]. However, the observed rounding must arise due to a different reason. If microscopic imperfections in a «single crystal» are severe enough to limit the maximum range of the correlations, the result could be an effective subdivision of the sample into an array of microcrystals. The micro-systems would not be identical and could have slightly different ordering temperatures. The specific heat of the entire sample would then be given by

$$C(T, T_c) = \sum_i C(T, T_{ci}) f(T_c, T_{ci})/\sum_j f(T_c, T_{cj})$$

where $C(T, T_c)$ is the asymptotic specific heat at $T$ and $f(T_c, T_{cj})$ the distribution function giving the fraction which order at $T_{cj}$. We have calculated the specific heat of our system using eq. (3) and a Gaussian distribution with half-width $\Gamma$ for $f(T_c, T_{cj})$. The same values for $A, B, \alpha$ were used in the expression for $C(T, T_c)$ since the specific heat is less sensitive to small changes in these parameters. For a half-width of $\Gamma = 1.5$ mK we reproduced the experimental results exactly (see Fig. 3). Moreover, outside of the

![Specific Heat of GdCl$_3$ Compared with Theory](image)

**Fig. 2.** — $\log [C/R-B]$ vs. $\log \varepsilon$ where $B' = 3.00, B = 0.94$; $T < T_c$ xxx, $T > T_c$ xxxx. Deviations from linearity near $\varepsilon = 9 \times 10^{-4}$ are due to the onset of rounding as discussed in the text. The scatter in the points arises from the fact that in the region of scatter $(C/R-B)$ is only about 10% of $C/R$.

rounded region the asymptotic form for the specific heat is unaffected. Because of the relatively good agreement obtained with other peaked distribution functions, we believe that results obtained with the true distribution function should agree well with our values. We conclude then, that the proposed distribution of ordering temperatures can account for the rounding in GdCl$_3$ and, in fact, could account for much of the observed rounding in other systems.

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References


[6] Fisher (see Ref. [7]) has shown that in a non-ideal system any number of «hidden variables» could convert the specific heat singularity to a cusp. If our values are actually renormalized, then the real values for an ideal system are $\alpha' \to 0.31$, $\alpha \to 0.15$ according to Fisher’s scheme for renormalization.
