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Experimental Test of the Extended-Scaling Hypothesis in the Spin-Flop System $CsMnBr_3 \cdot 2D_2O$

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This neutron-scattering study deals with the behavior of the order parameters $M_{st}^{\parallel}(H,T)$ and $M_{st}^{\perp}(H,T)$ in $CsMnBr_3 \cdot 2D_2O$. The data provide a first direct test of the extended-scaling theory near the bicritical point of a spin-flop system.

The (H,T) phase diagram for an antiferromagnet with weak orthorhombic spin anisotropy contains a bicritical point,[1] where two distinct types of critical behavior are simultaneously present, namely an ordering of the magnetic moments parallel to the easy axis in the antiferromagnetic (AF) phase, as well as an ordering perpendicular to it in the spin-flop (SF) phase. As the bicritical point is approached along either of the two critical lines separating the AF and SF phases from the paramagnetic (P) phase, an abrupt crossover is expected from Ising-like critical behavior (number of relevant spin components $n=1$) to XY-like bicritical behavior ($n=2$).[2] The crossover can be described by a scaling theory first introduced by Riedel and Wegner[3] and subsequently developed further by a so-called extended-scaling hypothesis.[4] On the basis of this hypothesis, which may be formulated[2,4] as the postulate that the Gibbs free energy and its derivatives are generalized homogeneous functions, the shape of the phase boundaries can also be predicted.

In the last few years, experimental studies on a number of spin-flop systems have been undertaken to verify the extended-scaling hypothesis and its implications. However, the results so far do not extend further than an accurate determination of the shape of the phase boundaries and a comparison of these data with the theoretical predictions.[5-7] Here we shall report the results of an extensive scaling analysis of the variation of the order parameters in the AF and SF phases, i.e., the components of the staggered magnetization parallel ($M_{st}^{\parallel}$) and perpendicular ($M_{st}^{\perp}$) to the easy axis, respectively, near the bicritical point in $CsMnBr_3 \cdot 2D_2O$ (CMB). To our knowledge such a direct experimental verification of the extended-scaling hypothesis for spin-flop systems has not been reported earlier.

CMB is a pseudo-one-dimensional ($d=1$) Heisenberg antiferromagnet.[8,9] The crystal structure is orthorhombic with space group $Pcca$. Below $T_N \simeq 6.30$ K the symmetry of the AF ordering is described by the magnetic space group $Pc'c'a'$.[10]

The application of a magnetic field $H \gtrsim 26$ kOe along the easy (b) axis brings the system in the SF state, where the moments are directed approximately parallel to the intermediate (c) axis.[11] The neutron-scattering experiments on CMB have been performed on a double-axis diffractometer at the Petten high-flux reactor. The disklike single crystal of $15 \times 15 \times 4$ mm^3 was mounted in a superconducting magnet with $\vec{H}$ parallel to the easy axis. In the experiments the scattering vector $\vec{\kappa}$ was confined to the a^*-c^* plane, as in this plane magnetic and nuclear scattering occurs at different reciprocal-lattice points. The order parameters in both the AF phase and the SF phase, i.e., $M_{st}^{\parallel}(H,T)$ and $M_{st}^{\perp}(H,T)$, could be obtained separately from the intensity of the magnetic reflections in the a^*-c^* plane,[12] which were recorded at a large number of fixed temperatures as a function of H. Figure 1(a) shows some typical results for $(M_{st}^{\parallel})^2$ and $(M_{st}^{\perp})^2$ close to the bicritical point, which have been used to verify the predicted scaling relations. Part of the corresponding (H^2,T) phase diagram is shown in Fig. 1(b).

The appropriate scaling fields for the analysis of the behavior close to a bicritical point are the two ordering (staggered) fields $H_{st}^{\parallel}$ and $H_{st}^{\perp}$, conjugated to the order parameters $M_{st}^{\parallel}$ and $M_{st}^{\perp}$, respectively, and two nonordering fields $\tilde{g}$ and $\tilde{t}$, which can be expressed in the more familiar variables H^2 and T.[13] [See Fig. 1(b).] Now the extended-scaling hypothesis can be formulated as a generalized-homogeneous-function postulate for the Gibbs free energy

$$G(H_{st}^{\parallel}, H_{st}^{\perp}, \tilde{g}, \tilde{t}) = |\tilde{t}|^{2-\alpha_b} \mathcal{G}_{\pm}\left\{ \frac{H_{st}^{\parallel}}{|\tilde{t}|^{\Delta_{\parallel}}}, \frac{H_{st}^{\perp}}{|\tilde{t}|^{\Delta_{\perp}}}, \frac{\tilde{g}}{|\tilde{t}|^{\varphi}} \right\}, \qquad (1)$$

or for its derivatives, as, for instance,

$$M_{st}^{\parallel,\perp}(\tilde{g},\tilde{t}) \equiv -\left(\frac{\partial G}{\partial H_{st}^{\parallel,\perp}}\right) = |\tilde{t}|^{\beta_b} \mathfrak{M}_{\pm}\left(\frac{\tilde{g}}{|\tilde{t}|^{\varphi}}\right), \qquad (2)$$

where we have set $H_{st}^{\parallel} = H_{st}^{\perp} = 0$. $\mathcal{G}$ and $\mathfrak{M}$ are so-called scaling functions, each consisting of two branches, indicated by the subscripts + and − for $\tilde{t} > 0$ and $\tilde{t} < 0$, respectively. The exponents α_b, β_b, φ, $\Delta_{\parallel}$, and $\Delta_{\perp}$ are defined as usual[2,4] and have bicritical values.

According to (2) the experimental data $M_{st}^{\parallel,\perp}(\tilde{g}, \tilde{t})$, scaled by $|\tilde{t}|^{\beta_b}$, should depend on only one single variable $x = |\tilde{g}|/w|\tilde{t}|^{\varphi}$. The constant w is introduced to normalize the variable x to unity at the paramagnetic phase boundaries, which for a system with orthorhombic spin anisotropy are given

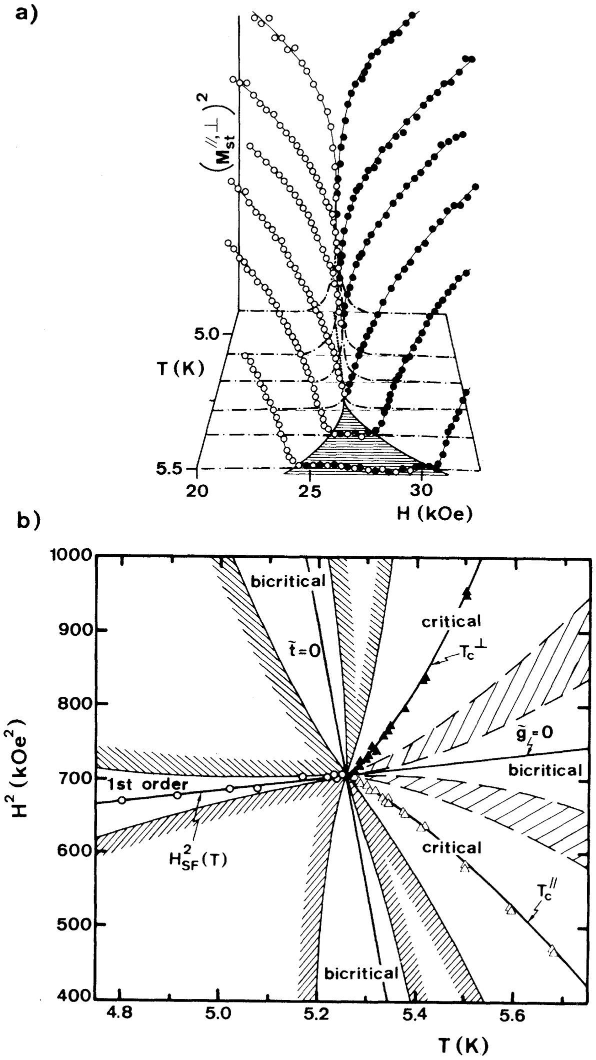

FIG. 1. (a) Typical field dependence of $(M_{st}^{\parallel})^2$ (open circles) and $(M_{st}^{\perp})^2$ (closed circles) close to the bicritical point. The continuation of the curves below their intersection point is indicated by a dash-dotted line. The shaded area and the dotted line represent the paramagnetic phase and the spin-flop line, respectively. (b) Part of the (H^2,T) diagram of CMB with the location of the $\tilde{g}$ and $\tilde{t}$ scaling axes. The shaded edges enclose the experimentally determined 25% crossover regions. For completeness, the crossover regions in the paramagnetic phase are also sketched.

by[4] $\tilde{g}_c = \pm w|\tilde{t}|^{\varphi}$. This scaling should result in a collapsing of the data for $\tilde{t} > 0$ on a curve $\mathfrak{M}_+^{\parallel,\perp}(x)$ and for $\tilde{t} < 0$ on a curve $\mathfrak{M}_-^{\parallel,\perp}(x)$. As the scaling function $\mathfrak{M}$ is expected to be universal,[4] in the present $n = 2$ spin-flop system the same scaling function should apply both to $M_{st}^{\parallel}(\tilde{g},\tilde{t})$ and $M_{st}^{\perp}(\tilde{g},\tilde{t})$. In addition it can be shown that the predicted critical behavior of $M_{st}^{\parallel,\perp}(\tilde{g},\tilde{t})$ along the scaling axes $\tilde{g}=0$ and $\tilde{t}=0$ close to the paramagnetic phase boundaries implies that $\mathfrak{M}_+^{\parallel,\perp}(x)$ and $\mathfrak{M}_-^{\parallel,\perp}(x)$ must display the following asymptotic behavior:

$$\mathfrak{M}_\pm^{\parallel,\perp}(x) \propto x^{\beta_b/\varphi}, \quad \text{for } x\to\infty ; \tag{3a}$$

$$\mathfrak{M}_-^{\parallel,\perp}(x) = \text{const}, \quad \text{for } x\to 0 ; \tag{3b}$$

$$\mathfrak{M}_+^{\parallel,\perp}(x) \propto (x-1)^{\beta}, \quad \text{for } x\to 1 , \tag{3c}$$

where β is the usual critical exponent.

In testing the above scaling relation, one can proceed in two ways. In the first approach one may vary all the parameters contained in the relation and process the experimental data according to (2) in order to find the optimum set of parameters which produce the expected data collapsing. In that case, one deliberately chooses to make no use of other available information. In our case the location and direction of the optimum scaling axes $\tilde{g}$ and $\tilde{t}$, the crossover exponent φ, and the constant w are coupled through the experimentally known shape of the critical phase boundaries $\tilde{g}_c=\pm w|\tilde{t}|^{\varphi}$, as we have shown in an earlier article.[14] Thus a change in, for instance, the location of the bicritical point (H_b,T_b), i.e., the origin of the $(\tilde{g},\tilde{t})$ coordinate system, entails a different set of φ and w values in order to retain the best description of the phase boundaries shown in Fig. 1(b). The *directions* of the $\tilde{g}$ and $\tilde{t}$ axes are hardly sensitive for such changes, as they are fully determined by the symmetry of the phase diagram. So, as a second method one may insert this relation between the parameters as a boundary condition and process the scaling data accordingly. In our testing procedure we have used both methods. Although it is difficult to quantify the criteria used in judging the data collapsing in the plots, we may state that the results obtained in both ways are essentially the same. The parameter set which gives the best results is

$$T_b=5.255(10)\text{ K},\quad H_b=26.55(3)\text{ kOe},$$
$$\varphi=1.20(3),\quad w=6.63(45)\times10^3\text{ kOe}^2, \tag{4}$$
$$\beta_b=0.34(2).$$

The estimated accuracy of these values is indicated between parentheses in units of the least significant digit. Earlier estimates for the first four values, obtained from the shape of the experimentally determined critical phase boundaries alone,[14] are in good agreement with this set. In Fig. 2 the experimental data, scaled as $\mathfrak{M}_\pm^{\parallel,\perp}(x)$ $=M_{st}^{\parallel,\perp}(\tilde{g},\tilde{t})/|\tilde{t}|^{\beta_b}$ with use of the parameter set (4), are presented as a function of $x-x_c$, with $x_c=1$ for $\mathfrak{M}_+$ and $x_c=0$ for $\mathfrak{M}_-$.

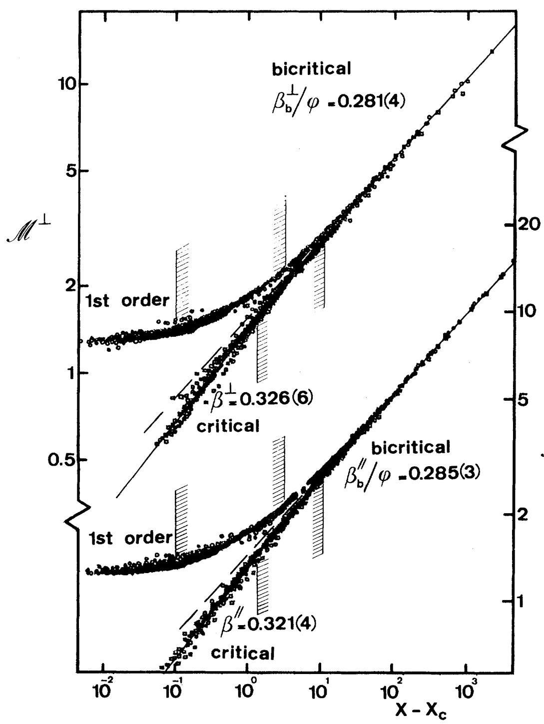

FIG. 2. Optimum collapsing of 5000 $M_{st}^{\parallel}$ and $M_{st}^{\perp}$ data points, scaled according to (2) to yield the scaling functions $\mathfrak{M}^{\parallel}$ and $\mathfrak{M}^{\perp}$, which both consist of two branches, viz. $\mathfrak{M}_+$ for $\tilde{t}>0$ and $\mathfrak{M}_-$ for $\tilde{t}<0$. $x_c=1$ for $\mathfrak{M}_+$ and $x_c=0$ for $\mathfrak{M}_-$. The parameter set (4) has been used. Solid lines represent the asymptotic behavior (3a) and (3c). The shaded edges enclose the 25% crossover regions.

In addition to the apparent data collapsing in Fig. 2, there is a striking symmetry between the $M_{st}^{\parallel}$ and $M_{st}^{\perp}$ data, as expected for an $n=2$ spin-flop system. The data are plotted on a double-logarithmic scale in order to display the asymptotic behavior in the critical and bicritical regions. The predicted behavior (3) appears to be satisfied indeed. Although the difference between the asymptotic slopes is only small, even the crossover from the critical behavior (3c) to the bicritical (3a) is observed, as is shown by the straight lines in the plot. Least-squares fits of the single-power laws (3a) to the data with $x-x_c \geqslant 30$ yield the slopes $(\beta_b/\varphi)_\perp=0.281(4)$ and $(\beta_b/\varphi)_\parallel=0.285(3)$ for $\mathfrak{M}^\perp$ and $\mathfrak{M}^\parallel$, respectively. Of course, these results may not be considered as independent estimates for β_b/φ, since the exponents β_b and φ separately have been used in the scaling procedure. However, these values demonstrate that the asymptotic behavior for $x \to \infty$ is described correctly by the exponent $\beta_b/\varphi = 0.34/1.20 = 0.283$, in agreement with (3a). Least-squares fits of (3c) to the data of $\mathfrak{M}_+^{\parallel,\perp}$ with $x - x_c \leq 1.4$ yield the exponents $\beta_\parallel = 0.321(4)$ and $\beta_\perp = 0.326(6)$. These values are completely consistent with the values $\beta_\parallel = 0.321(6)$ and $\beta_\perp = 0.326(7)$, which were determined in an independent way from the same data set.[14] With the asymptotic slopes indicated by α_1 and α_2, one can define 25% crossover regions as those regions where the local slope deviates more than $|\alpha_2 - \alpha_1|/4$ from either α_1 or α_2. These 25% crossover regions, centered around $x - x_c \simeq 5$ for $\mathfrak{M}_+$ and $x - x_c \simeq 0.7$ for $\mathfrak{M}_-$, are indicated both in Fig. 2 and in Fig. 1(b).

The values (4) of the bicritical exponents φ and β_b obtained from the scaling procedure are in good agreement with the theoretical predictions for an $n=2$ spin-flop system, viz. $\varphi = 1.175(15)$ (Ref. 4) and $\beta_b = 0.346(1)$, being the β value for a $d=3$ XY system.[15] The values for $\beta_\parallel$ and $\beta_\perp$ may be compared with the theoretical predictions for a $d=3$ Ising system, viz. $\beta = 0.311(7)$, an average value obtained from series expansions,[16] or $\beta = 0.325(1)$, the most recent estimate from ϵ expansions.[17] Especially the latter estimate compares very well with the experimental results.

A final comment is needed with regard to the accurate alignment of the magnetic field along the easy axis, which according to Rohrer and Gerber[5] is essential for a correct determination of bicritical exponents. In the present experiment the misalignment angle of $\vec{H}$ in the b-c (easy-intermediate) plane ($\psi \simeq 0.5$) exceeds the critical angle $\psi_c(T=0) \simeq 0.08°$, which corresponds to the maximum width of the first-order spin-flop "shelf."[5] One may wonder why this misalignment does not affect the results of the present analysis, whereas it has a pronounced influence on the observed variation of $M_{st}^\parallel$ and $M_{st}^\perp$ close to the spin-flop transition[12] and on the shape of the phase diagram.[14] In our opinion, this difference is related to the fact that in the logarithmic presentation of $\mathfrak{M}$ in Fig. 2, the asymptotic bicritical behavior close to the $\tilde{t} = 0$ axis is emphasized, which appears to be hardly sensitive to ψ. In contrast, also the alternative scaling function $\mathfrak{M}^{\parallel,\perp}(\tilde{t}/|\tilde{g}|^{1/\varphi}) \equiv M_{st}^{\parallel,\perp}/|\tilde{g}|^{\beta_b/\varphi}$ can be constructed, in which the behavior close to the $\tilde{g} = 0$ axis is stressed. Although again good data collapsing is found with the same parameter set (4), the expected asymptotic behavior of $\mathfrak{M}$ for $\tilde{g} \simeq 0$ is not observed. A more extensive report of the experiment and analyses will be published elsewhere.

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