Chiral Critical Exponents of the Triangular-Lattice Antiferromagnet CsMnBr$_3$ as Determined by Polarized Neutron Scattering

V.P. Plakhy,$^1$ J. Kulda,$^2$ D. Visser,$^{3,5,6}$ E. V. Moskvin,$^1$ and J. Wosnitza$^4$

$^1$Petersburg Nuclear Physics Institute RAS, Gatchina, St. Petersburg, 188350 Russia
$^2$Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex 9, France
$^3$Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom
$^4$Physikalisches Institut, Universität Karlsruhe, 76128 Karlsruhe, Germany
$^5$CLRC, ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, United Kingdom
$^6$IRI, TU Delft, Mekelweg 15, 2629 JB Delft, The Netherlands

(Received 30 May 2000)

The critical exponents $\gamma_c = 0.84(7)$ of the chiral susceptibility above the Néel temperature, $T_N$, and $\beta_c = 0.44(2)$ of the average chirality below $T_N$ have been determined for the triangular-lattice antiferromagnet CsMnBr$_3$ by means of polarized neutron scattering. These first experimental values of chiral critical exponents are in line with theoretical predictions and fulfill their scaling relation. The temperature at which the average chirality appears coincides with the spin-order transition temperature, $T_N$.

PACS numbers: 75.25.+z, 75.40.Cx, 75.50.Ee

The frustrated triangular-lattice antiferromagnets (TLA) have attracted considerable interest because of Kawamura’s conjecture on the existence of new chiral-universality classes of their spin-order transitions (see Ref. [1] and references therein). Specifically, for an easy-plane ($XY$) anisotropy as in CsMnBr$_3$, the frustration of the three nearest-neighbor Mn$^{2+}$ spins ($S = 5/2$) on a triangular plaquette in the hexagonal basal plane is resolved by a $120^\circ$ rotation of neighboring spins. There is a twofold degeneracy (Z$_2$) because the spin rotation may either be clockwise or counterclockwise when going around a triangle (Fig. 1). These two chiral states can be characterized by a two-point variable, the chirality $C = \langle S_i \times S_j \rangle$, where $S_i$ and $S_j$ are neighboring spins. For a chiral $XY$ system, $C$ is either parallel or antiparallel to the hexagonal $z$ axis. This additional twofold degeneracy changes the symmetry of the order parameter from SO(2) for an unfrustrated $XY$ magnet to Z$_2 \times$ SO(2). It was this modified symmetry which motivated the notion of new universality classes [2]. This conjecture is, however, still the subject of controversial debates. Based on Monte Carlo data [3] and on a nonlinear sigma model [4], a tricritical scenario was favored. Others suggest a weakly first-order transition which becomes apparent only very close to the critical temperature $T_N$, whereas further away from $T_N$ the data may be described by effective exponents [5–7]. An experimental indication for the latter scenario was found recently [8].

In any case, the chirality changes the critical behavior. Besides strongly modified conventional exponents $\alpha$, $\beta$, $\gamma$, and $\nu$ of the specific heat, the staggered magnetization, the susceptibility, and the correlation length, respectively, additional critical exponents appear. These new exponents, $\beta_c$, $\gamma_c$, and $\nu_c$ of the average chirality, the chiral susceptibility, and the chiral correlation length, respectively, describe how the chiral order develops and are unique to this class of phase transitions. Until now, no experimental values for the chiral critical exponents have been reported. Only the conventional ones have been measured, and their values were in reasonable agreement with those obtained by Monte Carlo simulations [1]. For CsMnBr$_3$, the experimental data are $\alpha = 0.39(9)–0.40(5)$ [9,10], $\beta = 0.22(2)–0.25(1)$ [11,12], $\gamma = 1.01(8)–1.10(5)$ [11,12], and $\nu = 0.54(3)–0.57(3)$ [11,12] in comparison with the Monte Carlo ones, $\alpha = 0.34(6)–0.46(10)$ [3,13], $\beta = 0.24(2)–0.253(10)$ [3,13], $\gamma = 1.03(4)–1.13(5)$ [3,13], and $\nu = 0.48(2)–0.54(2)$ [3,14].

An important question associated with the chirality is whether the spin and the chiral order occur simultaneously. In other words, the question is whether these two variables are components of a single order parameter or whether they are decoupled. For the two-dimensional (2D) $XY$ model, Monte Carlo simulations showed a spin-ordering phase transition occurring about 7% above the chiral critical temperature [15]. However, for the 3D $XY$ TLA, this difference was found to be less than $4 \times 10^{-4}$. Still, the authors claimed that the two variables might be decoupled based on a slight difference in the calculated critical exponents of the correlation lengths, $\nu = 0.50(1)$ and $\nu_c = 0.55(1)$ [3]. (Compare with $\nu_c/\nu = 2.85$ in the 2D case.) However, if the chirality would be decoupled from the spin a completely different criticality would be expected, i.e., a 3D Ising universality for the chiral phase transition [1].

![FIG. 1. A right-handed (a) and left-handed (b) chiral spin structure for a TLA, such as CsMnBr$_3$.](image-url)
These arguments could not be verified since no experimental technique was known to extract the chirality and chiral susceptibility.

The principal challenge to study the critical behavior of the chiral susceptibility is the necessity to investigate four-spin correlations which cannot be measured directly. An elegant way to circumvent this problem was proposed by Maleyev [16] who suggested studying the dynamical chirality, i.e., the projection of the chiral-fluctuation field onto the magnetization induced by an external magnetic field. Thereby, the dynamical chirality manifests in a polarization-dependent part of the neutron cross section that is completely inelastic and odd in energy transfer [16–18].

In previous papers, we were able to prove the feasibility of this proposal [17] and to study the chiral susceptibility above \( T_N = 8.37 \) K (from specific heat [10]) in CsMnBr\(_3\) [18]. Thereby, the main idea is to measure the difference, \( \Delta I(\omega, \mathbf{Q}) = I^1(\omega, \mathbf{Q}) - I^2(\omega, \mathbf{Q}) \), for incident neutron polarization parallel and antiparallel to the hexagonal axis as a function of energy transfer, \( \omega \), and momentum transfer, \( \mathbf{Q} \). (More details can be found in Refs. [17,18].) For \( \omega \ll T \) the polarization-dependent part of the cross section can be expressed as [18]

\[
\Delta I(\omega) = A(\omega) \left( \frac{\tau}{\Gamma_2} \right) \frac{1}{1 + (\omega/\Gamma_2)^2},
\]

where the scale factor \( A(\tau) \approx \tau^{-\phi} \) depends on the reduced temperature \( \tau = \left( T - T_N \right)/T_N \) and \( \Gamma_2 \) is the half-width of the squared Lorentzian. A least-squares fit of \( \Delta I(\omega) \) by use of Eq. (1) is shown in the inset of Fig. 2. From the temperature dependence of \( A(\tau) \) we were able to obtain the chiral crossover exponent \( \phi_c = \beta_c + \gamma_c = 1.29(7) \) for \( \tau > 0.1 \) [18].

For \( \tau < 0.1 \), a crossover to \( \phi_c = 0.3 \) was observed which was attributed to the limited instrumental energy resolution, since the width of a Lorentzian for the quasielastic polarization-independent scattering became equal to the resolution-function width in the vicinity of \( \tau = 0.1 \). (See Fig. 2 in Ref. [18].) Another possible explanation would be a saturation of \( A \) due to a large magnetic field \( B \). \( \Delta I \) is proportional to \( B \) only in the low-field approximation [16]. A rather crude estimate of the field limit is based on the fact that \( \Delta I \) must be smaller than the total differential cross section \( \Sigma I = I^1 + I^2 \). We found that the polarization-dependent part \( \Delta I \) is only of the order of a few percent even at \( \tau < 0.1 \). The proportionality up to \( B = 4 \) T was checked [18] at \( \tau = 0.11 \) using a beryllium-filter method described in [17]. However, there remained an uncertainty whether the low-field approximation was valid towards lower \( \tau \).

Another possible pitfall is the influence of an in-plane component of \( B \) which splits the chiral phase transition into two separate conventional ones [19,20]. In all our experiments the hexagonal axes were aligned to within \( \pm 2^\circ \) parallel to \( B \). As proven by high-resolution specific-heat experiments [20], the resulting in-plane field component of \( \approx 0.14 \) T at \( B = 4 \) T is by far too small to lead to any detectable splitting of the chiral phase transition.

In order to investigate the crossover of \( A(\tau) \) in more detail, we performed an improved experiment utilizing the higher energy resolution of \( \Delta \omega = 0.08 \) meV instead of \( \Delta \omega = 0.16 \) meV. Simultaneously, the external field was decreased from 4 to 2 T. The same crystal was measured on the IN12 triple-axis spectrometer installed at a cold-neutron guide at the ILL, Grenoble. As before, neutrons with \( k_i = 1.5 \) Å\(^{-1} \) were monochromated by a pyrolitic graphite (PG) crystal and subsequently polarized by a supermirror bender. The sample was mounted in a superconducting cryomagnet with the horizontal field along the hexagonal \( z \) axis. The measurements were carried out at the (1/3 1/3 1) reciprocal lattice point. The results are shown in Fig. 2 (closed circles) in comparison to the previous ones (open circles) [18]. The two data sets are very similar. A change of slope from 1.27(12) to 0.20(4) is visible for the new data at \( \tau = 0.1 \). This means that the crossover is neither a result of the energy resolution nor due to a violation of the low-field approximation. We, therefore, can conclude that the crossover in \( \phi_c \) is probably due to the limited resolution in momentum space. (The experimental spectra were corrected only for the energy resolution.) A correction of the data by taking into account the momentum-resolution ellipsoid as well as the \( T \) dependence of the correlation lengths along the \( z \) axis and in the \( xy \) plane [11] is possible only with large error bars. Therefore, the data for \( \tau > 0.1 \) have much more significance, since they neither depend on the energy resolution nor on the field strength. This finally leads to the

![FIG. 2. The scale factor \( A \) of the polarization-dependent part, \( \Delta I \), of the neutron scattering cross section as a function of reduced temperature, \( \tau = (T - T_N)/T_N \). The inset shows an example for the energy dependence of \( \Delta I \) with a least-squares fit by use of Eq. (1).](image-url)
conclusion that the ensemble of our data from both experiments is consistent with a mean chiral crossover exponent \( \phi_c = 1.28(7) \).

We now turn to the discussion of the polarization-dependent scattering in the ordered phase. Using the results of [21,22], the polarization-dependent intensity of a magnetic reflection can be expressed through the average chirality \( \langle C \rangle \) as

\[
I(Q) \propto f^2(Q)\{(S)^2[1 + (\hat{Q}\hat{C})^2] + 2\langle C \rangle (\hat{Q}\hat{C})(Q\hat{P}_0)\times(n_R - n_L)\delta(Q - k, b)\},
\]

where the scattering vector \( \hat{Q} \) is a unit vector along \( Q \), \( k \) is the propagation vector of the magnetic structure along \([110] \), \( b \) is a reciprocal-lattice vector, \( \hat{C} \) is the unit vector along \( C \) (either \([001] \) or \([00\bar{1}] \) ), \( P_0 \) is the initial neutron polarization, \( n_R \), and \( n_L \) are the populations of the right- and left-handed domains, respectively. The \( \delta \) function reflects the Bragg condition, \( Q = b \pm k \), for a magnetic reflection with propagation vector \( k \). In order to determine \( \beta_c \), one has to measure the \( T \) dependence of the second, polarization-dependent, term in (2), i.e., the difference \( \Delta I = I^1 - I^3 \) with \( P_0 \) parallel and antiparallel to \( Q \).

For \( \Delta I \neq 0 \), it is necessary to prepare a crystal with unequal domain population \( (n_R \neq n_L) \) in order to measure the average chirality. A few methods exist to influence the domain population. For ZnCr\(_2\)Se\(_4\) with a simple spiral spin structure, 95\% of single-handed domains were prepared by magneto-optical cooling [23]. Plumer et al. [24] have noticed that an electric field applied within the basal plane of a TLA breaks the chiral degeneracy, which can be used to prepare a single domain crystal by cooling in an electric field. This technique has been exploited for CsMnBr\(_3\) [25]. Thereby, the application of an electric field (1.5 kV/cm) changed the domain population only slightly, but \( \beta \) decreased by a factor of 1.5. Hence, there is no guarantee that \( \beta_c \) will not be influenced by an electric field. Fortunately, in some antiferromagnetic materials, including the chiral ones, a natural difference in domain population exists, as evidenced by polarized-neutron scattering and magneto-optical data [26].

Indeed, for the CsMnBr\(_3\) crystals used, we have observed relative intensity differences \( \Delta I/\Sigma I = (I^1 - I^3)/(I^1 + I^3) \) of up to about 10\% (data not shown). To avoid this kind of problem, we therefore measured much smaller crystals. Here, we report on the results of two crystals with average dimensions of \( 10 \times 5 \times 3 \) mm\(^3\) (crystal I) and \( 10 \times 2 \times 2 \) mm\(^3\) (crystal II) obtained on the IN20 spectrometer with \( k_i = 1.5 \) Å\(^{-1}\) and on the IN20 spectrometer with \( k_i = 2.662 \) Å\(^{-1}\). In both cases the samples were mounted in orange cryostats installed between Helmholtz coils, which produced the necessary guide field of about 10 Oe either parallel or antiparallel to the Bragg reflection vector \( Q = (1/3 1/3 1) \). These measurements of different-sized crystals with neutrons of different energy are important to exclude any extinction influence on the intensity. Another important source of error is the quasielastic critical scattering which is most intense at \( T_N \). To minimize its contribution on IN20, the usual PG analyzer was replaced by an elastically bent “perfect” silicon crystal with a strongly reduced bending radius (as compared to the usual optimum for real space focusing), resulting in an effective mosaic width of a few arc minutes. This allowed us to separate the Bragg scattering from most of the quasielastic critical scattering which enabled us to measure correctly \( \Delta I \) close to \( T_N \) and to determine a reliable value for \( T_N = 8.225(5) \) K [27].

The chiral domains appear at the chirality-ordering temperature, and their population remains stable as long as the system stays in the ordered phase. We, therefore, rapidly cooled the samples from \( \sim 20 \) K through \( T_N \) down to 5 K followed by a raise of \( T \) in small steps and measurement of \( \Delta I(T) \). For some crystals the magnitude and even the sign of the domain population were random in subsequent thermal cycles, whereas others showed reproducible \( \Delta I(T) \) values. Nevertheless, the \( T \) dependence of \( \Delta I \) was consistent within error bars in all cases.

Figure 3 shows the \( T \) dependence of the polarization-dependent part of the scattering intensity, \( \Delta I \), for two crystals measured on IN20. \( \Delta I \) has been obtained from the peak-height difference. The difference of the integrated intensities gave essentially the same result. The \( T \) dependence of \( \Delta I \), which is proportional to the average chirality \( \langle C \rangle \), follows perfectly well the power law \( \Delta I \propto \tau^{\beta_c} \) over more than 2 orders of magnitude in \( \tau \). We consistently
obtain the chiral critical exponent $\beta_c = 0.42(3)$ for crystal I and $\beta_c = 0.44(2)$ for crystal II with a chiral-ordering temperature of 8.217(3) K [27]. ($\Delta I$ vanishes completely above $T_N$.) The value $\beta_c = 0.45(3)$ obtained for crystal II on the IN12 spectrometer is within error bars compatible with the above-mentioned $\beta_c$’s obtained on IN20 (data not shown). Taking into account the factor of about 4 for the beam path lengths in the two crystals and recalling that the kinematic reflecting power scales with the cube of the wavelength factor of 1.8 between the two experiments, we can conclude that the effect of extinction is negligible. The average value of the chiral critical exponent is $\beta_c = 0.44(2)$. Using our value of the chiral crossover exponent $\phi_c = 1.28(7)$ given above, one obtains $\gamma_c = \phi_c - \beta_c = 0.84(7)$.

As a main result of this study, we have shown that the spin chirality follows a critical behavior. This fact itself rules out the possibility of a standard O(4) critical behavior as predicted in [28] as well as the mean-field tricritical behavior proposed in [4]. Although we cannot give a definitive statement, our result, $\beta_c = 0.44(2)$, agrees better with the Monte Carlo result of Kawamura [13], $\beta_c = 0.45(2)$, rather than with $\beta_c = 0.38(1)$ obtained by Plumer and Mailhot [3]. The chiral-susceptibility exponent, $\gamma_c = 0.84(7)$, is within the limits of the standard deviations of both results, $\gamma_c = 0.77(5)$ [13] and $\gamma_c = 0.90(9)$ [3]. Our $\beta_c$ and $\gamma_c$ values together with the specific-heat exponent $\alpha = 0.40(5)$ [10] satisfy within error bars the scaling relation $\alpha + 2\beta_c + \gamma_c = 2$ expected from renormalization-group arguments [29]. In our case, this sum is equal to 2.12(9).

As mentioned above, chiral XY systems have been predicted to show a weak first-order transition [6], which should become apparent very close to $T_N$ and would violate the scaling relations. From the data presented here we observe no indication for this scenario. Specific-heat data can be described by a second-order transition down to $\tau = 10^{-4}$ [10]. For a final statement data much closer to $T_N$ might be necessary.

Our ability to disentangle for the first time the spin and chiral variables allowed us to pursue the question of whether they are decoupled or not. The important result of this experiment is the agreement between the chiral-order temperature and $T_N$ within a relative precision of about $5 \times 10^{-4}$. This gives strong support for the notion of a single, coupled order parameter at a chiral phase transition. Our technique is highly sensitive to the exact value of $T_N$, since the polarization-dependent part $\Delta I$ disappears rather sharply at $T_N$. This is because quasielastic scattering (1) is canceled when being integrated over the energy resolution. The independent experimental determination of $T_N$, in turn, reduces the uncertainty in the critical exponents, where otherwise $T_N$ would be an implicit fit parameter. Correspondingly, we find for the conventional exponent $\beta = 0.21(1)$ which is at the lower end of the previously reported values [11,12].

In conclusion, we have determined for the first time experimental values of the critical exponents of the spin chirality and the chiral susceptibility for the TLA CsMnBr$_3$. Together with the fact that spin and chiral ordering occur at the same temperature and that the scaling relation is fulfilled, this gives evidence for the validity of the chiral-universality scenario of the magnetic phase transition in the XY TLA. The determination of $\gamma_c$ as well as $\nu_c$ at lower $\tau$ is desirable.

We thank R. K. Kremer for supplying some of the crystals and S. V. Maleyev for helpful discussions. We acknowledge partial support by the Russian Foundations for Fundamental Researches (No. 99-02-17273) and for Neutron Investigations on Condensed Matter (No. 00-15-96814), as well as by EPSRC and NWO.

[27] The difference of $T_N$ compared to the more precise value of [10] originates from thermal gradients in the cryostat.