CsMn(\(\text{Br}_x\text{I}_{1-x}\))\(_3\): Crossover from an \(XY\) to an Ising Chiral Antiferromagnet

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We report on high-resolution specific-heat and magnetocaloric-effect measurements of the triangular-lattice antiferromagnets CsMn(\(\text{Br}_x\text{I}_{1-x}\))\(_3\) with different \(x\). The evolution of the magnetic phase diagrams from the easy-axis system for \(x = 0\) to the easy-plane system for \(x = 1\) was studied in detail. The specific-heat critical exponent \(\alpha\) of the almost isotropic \(x = 0.19\) system agrees with the value predicted for a chiral Heisenberg scenario. In an applied magnetic field \((B = 6 \text{ T})\) a crossover to a weak first-order transition is detected.

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I. INTRODUCTION

The triangular-lattice antiferromagnets ABX\(_3\) with CsNiCl\(_3\) structure, where the magnetic B\(^{2+}\) ions form a triangular lattice, exhibit frustration due to the antiferromagnetic interactions on a triangular plaquette if the magnetic moments have a component in the triangular \(ab\) plane. The magnetic moments then form a 120° structure, with the extra two-fold degeneracy of chirality being broken at the antiferromagnetic transition. Simply speaking, the extra degeneracy arises from the possibility that the 120° spin structure on a given plaquette can be arranged clockwise or counterclockwise when moving around the plaquette. It has been suggested that the chiral degeneracy leads to new universality classes for three-dimensional \(XY\) and Heisenberg models \([3,4]\). The largest changes of the critical exponents are predicted to occur in the specific-heat exponent \(\alpha\), where the chiral \(XY\) and chiral Heisenberg universality classes are predicted to show \(\alpha = 0.34\) and 0.24, compared to \(\alpha = -0.01\) and -0.12 for the standard \(XY\) and Heisenberg models, respectively \([3,4]\). However, whether this concept of new universality classes is indeed applicable is a strongly debated question. Especially within recent years theoretical studies have supplied growing support for a weakly first-order scenario for both chiral phase transitions (see \([3,4]\) and references therein). An experimental indication for this behavior was found recently \([3,4]\).

In any case, the frustration enhances the degeneracy giving rise to different physics with rich phase diagrams and strongly modified critical behavior, which has been studied experimentally for a large number of different triangular-lattice antiferromagnets \([3,4]\). A well-studied example is the easy-plane system CsMnBr\(_3\), for which a number of experiments \([3,4]\) revealed a critical behavior in line with the theoretical prediction \([3,4]\). For ABX\(_3\) systems with easy-axis anisotropy like CsMnI\(_3\) (as well as CsNiCl\(_3\)), chiral behavior can be induced by applying a spin-flop field along the easy \(c\) direction thus forcing the spins into the \(ab\) planes. At the spin-flop field \(B_M\) (~6.4 T for CsMnI\(_3\) and ~2.3 T for CsNiCl\(_3\)) the magnetic energy is equal to the anisotropy energy, i.e., full isotropy in spin space is attained and chiral Heisenberg behavior is found \([3,4]\). For higher fields, an easy-plane anisotropy is induced and \(XY\) chirality occurs \([3,4]\).

Not many triangular-lattice antiferromagnets with negligible anisotropy exist. Besides the above-mentioned materials at their spin-flop fields, only the hexagonal antiferromagnet VBr\(_2\) is known. Indeed, for VBr\(_2\) critical exponents were found in line with the behavior predicted for a chiral Heisenberg system \([7]\). The possibility to tune a chiral Heisenberg system is offered by the solid solution CsMn(\(\text{Br}_x\text{I}_{1-x}\))\(_3\) which spans the range from an easy-axis system \((x = 0)\) to an easy-plane system \((x = 1)\). This system, therefore, allows to study the crossover in the magnetic phase diagrams and its influence on the critical behavior. In particular, the composition with \(x = 0.19\) presents an almost isotropic system and should therefore follow chiral Heisenberg behavior \([3,4]\). Magnetization measurements of CsMn(\(\text{Br}_x\text{I}_{1-x}\))\(_3\) which gave some information on the magnetic phase diagrams have already been reported by Ono et al. \([8]\). Here we report on detailed specific-heat and magnetocaloric-effect measurements.

The spin-Hamiltonian that describes the system is given by

\[
\mathcal{H} = -J_c \sum_{i,j} S_i \cdot S_j - J_{ab} \sum_{i,j} S_i \cdot S_j + D \sum_i (S_i^z)^2 - g\mu_B \sum_i B \cdot S_i. \tag{1}
\]

The summation \((i,j)\) is over nearest neighbors, with the first sum along the \(c\) direction and the second sum in the \(ab\) plane, with the exchange constants \(J_c\) and \(J_{ab}\), respectively. \(D < 0\) corresponds to an easy-axis system, \(D > 0\) to an easy-plane system. For CsMnBr\(_3\), \(J_c = -0.89\ \text{meV}\), \(J_{ab} = -1.7\ \mu\text{eV}\), and \(D = 12\ \mu\text{eV}\).
for CsMnI$_3$, $J_c = -1.5$ meV, $J_{ab} = -7.6$ $\mu$eV, and $D = -3.8$ $\mu$eV. For both materials we are dealing with the $S = 5/2$ spins of Mn$^{2+}$.

The topology of the magnetic phase-diagrams for ABX$_3$ antiferromagnets depends crucially on the sign of $D$ and on the ratio $D/J_{ab}$. Systems with Ising anisotropy ($D < 0$) show two successive phase transitions at $T_{N1}$ and $T_{N2}$ for $B = 0$. In the low-temperature phase ($T < T_{N2}$) the spins order in two sublattices where one third of the spins align along the $c$ axis, whereas the other two thirds are tilted by an angle $\Phi$ (which depends on the ratio $D/J_{ab}$) with respect to the $c$ axis [see Fig. 1(a)]. For CsMnI$_3$ this angle is $\Phi = 51^\circ$ [13]. All spins lie within a plane which includes the $c$ axis. At higher temperatures in the intermediate phase ($T_{N2} < T < T_{N1}$) the tilted spins have an additional degree of freedom, i.e., their components within the basal $ab$ plane is not defined. For $T > T_{N1}$ in the paramagnetic phase only short-range ordered independent spin chains exist along the $c$ axis. For a magnetic field applied within the basal plane ($B \perp c$) the two phase boundaries shift somewhat to higher temperatures (at least up to 6 T for CsMnI$_3$ and CsNiCl$_3$) without changing the principal spin topology [12-24].

Of much more relevance is the case when $B$ is applied along $c$ [Fig. 1(a)]. For $T < T_{N2}$, a first-order phase transition occurs at the spin-flop field $B_c$ above which the three sublattices form a $120^\circ$ umbrella-like structure. The $c$-axis spin component grows with further increasing $B$. For classical spins with $J_c \gg J_{ab}$, $B_c$ at $T = 0$ is given by

$$(g\mu_B B_c)^2 = 16|J_c D|S^2.$$

At the multicritical point $(T_M, B_M)$ the three phase lines merge tangentially into the first-order spin-flop line [21-23]. At this point full isotropy in spin space is achieved which leads to a chiral Heisenberg universality as experimentally observed for CsNiCl$_3$ [17-19] and CsMnI$_3$ [13].

For systems with easy-plane anisotropy ($D > 0$) only one phase transition at $T_N$ from the paramagnetic to the chiral $120^\circ$ structure exists at $B = 0$. The critical behavior, therefore, is of the chiral XY type. A magnetic field applied along the $c$ direction does not change the symmetry of the ground state. Consequently, the critical behavior stays essentially constant [13]. A much richer phase diagram can be observed for $B \perp c$. The phase diagram as predicted for $D < 3|J_{ab}|$ is shown in Fig. 1(b) [23]. At $T < T_N$ and $B < B_c$ the chiral phase exists. The competition between the Zeeman energy and the anisotropy energy leads to a spin-flop phase above $B_c$ which is also given by Eq. (2) [14]. Thereby, the spin triangle is oriented perpendicularly to $B$. Between the chiral low-temperature and the paramagnetic high-temperature phase, a collinear spin structure evolves where the spins remain in the $ab$ plane with two spins of a triangle pointing parallel and one in the opposite direction. With increasing field the spin components along $B$ become larger.

Experimentally, very little is known about the phase diagrams and the critical properties of easy-plane systems with such a small anisotropy ($D < 3|J_{ab}|$) [9,13,24]. Due to the stronger anisotropy $D$ the spin-flop phase is absent for such XY systems with only the chiral phase and the collinear structure remaining [13].

FIG. 1. (a) Schematic phase diagram of a triangular-lattice antiferromagnet with easy-axis anisotropy ($D < 0$) for $B||c$. The phase lines meet at the multicritical point $(T_M, B_M)$. $B_c$ is the critical field for the spin-flop transition. (b) The schematic phase diagram for systems with a small easy-plane anisotropy $0 < D < 3|J_{ab}|$ for $B \perp c$. The insets sketch the spin arrangements.

FIG. 2. Specific heat $C$ divided by temperature $T$ vs $T$ for CsMn(Br$_{0.1}$I$_{0.9}$)$_3$ in a magnetic field $B$ parallel to the $c$ direction. Data are shifted consecutively by 0.2 J/molK$^2$ with respect to $B = 0$. 

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II. EXPERIMENTAL

Single-crystalline samples of CsMn(Br$_{1-x}$I$_x$)$_3$ were grown by the Bridgman technique at the Tokyo Institute of Technology [13]. For the measurements pieces of 24 to 111 mg were cleaved from the crystals. The specific heat, \( C \), was measured by a standard semiadiabatic heat-pulse technique. Magnetic fields up to 14 T were applied either along or perpendicularly to the clearly visible \( c \) axis of the crystals. The magnetocaloric effect, \( \frac{\delta T}{\delta B} = -(T/C)\frac{\delta S}{\delta B} \), was measured in the same calorimeter. \( S \) denotes the entropy of the system. Upon changing the magnetic field by small steps \( \Delta B \), the resulting temperature variation \( \Delta T \) was recorded. Taking into account the small eddy-current heating the magnetocaloric effect \( \Delta T \) was extracted. For more details on the experiment see Ref. [25].

\[ \text{FIG. 3. } C/T \text{ vs } T \text{ for } \text{CsMn(Br}_{1-x}\text{I}_x)_3 \text{ in } B \perp c. \text{ Data are shifted consecutively by } 0.2 \text{ J/molK}^2 \text{ with respect to } B = 0. \text{ The arrows indicate the phase transitions between the spin-flop phase and the intermediate collinear phase. The inset shows an enlargement of the data at } B = 3.5 \text{T.} \]

III. RESULTS AND DISCUSSION

The specific heat of the sample with the smallest Br concentration, CsMn(Br$_{0.1}$I$_{0.9}$)$_3$, is shown in Fig. 3 for different fields \( B \) aligned along the \( c \) direction. This easy-axis system shows two consecutive zero-field transitions, which merge into one at the spin-flop field of about 4 T. The steep anomaly found beyond this field resembles that found for pure CsMnI$_3$ which was analyzed in terms of a chiral Heisenberg model at the spin-flop field \( B_M \approx 6.4 \text{T} \) [13]. The substitution of 10% of the I$^-$ ions by Br$^-$ with the concomitant increase of the (negative) anisotropy \( D \) towards zero reduces considerably the spin-flop field and likewise the width of the intermediate phase (\( T_N2 = 8.36 \text{ K} < T < T_N1 = 9.80 \text{ K} \)). This trend continues further for a sample with \( x = 0.18 \) (data not shown) where \( T_N1 = 8.50 \text{ K}, T_N2 = 8.40 \text{ K}, \) and \( B_M \approx 1 \text{T} \) (see also Figs. 4 and 5 below).

\[ \text{FIG. 4. } C/T \text{ vs } T \text{ for } \text{CsMn(Br}_{0.19}\text{I}_{0.81})_3 \text{ in a magnetic field } B \parallel c. \text{ Data are shifted consecutively by } 0.2 \text{ J/molK}^2 \text{ with respect to } B = 0. \text{ Consequently, the anisotropy } D \text{ should become zero somewhere between } x = 0.18 \text{ and } x = 0.20. \text{ A good candidate for such a chiral Heisenberg system is therefore CsMn(Br}_{0.19}\text{I}_{0.81})_3. \text{ For an isotropic Heisenberg system only one phase-transition line from the paramag-} \]
netic to the chiral phase is expected, independent of the magnetic-field orientation. Indeed, magnetization and susceptibility data could not observe any spin-flop line or splitting of the zero-field transition. Our specific-heat data for \( B \parallel c \) (Fig. 3) are in line with these observations. We particularly can resolve only a single strong anomaly at \( T_N \) which becomes somewhat larger with increasing field up to 6 T, similar to what is observed for \( x = 0.1 \) above \( B_M \).

However, measurements of CsMn\((\text{Br}_{0.19}\text{I}_{0.81})_3\) for \( B \perp c \) (Fig. 5) reflect a small residual planar anisotropy, as evidenced by a slight splitting of the transition in fields between 1 and 3 T. The anomaly at lower temperatures is still relatively sharp and large at \( B = 1 \) T, but becomes clearly reduced at 1.2 T which indicates the junction with the spin-flop phase line. At higher fields the phase lines merge and only one anomaly remains at 6 T.

In order to determine the complete phase diagrams including the expected spin-flop lines (see Fig. 1), we measured the magnetocaloric effect for all samples. Since the spin-flop transition at \( B_c \) is almost temperature independent, the specific heat is not sensitive to this transition, contrary to magnetocaloric-effect measurements which cross the corresponding phase line at an approximately right angle. Figure 6(a) shows the magnetocaloric effect for the samples with \( x = 0.18 \) and \( x = 0.19 \) at \( T \approx 7 \) K in fields aligned parallel to the \( c \) axis. For \( x = 0.18 \), a clear step at about 0.9 T is visible which signals the spin-flop transition in line with the data of Ono et al. [18]. The spin-flop field at each temperature was estimated from the position of the maximum in the derivative of the magnetocaloric-effect data. For \( x = 0.19 \), \( \Delta T/\Delta B \) increases monotonically without any detectable step or anomaly. This confirms that CsMn\((\text{Br}_{0.19}\text{I}_{0.81})_3\) has no Ising-like anisotropy.

Instead the anisotropy \( D \) has switched to an \( XY \) type, as the specific-heat data (Fig. 3) show. Consequently, a spin-flop line is expected for fields perpendicular to \( c \) [see Fig. 1(b)]. Indeed, magnetocaloric-effect data could verify this phase diagram by showing a step-like feature at about 1.2 T almost independent of temperature [Fig. 6(b)]. Therefore, the critical concentration for which \( D = 0 \) should be just below \( x = 0.19 \).

Figure 6 summarizes the results in terms of \((B, T)\) phase diagrams for various \( x \). In Fig. 6(a), the absolute magnitude of the easy-axis anisotropy \((D < 0)\) decreases with increasing \( x \), getting close to zero for \( x = 0.18 \). From the reduced spin-flop field \( B_c \approx 1 \) T for \( x = 0.18 \) one can estimate with Eq. (3) that \(|J_cD|\) has reduced to about 2.5\% of the value for CsMn\(3\). Since \( J_c \) should depend little on \( x \), this means that \(|D|\) has reduced to about 95 neV corresponding to 1.1 mK. The phase diagrams for \( x = 0.10 \) and \( x = 0.18 \) fully agree with the predicted behavior for easy-axis systems [Fig. 6(a)].

The phase-diagram topology changes for easy-plane systems with \( D > 0 \). In Fig. 6(b), the absolute magnitude of \( D \) increases with \( x \), with a small anisotropy present for \( x = 0.19 \). As for \( B \parallel c \), the phase diagrams for \( B \perp c \) \((D > 0)\) are in full agreement with mean-field calculations and verify nicely the predictions for easy-plane systems with \( D < 3|J_{ab}| \) [23].

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**FIG. 5.** \( C/T \) vs \( T \) for CsMn\((\text{Br}_{0.19}\text{I}_{0.81})_3\) in a magnetic field \( B \) perpendicular to the \( c \) direction. Data are shifted by different amounts with respect to \( B = 0 \). The arrows indicate the small anomaly indicating the transition from the intermediate phase to the paramagnetic phase.

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**FIG. 6.** Magnetocaloric effect of (a) CsMn\((\text{Br}_{0.19}\text{I}_{0.81})_3\) and CsMn\((\text{Br}_{0.18}\text{I}_{0.82})_3\) in \( B \parallel c \) and (b) of CsMn\((\text{Br}_{0.19}\text{I}_{0.81})_3\) for three different temperatures in \( B \perp c \). Data in (b) are shifted consecutively by 5 mK/T with respect to \( T = 8.1 \) K.
FIG. 7. Phase diagrams of CsMn(Br$_x$I$_{1-x}$)$_3$ (a) for fields along the $c$ direction for $x \leq 0.19$ and (b) for fields perpendicular to $c$ for $x \geq 19$.

The phase diagram of the transition temperatures vs Br concentration $x$ is shown in Fig. 8. The lower Néel temperature $T_{N2}$ increases slightly with $x$, whereas $T_{N1}$ rapidly decreases. The two phase lines merge at a critical concentration, $x_c$, somewhere between $x = 0.18$ and $x = 0.19$. The phase diagram is in line with that reported by Ono et al. [18]. With our specific-heat and magnetocaloric-effect measurements, however, we were able to resolve the spin-flop line and the intermediate collinear phase for $x = 0.19$ at fields between 1 and 3 T proving that the critical concentration with $D = 0$ must be slightly less than $x = 0.19$.

For completeness, the inset of Fig. 8 shows a comparative $B-T$ phase diagram for different chiral XY systems with $B$ aligned along the $c$ direction. The data for CsMnBr$_3$ are from [20]. With increasing field the phase transition from the paramagnetic to the chiral

phase shifts to higher temperatures. This effect is less prominent for larger $x$ indicating that the field-induced $T_N$ increase becomes larger for reduced $D$.

As a final point, we discuss the critical behavior of CsMn(Br$_x$I$_{1-x}$)$_3$. In order to describe the specific-heat data close to the critical temperature $T_c$, we applied the usual fit function [27]

$$C^\pm = (A^\pm/\alpha)|t|^{-\alpha} + B + Et,$$

where $t = (T - T_c)/T_c$ and the superscript $+$ ($-$) refers to $t > 0$ ($t < 0$). The first term describes the leading contribution to the singularity in $C$ and the non-singular contribution to the specific heat is approximated by $B + Et$. After a good fit of the data had been achieved except very close to $T_c$, a Gaussian distribution of $T_c$ with width $\delta T_c$ was introduced. This procedure is able to describe a rounding of the transitions caused by sample inhomogeneities (see also Refs. [13,14]).

Figure 9(a) shows the specific heat $C$ for $x = 0.19$ vs the reduced temperature $|t|$ at $B = 0$. The data are compatible with an exponent $\alpha = 0.23(7)$ of the specific heat if we include a Gaussian broadening of $\delta T_c/T_c \approx 4.2 \times 10^{-4}$. The dashed lines indicate the fit with these parameters, while the solid lines represent a fit with $\delta T_c = 0$. The exponent $\alpha$ as well as the experimental amplitude ratio $A^+//A^- = 0.54(13)$ are in line with the chiral Heisenberg model, which predicts $\alpha = 0.24(8)$ and $A^+/A^- = 0.54(20)$ [2]. This is in accordance with the small easy-plane anisotropy which obviously is too small to force the system to XY chirality. The available
results of neutron-scattering experiments for \( x = 0.19 \) are rather inconclusive [2]. While the exponent \( \beta = 0.28(2) \) of the sublattice magnetization agrees well with the theoretical value of a chiral Heisenberg system \( (\beta = 0.30) \), the exponents of the susceptibility, \( \gamma = 0.75(4) \), and of the correlation length, \( \nu = 0.42(3) \), are at variance with the predictions \( (\gamma = 1.17 \text{ and } \nu = 0.59) \). Furthermore, the experimentally found exponents are in contradiction to the fundamental scaling laws \( \alpha + 2 \beta + \gamma = 2 \) and \( \alpha + d \nu = 2 \) \((d \text{ is the dimension})\), which should be fulfilled at universal second-order phase transitions.

Theoretically, the region around the multicritical point where a chiral Heisenberg scenario is expected should not be large [2]. This would imply that either the increase of the Br concentration \( (x > x_c) \) or the application of a large magnetic field \( (B > B_M) \) should drive the system quickly to chiral \( XY \) behavior with \( \alpha = 0.34(6) \), which for \( \text{CsMnBr}_3 \) \((x = 1)\) has been observed [10][11]. Nevertheless, for \( x = 0.20 \) and for \( x = 0.25 \) the critical exponents remain approximately constant with \( \alpha = 0.25(7) \) and \( \alpha = 0.20(6) \), respectively, suggesting that the chiral Heisenberg behavior is rather stable.

Another unexpected result becomes obvious from the analysis of the specific-heat data of \( x = 0.19 \) in \( B = 6 \, \text{T} \) applied along the \( c \) direction [Fig. 3(b)]. A magnetic field of this strength, i.e., much larger than \( B_M \), should induce \( XY \) chirality for a system with Ising anisotropy as previously observed for \( \text{CsNiCl}_3 \) \([13][14] \) and, in this work, with \( \alpha = 0.37(10) \) for \( \text{CsMn(Br)}_{1.10.9} \) at \( B = 6 \, \text{T} \) (not shown). Likewise, for an easy-plane system like \( \text{CsMnBr}_3 \) the critical behavior remains chiral \( XY \)-like for fields applied along \( c \) [7]. However, for \( x = 0.19 \) (as well as for \( x = 0.18 \), data not shown) in a magnetic field of \( 6 \, \text{T} \), \( C \) for \( t \geq 10^{-3} \) still follows a chiral Heisenberg-like behavior with \( \alpha = 0.21(8) \) \([\alpha = 0.23(6) \text{ for } x = 0.18]\) and increases much more strongly for \( t \to 0 \) which can neither be described by a reasonable critical exponent nor by a \( T_c \) distribution. This possibly indicates a crossover to a weakly first-order transition, similar as previously observed for \( \text{CsCuCl}_3 \) close to \( T_c \) at \( B = 0 \) [8].

In conclusion, we have mapped out in detail the impact of an axial versus a planar anisotropy on the magnetic phase diagrams of triangular-lattice antiferromagnets by fine-tuning \( x \) of the system \( \text{CsMn(Br)}_{1-x} \). In particular, the predicted phase diagram [Fig. 3(b)] of easy-plane systems with small anisotropy could be accurately verified. The critical concentration for which the spin anisotropy vanishes was found to be located between \( x = 0.18 \), a system with small axial anisotropy \( (D < 0) \), and \( x = 0.19 \), a system with small planar anisotropy \( (D > 0) \). The critical behavior at \( B = 0 \) for \( x = 0.19, x = 0.20, \) and \( x = 0.25 \) can be described with critical exponents \( \alpha \) as predicted from Monte-Carlo simulations for the chiral Heisenberg universality class [2]. Thereby, rounding effects due to sample inhomogeneities prevent the possible detection of a crossover to a first-order scenario as proposed recently [9]. In a magnetic field \( B = 6 \, \text{T} \), the samples with \( x = 0.18 \) and \( x = 0.19 \) show a weakly first-order phase transition. For \( 10^{-3} < t < 0.1 \), the data can be described by chiral Heisenberg critical exponents. For all other samples with either larger planar or larger axial symmetry, no indication for a first-order phase transition was detected.

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