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LETTER TO THE EDITOR

Chiral universality in $CsMnI_3$ and $CsNiCl_3$

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Abstract. $CsMnI_3$ and $CsNiCl_3$ are hexagonal Heisenberg antiferromagnets with small Ising-like anisotropy. The magnetic specific heat of $CsNiCl_3$, measured by linear magnetic birefringence (LMB), follows chiral XY behaviour at the phase transition between the paramagnetic and spin-flop phase up to 14.5 T, and Heisenberg behaviour close to the multicritical point. The magnetization discontinuity of $CsMnI_3$ at the spin-flop transition was investigated by Faraday rotation. Its temperature dependence is consistent with the prediction for the chiral Heisenberg universality class.

Recently there has been growing interest in the phase transitions of hexagonal antiferromagnets because new chiral universality classes have been proposed for antiferromagnets with continuous symmetries (XY, Heisenberg) on a triangular or hexagonal lattice. The topology of the H–T phase diagram and the universality of the phase transitions of weakly anisotropic Heisenberg antiferromagnets depend on whether the lattice is triangular or rectangular [1, 2]. Table 1 shows the critical exponents on rectangular and triangular lattices [1].

Table 1. Critical exponents of antiferromagnets on rectangular and triangular lattices.

Model	α	β	γ	ν	A^+/A^-
Rectangular Ising	0.1098(29)	0.325(1)	1.2402(9)	0.6300(8)	0.55
Rectangular XY	−0.0080(32)	0.346(1)	1.3160(12)	0.6693(10)	0.99
Rectangular Heisenberg	−0.1160(36)	0.3647(12)	1.3866(12)	0.7054(11)	1.36
Chiral XY	0.34(6)	0.253(10)	1.13(5)	0.54(2)	0.36(2)
Chiral Heisenberg	0.24(8)	0.30(2)	1.17(7)	0.59(2)	0.54(2)

Evidence of chiral XY critical behaviour was observed in $CsMnBr_3$ [3] (XY anisotropy). Heisenberg antiferromagnets with Ising anisotropy ($\| c$) display, for $H \| c$, a field dependent phase transition from the low-temperature low-field phase (spins in the ac-plane) to the spin-flop phase, where the spins become perpendicular to B (spin-flop transition). On a triangular lattice, they form a 120° spin structure in the ab-plane. This spin structure has chiral XY symmetry. The temperature dependent phase transition between spin-flop and paramagnetic phase belongs to the chiral XY universality class [2]. A crossover to chiral Heisenberg universal behaviour is expected towards the multicritical point, the merging

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point of the boundaries of paramagnetic, spin-flop and the two antiferromagnetic low-field phases (cf. figure 1). On a rectangular lattice, the transition from spin-flop to paramagnetic phase has the usual XY universality, with a crossover to Heisenberg behaviour close to the critical point. Chiral XY behaviour at the transition between paramagnetic and spin-flop phase has been observed in specific heat experiments on $CsNiCl_3$ in magnetic fields up to 6 T [4]. However, the data seem to indicate an increase of the specific heat exponent with increasing field strength. We extended the investigation of this phase transition to magnetic fields up to 14.5 T. Chiral/usual Heisenberg universality is also reflected by the temperature dependence of the magnetization discontinuity at the spin-flop transition towards the multicritical point [2]. Here, the critical behaviour at this phase transition is studied for the first time. Obviously the specific heat exponent α varies most for the different universality classes. We investigated α on $CsNiCl_3$ and $CsMnI_3$, which are Heisenberg antiferromagnets with small Ising anisotropy, in magnetic fields parallel to the Ising axis. The magnetization discontinuity of $CsMnI_3$ at the spin-flop transition was studied by Faraday rotation, the magnetic specific heat of $CsNiCl_3$ at the paramagnetic to spin-flop transition by LMB.

The optical experiments were performed in a superconducting solenoid, which produced magnetic fields up to 14.5 T. LMB experiments were performed with the Sénarmont method as described in [5]. The He–Ne laser beam was deflected into and out of the cryostat by two pairs of mirrors. The Faraday rotation was measured in an inversed geometry (circularly polarized source–analyser–photoelastic modulator–sample–fixed polarizer). A Babinet–Soleil compensator was used to compensate the polarization dependent phase shift produced by the mirrors. For the Faraday experiments, $CsMnI_3$ was cut and polished perpendicular to the c axis. The orientation of the sample was checked in a conoscopic beam.

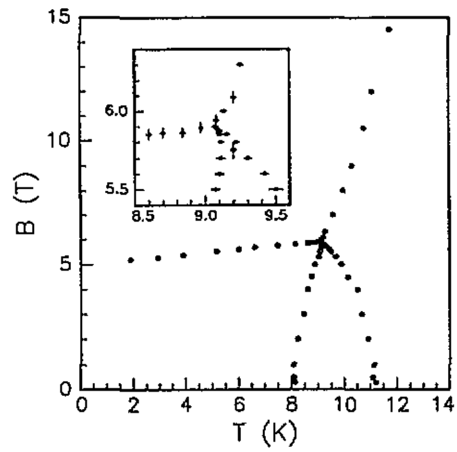

Figure 1. Phase diagram of $CsMnI_3$ determined by Faraday rotation. Inset: region of the multicritical point.

The phase diagrams of $CsNiCl_3$ and $CsMnI_3$ look very similar. Figure 1 shows the phase diagram of $CsMnI_3$ obtained by Faraday rotation (θ). The multicritical point is found at $T_m = 9.02(5)$ K, $B_m = 5.86(1)$ T. Although our data are much more detailed in the multicritical region than earlier published results [6], it is still not clear whether all three phase boundaries become exactly tangential to the spin-flop transition line at the multicritical point, as predicted in [1]. In any case, our data are consistent with a crossover exponent

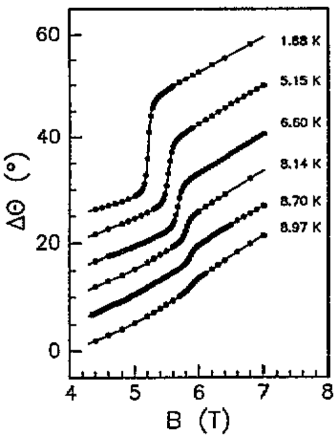

Figure 2. Faraday rotation of $CsMnI_3$ at the spin-flop transition. The discontinuity at the spin-flop transition is proportional to the magnetization discontinuity.

ϕ close to 1. Figure 2 displays typical field scans across the spin-flop transition. The small width of the spin-flop transition (FWHM of $\mathrm{d}\theta/\mathrm{d}H$: 0.06 T at $T = 1.88$ K) at low temperatures indicates the quality of the alignment $H \| c$, which is crucial for the investigation of chiral critical properties. About the same width was obtained in LMB experiments on $CsNiCl_3$ (FWHM of $\mathrm{d}\Delta n/\mathrm{d}H$: 0.07 T at 2.6 K). This should be compared to a width of about 0.5 T in the previous experiments [6]. The Faraday rotation discontinuity $\Delta\theta$ is shown in figure 3. $\Delta\theta$ is the difference of the linear extrapolations of the measured rotation below and above the transition, taken at the turning point of $\theta(H)$.

The Faraday rotation θ is proportional to the components of magnetization and magnetic field along the propagation vector of the light, $\theta = AM^z + CB^z_{\mathrm{loc}}$, where the first term is due to the different thermal occupation of ground state sublevels and the second arises from the Zeeman splitting of the levels. A and C can be of the same order of magnitude for antiferromagnets with zero angular momentum [7]. The steep change of slope at the spin-flop transition shows that C must be rather small for $CsMnI_3$ (figure 2). In any case the discontinuity of θ is proportional to the magnetization discontinuity. It is expected to vary with temperature according to $t^{\tilde{\beta}}(1 + ct^{\phi-1})$, where $t = |T - T_{\mathrm{m}}|/T_{\mathrm{m}}$, $\phi \approx 1.06$ and $\tilde{\beta} = 2 - \alpha - \phi$ [2]. For ϕ close to 1, the discontinuity can be fitted with $\Delta\theta = at^{\tilde{\beta}}$. We obtain $\tilde{\beta} = 0.50 \pm 0.03$, corresponding to $\alpha = 0.44$. T_{m} was fixed to 9.02 K. The value of α points to chiral universality (see table 1), but is larger than expected for the Heisenberg universality class. Even larger values of α are found if $\tilde{\beta}$ is determined from a region closer to T_{m}. This points to a value of ϕ slightly larger than 1.06. Even then, $t^{\tilde{\beta}}$ remains the dominant term in the discontinuity. Note that the rectangular Heisenberg model would require ϕ as large as 1.6. Hence the temperature variation of the magnetization discontinuity is consistent with chiral critical behaviour.

On $CsNiCl_3$, we studied the transition between spin-flop and paramagnetic phase by LMB. Figure 4 shows a typical example of the birefringence derivative $\mathrm{d}\Delta n/\mathrm{d}T$. The anomaly was fitted to an inverse power law in the region $|T - T_{\mathrm{N}}|/T_{\mathrm{N}} \leqslant 0.35$. As discussed below, the critical exponent of this power law is the specific heat exponent α for $T \geqslant T_{\mathrm{N}}$. The non-critical part — contributions from 1D correlations and the lattice — was approximated by a straight line, $bT + c$. $\mathrm{d}\Delta n/\mathrm{d}T$ is linear between 8 K and 14 K, above the phase transition, but should vanish smoothly as T approaches 0. Hence not only T_{N} and the

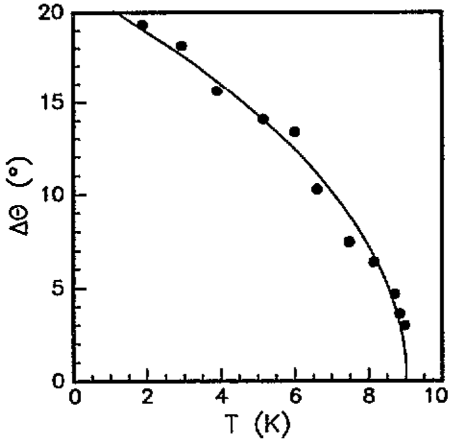

Figure 3. Magnetization discontinuity at the spin-flop transition, measured by Faraday rotation on $CsMnI_3$, in dependence of temperature. Solid line: fit $\Delta\theta = a|t|^{\tilde{\beta}}$ with $\tilde{\beta} = 2 - \phi - \alpha = 0.5$. This value of $\tilde{\beta}$ indicates an unusually large specific heat exponent α.

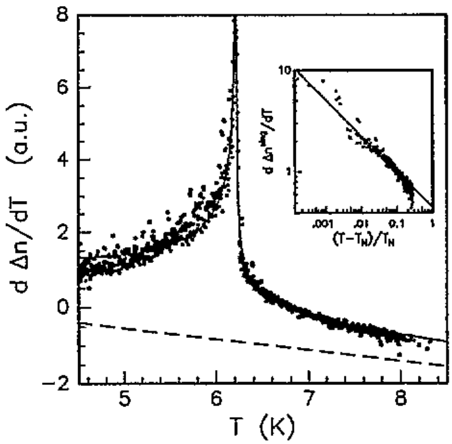

Figure 4. Temperature derivative of the LMB of $CsNiCl_3$, ($\mathrm{d}\Delta n/\mathrm{d}T$), at $B = 11$ T ($\parallel c$) at the transition between spin-flop and paramagnetic phase. Solid line: fit with $\alpha = 0.35$. Inset: singular part of $\mathrm{d}\Delta n/\mathrm{d}T$ in the paramagnetic phase.

amplitudes of the power law, but also b, c were varied in the fit. The latter were restricted to $B \leqslant b \leqslant 0$ and $0 \leqslant c \leqslant C$, where B and C are determined from a linear fit of $\mathrm{d}\Delta n/\mathrm{d}T$ between 8 and 14 K. α was varied in steps of 0.01 until b or c left the reasonable interval. This defines the error bars of α. T_N was usually stable to 1–2 $\times 10^{-3}$ K. To obtain good agreement *above* the phase transition, the temperature region below T_N was reduced and α was checked with fixed T_N in the region $T \geqslant T_N$. Figure 5 shows the obtained values of α. The large error bars result from the uncertainty in the determination of b, c.

In uniaxial antiferromagnets with quenched orbital momentum and nearly Heisenberg exchange interaction of the magnetic ions, the temperature dependence of the LMB at $H = 0$ phase transitions is proportional to the isotropic exchange interaction and hence $\mathrm{d}\Delta n/\mathrm{d}T$ varies as the magnetic specific heat [8]. At finite magnetic field, in the spin-flop phase, anisotropic contributions like $\mathrm{d}\langle (S^z)^2(T)\rangle/\mathrm{d}T$ with deviating critical behaviour become

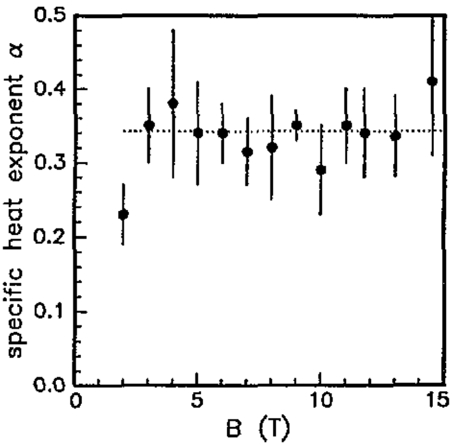

Figure 5. Specific heat exponent α determined from LMB experiments on $CsNiCl_3$ as described in the text. Broken line: mean value at the transition between spin-flop and paramagnetic phase (3 T $\leqslant B \leqslant$ 14.5 T), in excellent agreement with the prediction for the chiral XY universality class (0.34 ± 0.06). The value at $B = 2$ T, close to the multicritical point ($B_m = 2.1$ T), follows the prediction for the Heisenberg universality class (0.24 ± 0.08).

finite, because of the temperature dependence of the order parameter. In the paramagnetic phase, the order parameter is zero and the anisotropic terms are not expected to vary with temperature. The remaining field induced and temperature dependent contributions to the LMB in the paramagnetic phase are proportional to MH and M^2. Since

$$\frac{\partial M(H,T)}{\partial T} + \frac{\mathrm{d}T_c}{\mathrm{d}H}\frac{C_m}{T} = A(H,T) \tag{0.1}$$

at second-order phase transitions, where $A(H,T)$ is a continuous function at the phase transition [9], $(\partial M/\partial T)$ and $(\partial M^2/\partial T) = 2M(\partial M/\partial T)$ diverge as C_m at the phase transition from above and $\mathrm{d}\Delta n/\mathrm{d}T \sim C_m$ above T_N. Hence the specific heat exponent can be derived from $\mathrm{d}\Delta n/\mathrm{d}T$ above the phase transition.

For 3 T $\leqslant B \leqslant$ 14.5 T the values of α range between 0.3 and 0.4 with $\bar{\alpha} = 0.342 \pm 0.005$. They are clearly different from the exponents of the rectangular universality classes, but in excellent agreement with the proposed exponent of the chiral XY universality class. There is no increase of the specific heat exponent with the magnetic field within the error bars. At $B = 2$ T, which is close to the multicritical point $B_m = 2.1$ T of $CsNiCl_3$ [10], we obtain $\alpha = 0.23 \pm 0.04$ as predicted for chiral Heisenberg universality. Since the determined specific heat exponents are far away from those of rectangular universality classes, chiral critical behaviour is confirmed by the value of α alone.

To conclude, we studied the magnetic specific heat of $CsNiCl_3$ via LMB and the temperature dependence of the magnetization discontinuity at the spin-flop transition of $CsMnI_3$ via Faraday rotation. Up to the highest magnetic fields, we confirmed chiral XY critical behaviour along the spin-flop to paramagnetic transition of a hexagonal Heisenberg antiferromagnet with small Ising anisotropy, and chiral Heisenberg behaviour close to the multicritical point.

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