Quenched chirality in RbNiCl$_3$

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The critical behaviour of stacked-triangular antiferromagnets has been intensely studied since Kawamura predicted new universality classes for triangular and helical antiferromagnets. The new universality classes are linked to an additional discrete degree of freedom, chirality, which is not present on rectangular lattices, nor in ferromagnets. However, the theoretical as well as experimental situation is discussed controversially, and generic scaling without universality has been proposed as an alternative scenario. Here we present a careful investigation of the zero-field critical behaviour of RbNiCl$_3$, a stacked-triangular Heisenberg antiferromagnet with very small Ising anisotropy. From linear birefringence experiments we determine the specific heat exponent $\alpha$ as well as the critical amplitude ratio $A^+/A^-$. Our high-resolution measurements point to a single second order phase transition with standard Heisenberg critical behaviour, contrary to all theoretical predictions. From a supplementary neutron diffraction study we can exclude a structural phase transition at $T_N$. We discuss our results in the context of other available experimental results on RbNiCl$_3$ and related compounds. We arrive at a simple intuitive explanation which may be relevant for other discrepancies observed in the critical behaviour of stacked-triangular antiferromagnets. In RbNiCl$_3$ the ordering of the chirality is suppressed by strong spin fluctuations, yielding to a different phase diagram, as compared to e.g. CsNiCl$_3$, where the Ising anisotropy prevents these fluctuations.

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On a hexagonal lattice, an antiferromagnet can never entirely satisfy its interactions, they will be at least partially frustrated. A stacked set of triangular planes will nevertheless develop long-range order for any finite interplane interaction. In the perfectly isotropic case (Heisenberg antiferromagnet), neighboring spins on a triangle compromise the antiferromagnetic interaction by including 120°, along the hexagonal axis the spins will be collinear. The magnetic structure is then defined by two continuous degrees of freedom (the polar and azimuthal angle of one chosen spin) and one additional discrete degree of freedom, the chirality, the sense of rotation of the spin direction on a chosen triangle. This chirality vanishes in collinear structures, on rectangular lattices and in ferromagnets. It is still present for easy-plane antiferromagnets, and in the spin-flop phases of antiferromagnets with a small Ising-anisotropy. A large family of hexagonal compounds with a chiral degree of freedom can be described by the Hamiltonian

$$H = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{i,k} \mathbf{S}_i \cdot \mathbf{S}_k - D \sum_i (S_i^z)^2.$$  (1)

Here, $J > 0$ denotes the antiferromagnetic exchange interaction between nearest neighbours along the symmetry axis, $J' > 0$ the antiferromagnetic interaction between nearest neighbours on a triangle. The single ion anisotropy constant $D$ favors an easy-axis ($D > 0$) or plane ($D < 0$). Kawamura predicted that the chiral degree of freedom provokes not only a different topology of the field-temperature phase diagrams, but also new types of universal critical behaviour, the $n = 2$ chiral and the $n = 3$ chiral universality classes. This prediction is discussed controversially, and arguments have been given for quite different scenarios, as e.g. generic non-universal behavior.

Table I lists Kawamura’s predictions for the critical exponents $\alpha, \beta, \gamma$ and $\delta$ and the ratio $A^+/A^-$ for antiferromagnets on rectangular and triangular lattices as a survey.

ABX$_3$ compounds with easy-axis anisotropy, as CsNiCl$_3$, RbNiCl$_3$, CsMnI$_3$ and CsNiBr$_3$ are well described by the Hamiltonian in Eq. (1) and have developed into model systems for low dimensional fluctuations and ordering, see e.g. [3] for a recent review. These compounds show quasi one-dimensional (1D) magnetic behavior, because the intrachain interaction $J$ is much larger than the interchain interaction $J'$, typically $J'/J \approx 10^{-2}$. One-dimensional short-range antiferromagnetic order within the 1D spin chains develops below about 40 K. At lower temperatures there is a phase transition into a three-dimensionally (3D) magnetically ordered structure. Without an external field, Heisenberg antiferromagnets with an Ising anisotropy on a triangular lattice undergo two successive phase transitions, where ordering of the spin components parallel and perpendicular to the hexagonal $c$-axis occurs at $T_{N1}$ and $T_{N2}$ ($=T_{N1}$), respectively. Below $T_{N2}$, the spins form a 120° structure in the $ac$ plane. The predicted B-T phase diagram is schematically shown in Fig. 4. The two zero-field
phase transitions should show 3D XY-critical behavior. On a rectangular lattice, there is just one transition with Ising-type critical behavior\textsuperscript{3,4,5}.

In order to clarify the number of phase transitions in RbNiCl\textsubscript{3}, and their criticality, we performed linear magnetic birefringence (LMB) experiments with a high temperature resolution to measure the critical exponent $\alpha$ and the amplitude ratio $A^+/A^-$. The paper is organized as follows: The properties of RbNiCl\textsubscript{3} are discussed in Sect. I. Experimental details of the birefringence set-up are presented in Sect. II, the LMB results and the outcome of a supplementary neutron diffraction study are shown and discussed in Sect. III. The anomalous behavior of RbNiCl\textsubscript{3} as compared to other members of the above mentioned ABX\textsubscript{3} family, is discussed in the discussion in Sect. IV.

I. RbNiCl\textsubscript{3}

RbNiCl\textsubscript{3} is a quasi 1D $S$=1 Heisenberg antiferromagnet with a weak Ising anisotropy on a triangular lattice

( )

| & & & & & |
|---|---|---|---|---|---|
| & Ising & $\alpha$ | $\beta$ | $\gamma$ | $\nu$ | $A^+/A^-$ |
|□ | XY & $-0.0080(32)$ & $0.346(1)$ & $1.3160(12)$ & $0.6693(10)$ | $0.99$ |
| & Heisenberg & $-0.1160(36)$ & $0.3647(12)$ & $1.3866(12)$ & $0.7054(11)$ | $1.36$ |
|△ | n=2 chiral & $0.34(6)$ & $0.253(10)$ & $1.13(5)$ & $0.54(2)$ | $0.36(2)$ |
| | n=3 chiral & $0.24(8)$ & $0.30(2)$ & $1.17(7)$ & $0.59(2)$ | $0.54(2)$ |

TABLE I: Critical exponents for antiferromagnets on square and triangular lattices after Kawamura, see \textsuperscript{3,4,5} and references therein.

\textbf{FIG. 1:} Predicted phase diagram for $\text{ABX}_3$ with easy-axis anisotropy. In zero magnetic field, two successive phase transitions are expected, connected with ordering of the spin components parallel and perpendicular to the hexagonal $c$-axis at $T_{N1}$ and $T_{N2}$ ($<T_{N1}$), respectively. Both transitions should show XY critical behavior.

\textbf{FIG. 2:} In RbNiCl\textsubscript{3}, magnetic exchange $J$ along the easy-axis is two orders of magnitude larger than exchange in the basal plane $J'$, which involves two Cl\textsuperscript{-}-ions (as compared to one along c).

(hexagonal space group $P6_3/mmc$). As in other members of the ABX\textsubscript{3} family, CsNiCl\textsubscript{3}, CsMnI\textsubscript{3}, CsNiBr\textsubscript{3} and RbNiBr\textsubscript{3}, the magnetic Ni\textsuperscript{2+}-ions form strongly coupled chains along the crystallographic $c$-axis. The chains are characterized by an intrachain exchange parameter $J$, which is much larger than the interchain exchange parameter $J'$ because magnetic exchange in the basal plane is mediated via two X-ions compared to only one along $c$, as pictured in Fig. 2. $J'/J = 0.38 K/23.8 K = 1.6 \cdot 10^{-2}$ in RbNiCl\textsubscript{3}. The magnetic behavior therefore is quasi 1D. At $T_N \simeq 11 K$, there is a phase transition into a 3D magnetically ordered structure.

Magnetic ordering in RbNiCl\textsubscript{3} can be discussed in the context of other members of the ABX\textsubscript{3} family. In e.g. CsNiCl\textsubscript{3}, two successive phase transitions are found in neutron scattering, magnetic birefringence and specific heat capacity experiments and display 3D XY-critical behavior with the corresponding critical exponent\textsuperscript{3,18}, as predicted by Kawamura. For RbNiCl\textsubscript{3}, with most experimental techniques just one transition is observed. The criticality of this transition is not clear: Different meth-
methods obtained disagreeing values of the critical exponents and accordingly different universality classes have been proposed for the transition. None of the experimentally determined values agree with the prediction for the 3D XY class. Table III summarizes experimental techniques and the values determined for $T_N$, $\alpha$ and $\beta$, as found in literature. Apart from a neutron scattering study by Oohara et al., all measuring techniques report only one phase transition. The temperature resolution in all experiments was better than 0.02 K, considerably smaller than 0.15 K, claimed as the distance between $T_{N1}$ and $T_{N2}$ in the neutron scattering study. The anomalies in all techniques (except [8]) appear very sharp while the overlap of two close lying divergences would lead to a rounded and broad anomaly. Furthermore, the measured critical exponents do not coincide with the predicted 3D XY-critical behavior. If the two transitions would fall together at the same temperature, the transition from the paramagnetic directly into the chiral ordered state should show $n=3$ chiral exponents, against the experimental evidence.

RbNiCl$_3$ has a very small Ising anisotropy $D$, as compared to other members of the ABX$_3$ family. We argue that the pronounced Heisenberg character plays the key role for the understanding of phase transitions and criticality in RbNiCl$_3$. In the next section, we present and discuss the results of our high resolution LMB experiments.

### III. RESULTS

The linear birefringence $n_{ac} = n_c - n_a$ has been measured using a Sénarmont set-up with a He-Ne laser at $\lambda=632.8$ nm. Before and behind the sample, apertures with a diameter of 0.3 mm were installed. The sensitivity of the Sénarmont set-up was increased by modulating the incoming polarisation with 50 kHz and lock-in detection of the intensity. Single crystals of RbNiCl$_3$ were grown by the Bridgeman method. The slightly hygroscopic samples were prepared by cleaving in a glovebox under He-atmosphere. The natural cleavage planes contain the c-axis, and correspond probably to {1010}. The typical sample size was 4x4x1.5 mm$^3$. The cleft samples were used without further polishing and were mounted stress-free in an optical $^4$He continuous flow cryostat with a temperature stability of 0.001 K. The sample temperature was measured with a Cernox semiconductor thermometer in lock-in technique with a relative accuracy of 0.001 K.

Under certain conditions, the derivative $dn_{ac}/dT$ is proportional to the magnetic part of the specific heat capacity, see e.g. [21] and references therein. This relation is in particular valid close to the phase transitions of the antiferromagnetic triangular $\text{ABX}_3$ compounds and with and without easy-axis anisotropy, as CsNiCl$_3$ and RbNiCl$_3$. In the temperature range of the phase transition in RbNiCl$_3$ at $T_N \approx 11$ K, the specific heat capacity is already dominated by contributions of the crystal lattice. The critical properties of the magnetic specific heat are therefore difficult to measure in a standard specific heat capacity setup. Here the birefringence is an elegant way to determine the critical exponent $\alpha$ as well as the amplitude ratio $A^+/A^−$ of the critical part of the specific heat capacity above and below the phase transition.


\[
\frac{dn_{ac}}{dT} = A^\pm \left| \frac{T - T_N}{T_N} \right|^{-\alpha^\pm}.
\]
FIG. 3: Temperature dependence of the birefringence $n_{ac} = n_c - n_a$ over a broad temperature range. The inset shows the range of the phase transition in magnification. The transition is marked by an arrow.

FIG. 4: Temperature-derivative of the critical part of the birefringence, $dn_{ac}/dT$, which is proportional to the magnetic specific heat. The solid line is the resulting fit with Eq. (2).

Figure 4 shows $dn_{ac}/dT$, the solid line is a fit after Eq. (2). The noncritical contribution due to 1D correlations and lattice natural birefringence was taken into account by a polynomial of the form $a + bT + cT^2 + dT^3 + eT^4$ which was subtracted from the data. We observe only one transition as the fitted transition temperatures for the range below and above $T_N$ perfectly coincide. The good temperature resolution allows to measure as close to the phase transition as $10^{-4}$ in reduced temperature, considerably closer than all previous experiments. Even if two different transition temperatures were allowed for the high and the low temperature side, they converge to a single one in the fit. We do not observe any signs of crossover effects. To check the quality of the fits, Fig. 5 shows log-log plots of the critical part of $n_{ac}$ vs. reduced temperature $|t| = |(T - T_N)/T_N|$ for (a) $T > T_N$ and (b) $T < T_N$. Solid lines are fits with Eq. (2), the fitted values for $\alpha^\pm$, $T_N$ and the ratio $A^+/A^-$ are given in the figure.

FIG. 5: Log-log plots of the critical part of the birefringence $n_{ac}$ vs. reduced temperature $|t| = |(T - T_N)/T_N|$ for (a) $T > T_N$ and (b) $T < T_N$. Solid lines are fits with Eq. (2), the fitted values for $\alpha^\pm$, $T_N$ and the ratio $A^+/A^-$ are given in the figure.

The unusual behavior of RbNiCl$_3$ might be explained by a lift of degeneracy of the magnetic exchange interactions in the hexagonal basal plane. This scenario has been discussed for other ABX$_3$ compounds in e.g. [3,22].
ment angular resolution of $10'$ nor the appearance of additional superlattice reflections which are not indexed by the tripled hexagonal cell. Even a small orthorhombic or monoclinic distortion would lead to the appearance of Bragg peaks at former forbidden positions and should have been detected. Our measurements therefore confirm the previous results by Yelon and Cox. Moreover, the zero-field birefringence $n_{a,b}$ in the hexagonal basal plane vanishes, which independently excludes any orthorhombic or monoclinic distortion in the ordered phase.

IV. DISCUSSION

The question arises, why RbNiCl$_3$ - which orders into the same magnetic structure as CsNiCl$_3$ - does not show two successive phase transitions and the predicted chiral critical behavior. Two successive phase transitions can be excluded from our high resolution birefringence measurements as well as from most of the previously reported experimental results. Close lying divergences due to two close lying phase transition should lead to a rounded anomaly in the measurements. But even in the high resolved data of Fig. 4 the anomaly remains sharp and pronounced confirming the single phase transition observed in a previous LMB study and other techniques (see the listing in Tab II). The critical exponent $\alpha$ and the ratio $A^+/A^-$ correspond to conventional Heisenberg critical behavior and therefore point to a disordered chirality below $T_N$. A vanishing chirality due to a collinear structure can be excluded from the structural data. If there was only one transition, connected with ordering of the spin components parallel and perpendicular to the 1D axis but no static ordering of the chirality, the corresponding transition should indeed show conventional Heisenberg critical behavior like for antiferromagnets on rectangular lattices. We argue in the following that spin fluctuations suppress long ranged chiral order in RbNiCl$_3$ below $T_N$.

The chirality, which basically takes into account the sense of rotation of the spin direction on a chosen triangle, is defined as:

$$\vec{\kappa} = \frac{2}{3\sqrt{3}} (S_i \times S_j + S_j \times S_k + S_k \times S_i).$$  \hspace{1cm} (3)

Figure 7 shows the ordered spin structure of RbNiCl$_3$ in the hexagonal basal plane, as proposed in the literature. The spins lie in a [001][110] plane with 2/3 of the spins canted away from $c$ by an angle $\theta$. $\theta$ depends on the ratio $D'/J$ and is determined to $\theta=57.5^\circ$ in RbNiCl$_3$, very close to the ideal value of 60$^\circ$. In this model, the chirality $\vec{\kappa}$ is long ranged ordered and changes sign from one to the neighboring triangle, respectively. Anti-phase domains of the chirality contribute equally in a neutron scattering experiment. Oohara and Iio investigated the RbNi$_{1-x}$Co$_x$Cl$_3$ system with LMB. By replacing Ni$^{2+}$ by Co$^{2+}$, the magnitude of the Ising anisotropy $D$, which is very small in pure RbNiCl$_3$ (70% that of CsNiCl$_3$), can

FIG. 6: Laue-photographs taken at different temperatures. (a) $T=20$ K, above phase transition; (b) $T=2$ K, in the magnetically ordered phase (some of the magnetic superlattice reflections are marked by the arrow); (c) $T=2$ K with predicted Laue-pattern.

Considering the crystal structure of RbNiCl$_3$, as pictured in Fig. 2 a lift of degeneracy is inseparable from changes in the crystal lattice. We therefore carried out supplementary single-crystal neutron diffraction experiments at the new Vivaldi Laue-diffractometer at the high flux reactor of the ILL in Grenoble, France, to detect a possible change in the lattice symmetry below $T_N$. Vivaldi’s large image plate thereby allows to survey large areas of reciprocal space to detect possible superlattice reflections in the ordered phase. Typical sample crystals of about 1x1x2 mm$^3$ were mounted in a helium orange cryostat. We took exposures at $T=20$ K, in the paramagnetic, and in the ordered phase, at 2 K. The corresponding Laue patterns are shown in Fig. 6. The reflections of the $T=20$ K exposure in Fig. 6 (a) could be indexed by a primitive hexagonal cell with lattice parameters $a=6.93\AA$ and $c=5.89\AA$. The reflections in the magnetically ordered phase can be described in terms of a tripled hexagonal cell $(a\sqrt{3}, a\sqrt{3}, c)$. We could not detect any splitting of the reflections below the phase transition within the experi-
gradually be increased. With increasing $D$, two anomalies become visible in the LMB experiments and the distance $T_{N1}-T_{N2}$ increases. The latter study clearly shows that the small Ising anisotropy $D$ plays the crucial role for the understanding of criticality and phase transitions in RbNiCl$_3$. It also proves that LMB is capable to detect the upper transition, if it exists.

The anisotropy $D$ confines the 120° spin structure in the $ac$ plane. Depending on the ratio $D/J'$, the structure might exhibit an additional degree of freedom connected with the rotation of the 120° structure in the $ac$ plane. This quasidegeneracy has been predicted and experimental evidence was found for the case of CsNiCl$_3$. The energy barrier for a rotation of the spin-star in the $ac$ plane is of the order of $D(D/6J')^2$. Miyashita suggested, this quasidegeneracy exists, if $(D/J')<1$ $(D/J'=0.06$ in RbNiCl$_3$). Even though $D_{RbNiCl3} = 0.7D_{CsNiCl3}$, $D(D/6J')^2$ for RbNiCl$_3$ is just 7% of that of CsNiCl$_3$; the quasidegeneracy should therefore be strongly enhanced in the paramagnetic phase of pure RbNiCl$_3$.

NMR and measurements of the specific heat capacity give evidence for strong spin fluctuations also in the ordered phase of RbNiCl$_3$. Figure 8 schematically shows the two basic spin relaxation mechanisms. Type I fluctuations are rotations of the spin-star around an axis perpendicular to the spin plane, i.e. parallel to the chirality vector $\vec{\kappa}$. This is the quasidegeneracy that has been discussed above. As indicated in the figure, these fluctuations preserve the chirality of the triangle; $\vec{\kappa}$ can still show long ranged order. All fluctuations with axis of rotation perpendicular to $\vec{\kappa}$ (Type II fluctuations) change the sign of $\vec{\kappa}$. If these fluctuations occur incoherently, $\vec{\kappa}$ cannot order. The phase transition should be of conventional type, as suggested by the LMB experiment.

Type II fluctuations seem not to depend directly on the Ising anisotropy $D$ because the canting angle of the respective spins does not change during the rotation. Their incoherent occurrence in the ordered structure, however, may be emphasized by the presence of Type I fluctuations. When the Ising anisotropy is enlarged in CsNiCl$_3$ or in the RbNi$_{1-x}$Co$_x$Cl system, the contribution of Type II fluctuations is obviously negligible, as these compounds show chiral ordering as predicted by theory. This seems to imply that Type II fluctuations play a major role only when Type I fluctuations are already strongly enhanced (as in pure RbNiCl$_3$).

The basic idea of fluctuations which on the one hand preserve (Type I) and on the other hand suppress (Type II) long ranged chiral order seems to account well for phase transitions and critical behavior observed in RbNiCl$_3$. The separate ordering of the spin components parallel to the 1D axis, is presumably suppressed by Type I fluctuations; Type II fluctuations do not affect the projection of the magnetic moment onto the $c$-axis. But fluctuations of Type II might suppress ordering of the chirality $\vec{\kappa}$ at the phase transition $T_N$ where the magnetic moment shows 3D ordering (whereas Type I fluctuations have no effect on the sign of $\vec{\kappa}$). If both types of fluctuations are strongly enhanced, we imagine that the domain walls between chirality domains of opposite sign move freely through the otherwise magnetically long-range ordered structure. If the chirality domain walls in the ordered phase behave liquid-like, the transition should show conventional Heisenberg critical behavior, as it is observed in the LMB experiment. In this language, the chirality domain walls in CsNiCl$_3$ or CsMnBr$_3$ are quasi-static. This liquid-like behavior of the domain walls, which leads to a different phase diagram and different critical behavior, as compared to other members of the ABX$_3$ family, must crucially depend on the almost perfect Heisenberg character of RbNiCl$_3$.

**V. CONCLUSIONS**

We present a linear magnetic birefringence study in RbNiCl$_3$. Our high resolution determination of the critical parameters $\alpha$ and the amplitude ratio $A^+/A^-$ shows conventional Heisenberg critical behavior like antiferro-
magnets on rectangular lattices (which have no ordered chirality) as opposed to theoretical predictions. There is just one phase transition in RbNiCl$_3$. From a neutron diffraction study we can exclude a structural phase transition and a lift of the degeneracy of the magnetic exchange interactions in the basal plane at $T_N$. We discuss RbNiCl$_3$ in the framework of previous experimental and theoretical results and other members of the ABX$_3$ family. We finally argue that spin fluctuations lead to the unusual behavior of RbNiCl$_3$. A separate phase transition of the spin component parallel to the easy-axis might be suppressed by spin fluctuations with axis of rotation parallel to the chirality vector $\vec{\kappa}$ (Type I fluctuations). Fluctuations of Type II, with axis of rotation perpendicular to $\vec{\kappa}$, presumably suppress long ranged order of the chirality $\vec{\kappa}$ below $T_N$. The resulting single phase transition shows conventional Heisenberg critical behavior, as evidenced by the critical exponents and phase transitions observed in the LMB experiment.

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