Quenched chirality in $RbNiCl_3$: Linear birefringence and neutron diffraction

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The critical behavior 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 behavior of $RbNiCl_3$, a stacked-triangular Heisenberg antiferromagnet with very small Ising anisotropy. From linear birefringence experiments we determine the specific-heat exponent α 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 behavior, 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 behavior of stacked-triangular antiferromagnets. In $RbNiCl_3$ the ordering of the chirality is suppressed by strong spin fluctuations, yielding a different phase diagram, as compared to, e.g., $CsNiCl_3$, where the Ising anisotropy prevents these fluctuations.

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I. INTRODUCTION

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°. Neighbors along the hexagonal c axis are aligned antiparallel.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

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

Here, $J>0$ denotes the antiferromagnetic exchange interaction between nearest neighbors along the symmetry axis and $J'>0$ the antiferromagnetic interaction between nearest neighbors on a triangle. The single ion anisotropy constant D favors an easy axis $(D>0)$ or plane $(D<0)$. Kawamura[1] 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 behavior, 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, such as, e.g., generic nonuniversal behavior.[2] Table I lists Kawamura's predictions for the critical exponents α, β, γ, and δ and the ratio A^+/A^- of the amplitudes above and below T_N for antiferromagnets on rectangular and triangular lattices as a survey. Here rectangular stands for all collinear structures, as they usually develop on a square or orthorhombic lattice.

ABX_3 compounds with easy-axis anisotropy, such 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., Ref. 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. 1.

TABLE I. Critical exponents for antiferromagnets on square and triangular lattices after Kawamura, see Refs. 3–5, and references therein.

		α	β	γ	ν	A^+/A^-
□	Ising	0.1098(29)	0.325(1)	1.2402(9)	0.6300(8)	0.55
	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)

The two zero-field phase transitions should show 3D *XY* critical behavior.[1] On a rectangular lattice, there is just one transition with Ising-type critical behavior.[3–5]

In order to clarify the number of phase transitions in $RbNiCl_3$, and their criticality, we performed linear magnetic birefringence (LMB) experiments with high temperature resolution to measure the critical exponent α and the amplitude ratio A^+/A^-. The paper is organized as follows. The properties of $RbNiCl_3$ are discussed in Sec. II. Experimental details of the birefringence setup are presented in Sec. III, the LMB results and the outcome of a supplementary neutron diffraction study are shown and discussed in Sec. IV. The anomalous behavior of $RbNiCl_3$ as compared to other members of the abovementioned ABX_3 family, is discussed in Sec. V.

II. RBNICL$_3$

$RbNiCl_3$ is a quasi-1D $S=1$ Heisenberg antiferromagnet with a weak Ising anisotropy on a triangular lattice (hexagonal space group $P6_3/mmc$). As in other members of the ABX_3 family, $CsNiCl_3$, $CsMnI_3$, $CsNiBr_3$, and $RbNiBr_3$, the magnetic Ni^{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. $J'/J = 0.38\ \text{K}/23.8\ \text{K} = 1.6\times10^{-2}$ in $RbNiCl_3$.[6] The magnetic behavior therefore is quasi-1D.

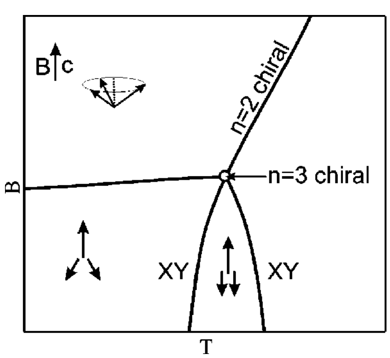

FIG. 1. Predicted phase diagram for ABX_3 with easy-axis anisotropy. In zero magnetic field, two successive phase transitions are expected, connected with ordering of the spin components parallel and perpendicular to the hexagonal c axis at T_{N1} and T_{N2} ($<T_{N1}$), respectively. Both transitions should show *XY* critical behavior.

At $T_N \simeq 11$ K, there is a phase transition into a 3D magnetically ordered structure. Magnetic ordering in $RbNiCl_3$ can be discussed in the context of other members of the ABX_3 family. In, e.g., $CsNiCl_3$, 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 exponents,[3,7] as predicted by Kawamura. For $RbNiCl_3$, most experimental report only one transition. The criticality of this transition is not clear: Different methods obtained disagreeing values of the critical exponents and accordingly different universality classes have been proposed for the transition. The experimentally determined values do not coincide with 3D *XY* critical behavior. Table II summarizes experimental techniques and the values determined for T_N, α and β, as found in the literature. Apart from a neutron scattering study by Oohara *et al.*,[8] 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 for Ref. 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

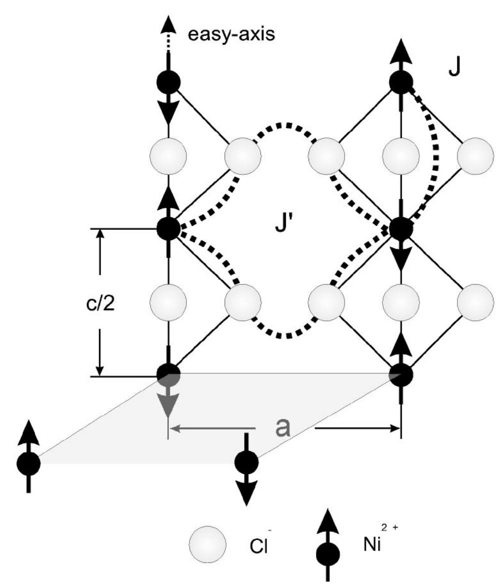

FIG. 2. In $RbNiCl_3$ magnetic exchange J along the easy axis is two orders of magnitude larger than exchange in the basal plane J', which involves two Cl^- ions (as compared to one along c).

TABLE II. Reported results for the zero-field phase transition in $RbNiCl_3$. Values for the relation A^+/A^- are not given in the references.

Technique	Ref.	T_N (K)	α	β
neutron diffraction	14	11.15		$\beta=0.30\pm0.01$
	8	11.11,11.25		$\beta_{\parallel,\perp}=0.27+0.01,\ 0.28\pm0.01$
LMB	12	11		
	15	11.3	0.06 ± 0.04	
	20	11		
NMR	21	11.18,11.36 (?)		
susceptibility, torque	17 and 22	11.38		
magnetization, susceptibility	23	11		
thermal expansion	24	11.2		
specific heat capacity	24 and 25	11.0		

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. EXPERIMENTAL

Single crystals of $RbNiCl_3$ were grown by the Bridgman 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 $\{10\underline{1}0\}$. The typical sample size was $4\times4\times1.5\ \mathrm{mm}^3$ (with thickness $d=1.5$ mm). The linear birefringence $n_{ac}=n_c-n_a$ has been measured using a Sénarmont setup[9,10] with a He-Ne laser at $\lambda=632.8$ nm. 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. Before and behind the sample, apertures with a diameter of 0.3 mm were installed. The sensitivity of the Sénarmont setup was increased by modulating the incoming polarisation with 50 kHz and lock-in detection of the intensity.

Under certain conditions, the derivative dn_{ac}/dT is proportional to the magnetic part of the specific-heat capacity, see, e.g., Ref. 11 and references therein. This relation is in particular valid close to the phase transitions of the antiferromagnetic triangular ABX_3 compounds with and without easy-axis anisotropy, such as $CsNiCl_3$ and $RbNiCl_3$. In the temperature range of the phase transition in $RbNiCl_3$ at $T_N\approx11$ 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 α as well as the amplitude ratio A^+/A^- of the critical part of the specific heat capacity above and below the phase transition.

IV. RESULTS

Figure 3 shows the temperature dependence of n_{ac} over a broad temperature range. At high temperatures, n_{ac} linearly decreases with lowering temperature. Below about $T=70$ K there is distinct deviation from linear behavior due to the onset of short-ranged 1D correlations along the Ni chains.[12] The inset in Fig. 3 shows the temperature range of the 3D phase transition in magnification. The onset of 3D correlations close to $T_N=10.89$ K, which finally leads to a 3D mag-

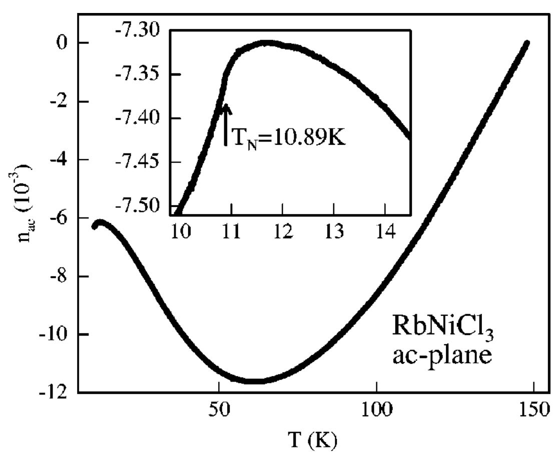

FIG. 3. Temperature dependence of the birefringence $n_{ac}=n_c-n_a$ over a broad temperature range. The inset shows the range of the phase transition in magnification. The transition is marked by an arrow.

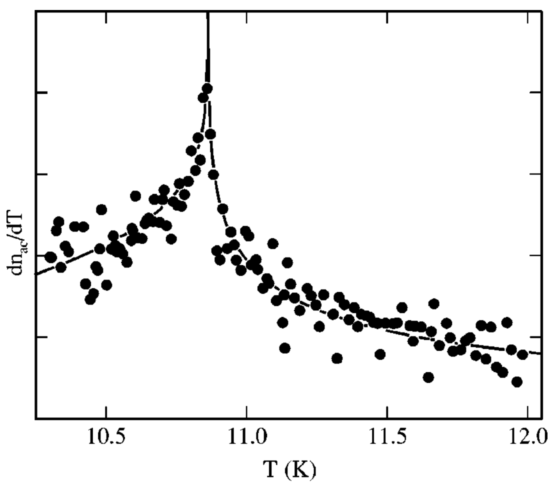

FIG. 4. Temperature derivative of the critical part of the birefringence dn_{ac}/dT, which is proportional to the magnetic specific heat. The solid line is the resulting fit with Eq. (2).

netically ordered structure, is indicated by the drop of the birefringence below 11 K.

Close to the phase transition the derivative of the birefringence with respect to temperature is described by a power law

$$\frac{dn_{ac}}{dT} = A^{\pm}\left|\frac{T-T_N}{T_N}\right|^{-\alpha^{\pm}} + \text{noncritical contribution}. \quad (2)$$

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 one 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 $T \lesseqgtr T_N$ in the range close to the phase transition. T_N is determined to be $T_N=10.888\pm0.001$ K, the values for α from the high- and the low-temperature side to be $\alpha^+=-0.084\pm0.001$ and $\alpha^-=-0.080\pm0.007$. The ratio A^+/A^- obtained is 1.38 ± 0.07. Comparing these values with those from Table I, the determined critical exponent and A^+/A^- agree remarkably well with those of a Heisenberg antiferromagnet on the rectangular lattice. Chiral or XY behavior, as predicted in the chiral theory, can be excluded. Two close lying successive phase transitions that would lead to a rounded anomaly in the measurements can obviously be excluded by our measurements in Figs. 4 and 5.

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

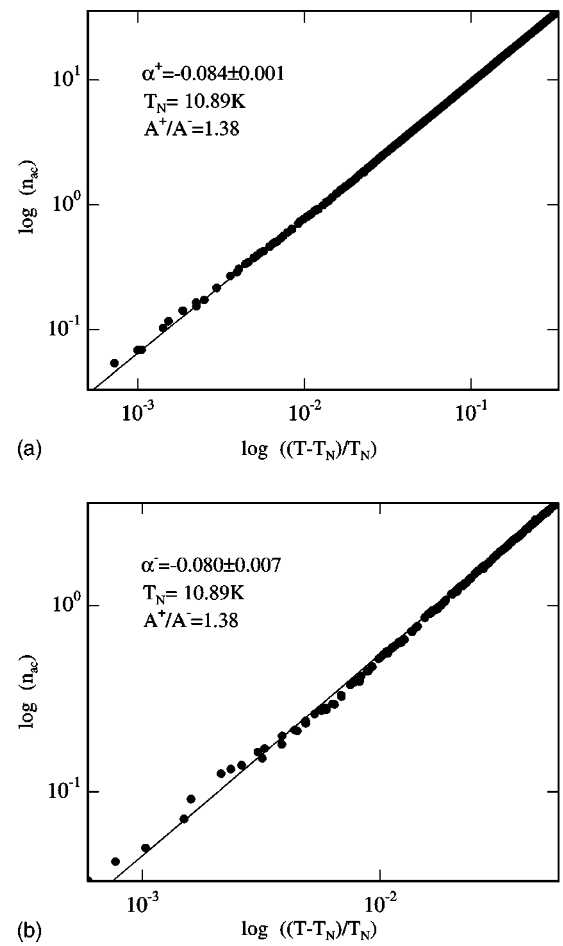

FIG. 5. Log-log plots of the critical part of the birefringence n_{ac} vs reduced temperature $|t|=|(T-T_N)/T_N|$ for (a) $T>T_N$ and (b) $T<T_N$. Solid lines are fits with Eq. (2), the fitted values for $\alpha^{\pm}$, T_N and the ratio A^+/A^- are given in the figure.

for other ABX_3 compounds in, e.g., Ref. 3 and 13. 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 detector allows us to survey large areas of reciprocal space to detect possible superlattice reflections in the ordered phase.

Typical sample crystals of about $1\times1\times2$ mm^3 were mounted in a helium 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 $a=6.93$ Å and $c=5.89$ Å. The reflections in the magnetically ordered phase in Fig. 6(b) can be described in terms of a tripled hexagonal cell $(a\sqrt{3},a\sqrt{3},c)$. Figure 6(c) shows the measured reflections at $T=2$ K and the corresponding simulated Laue pattern superposed. We could not detect any splitting of the reflections below the phase transition within the experimental angular resolution of 10′ nor the appearance of

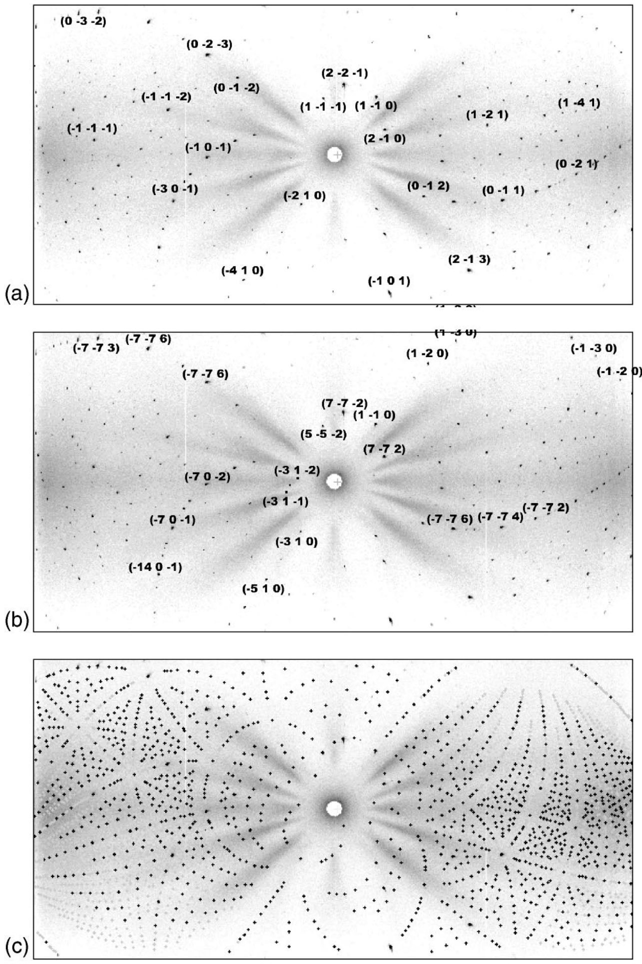

FIG. 6. Laue photographs taken at different temperatures. (a) $T=20$ K, above the phase transition. Some of the reflections are labeled in the hexagonal unit cell with $a=6.93$ Å and $c=5.89$ Å. (b) $T=2$ K, in the magnetically ordered phase. Some reflections are labeled in the tripled magnetic orthorhombic cell with lattice parameters $a=20.78$ Å, $b=12.00$ Å, and $c=5.89$ Å. (c) $T=2$ K with the predicted Laue pattern superposed.

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.[14] Moreover, the zero-field birefringence n_{ab} in the hexagonal basal plane[15] vanishes, which independently excludes any orthorhombic or monoclinic distortion in the ordered phase.

V. DISCUSSION

The question arises, why does $RbNiCl_3$—which orders into the same magnetic structure as $CsNiCl_3$—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 transi-

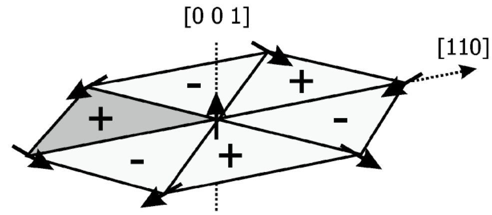

FIG. 7. In the magnetically ordered phase of $RbNiCl_3$, 2/3 of the spins are canted away from c in the [110] direction [as proposed by Yelon and Cox (Ref. 14)]. The chirality $\vec{\kappa}$ changes sign from one triangle to the neighboring triangle.

tion should lead to a rounded anomaly in the measurements. But even in the highly resolved data of Fig. 4, the anomaly remains sharp and pronounced confirming the single phase transition observed in a previous LMB study[15] and other techniques (see the listing in Table II). The critical exponent α 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 similar to 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[16]

$$\vec{\kappa} = \frac{2}{3\sqrt{3}}(\mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_j \times \mathbf{S}_k + \mathbf{S}_k \times \mathbf{S}_i). \qquad (3)$$

Figure 7 shows the ordered spin structure of $RbNiCl_3$ in the hexagonal basal plane, as proposed in the literature by Yelon and Cox.[14] The spins lie in a [001] [110] plane with 2/3 of the spins canted away from c by an angle θ. θ depends on the ratio D/J' and is determined to $\theta=57.5°$ (Ref. 14) in $RbNiCl_3$, very close to the ideal value of 60°. In this model, the chirality $\vec{\kappa}$ is long-ranged ordered and changes sign from one triangle to the neighboring triangle. Note that antiphase domains of the chirality contribute equally in a neutron-scattering experiment.

Oohara and Iio investigated the $RbNi_{1-x}Co_xCl_3$ system[17] 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 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 of detecting the upper transition, if it exists.

The anisotropy D confines the 120° spin structure to 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 *quaside-*

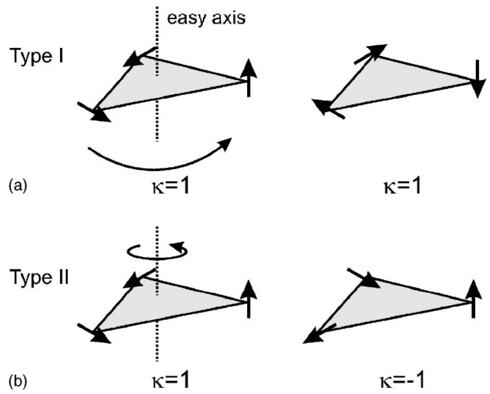

FIG. 8. Fluctuations of the triangle marked in Fig. 7. (a) Type I: Rotations about an axis parallel to the vector of chirality $\vec{\kappa}$ preserve the chirality. (b) Type II: Fluctuations around the easy axis, perpendicular to $\vec{\kappa}$, change sign of the chirality.

generacy has been predicted[18] and experimental evidence was found for the case of $CsNiCl_3$.[19] The energy barrier for a rotation of the spin-star in the *ac* plane is of the order of $D(D/6J')^2$.[20] Miyashita[18] suggested that this quasidegeneracy exists if $(D/J')<1$ ($D/J'=0.06$ in $RbNiCl_3$). Even though $D_{RbNiCl_3}=0.7D_{CsNiCl_3}$, $D(D/6J')^2$ 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 (see Table II) 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 $RbNi_{1-x}Co_xCl$ 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 is observed in the LMB experiment. In this language, the chirality domain walls in $CsNiCl_3$ or $CsMnBr_3$ are quasistatic. 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 $RbNiCl_3$.

VI. CONCLUSIONS

We present a linear magnetic birefringence study in $RbNiCl_3$. Our high-resolution determination of the critical parameters α and the amplitude ratio A^+/A^- show conventional Heisenberg critical behavior similar to antiferromagnets 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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