Linear birefringence behavior of A_2BX_4-type ferroic crystals in the normal-incommensurate phase transition

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Critical behavior of linear birefringence was studied in the normal-incommensurate phase transition of various A_2BX_4-type crystals. We considered the fluctuation effect in the scaling analysis so that we could obtain a precise value of the critical exponent β from linear birefringence measurements. The numerical values of 2β obtained from our experiment were $0.72\sim0.75$ (±0.02). These results are in agreement with the previous theoretical prediction which suggests that the normal-incommensurate phase transition of A_2BX_4-type crystals belongs to the universality class of the three-dimensional (3D) XY model. Beyond the critical region, we observed crossover from 3D XY-like behavior into classical Landau-like behavior, $2\beta=1$, in Rb_2ZnCl_4 and K_2ZnCl_4. [S0163-1829(98)03318-9]

I. INTRODUCTION

Physical properties of A_2BX_4-type crystals have been widely studied by various experimental method because of the interesting characteristics of their incommensurate phase.[1] ($A=$Rb, K, NH_4, $N(CH_3)_4$; $B=$Zn, Mn, Cu, Co; and $X=$Cl, Br.) The A_2BX_4-type crystals have the same phase transition sequence as the normal (N)-incommensurate (IC)-commensurate (C) phase with descending temperature.[1,2] In the N phase, the A_2BX_4-type crystals have β-K_2SO_4-type $Pmcn$ structure.[3,4] In the IC phase, modulated polarization turns up with a period irrational to the period of crystal lattice. Some characteristics of the IC phase of the A_2BX_4-type crystals are given in Table I. In the C phase, some A_2BX_4-type crystals have ferroelectricity or ferroelasticity.

The N-IC phase transition was theoretically studied by Cowley and Bruce, who predicted that the N-IC phase transition belongs to the universality class of the three-dimensional (3D) XY model.[5] Their theoretical study was followed by many experimental investigations, some of which agree with the expectation of the 3D XY model.[6–10] However, there also exist other experimental results which agree with the 3D Ising model rather than with the 3D XY model.[11–13] Up to now, it is not yet conclusive experimentally whether or not the N-IC phase transition of A_2BX_4-type crystals belongs to the universality class of the 3D XY model.[13]

For the investigation of critical phenomena, linear birefringence (LB) is a very useful tool because it has a direct relation to the order parameter of the N-IC phase transition.[14–18] Therefore, the critical exponent β for the order parameter can be obtained from LB measurement and scaling analysis. In our previous studies, we have reported that the LB behavior of some A_2BX_4-type crystals were 3D-XY-like near the N-IC phase transition temperature (T_i).[19–21]

In this paper, we report the LB behavior near the N-IC phase transition of various A_2BX_4-type crystals. The critical LB behavior has been investigated systematically by considering the fluctuation effect and crossover behavior.

II. EXPERIMENTAL PROCEDURE

In this report, we have measured the LB behavior of Rb_2ZnCl_4 (RZC), K_2ZnCl_4 (KZC), $(NH_4)_2ZnCl_4$ (AZC), $[N(CH_3)_4]_2ZnCl_4$ (TMAZC), $[N(CH_3)_4]_2CuCl_4$

TABLE I. Characteristics of the IC phase of A_2BX_4-type crystals studied in this paper. T_i and T_c are the N-IC and the IC-C phase transition temperature, respectively. The ferro-axis means the axis along which spontaneous polarization or spontaneous strain appears. All of the LB behaviors in this study were measured along these ferro-axes. The crystals have ferroelectricity or ferroelasticity in the temperature region given in the parentheses.

	modulation wave vector	T_i	T_c	ferro-axis	
Rb_2ZnCl_4	$\mathbf{q}=(1/3-\delta)\mathbf{c}^*$	302 K	195 K	a axis	ferroelectricity($\leqslant$195 K)
K_2ZnCl_4	$\mathbf{q}=(1/3-\delta)\mathbf{c}^*$	553 K	403 K	a axis	ferroelectricity($\leqslant$403 K)
$(NH_4)_2ZnCl_4$	$\mathbf{q}=(1/4+\delta)\mathbf{c}^*$	406 K	364 K	a axis	ferroelectricity($\leqslant$268 K)
$[N(CH_3)_4]_2ZnCl_4$	$\mathbf{q}=(2/5\pm\delta)\mathbf{a}^*$	297 K	280 K	b axis	ferroelectricity($\leqslant$280 K)
$[N(CH_3)_4]_2CuCl_4$	$\mathbf{q}=(1/3-\delta)\mathbf{a}^*$	296 K	293 K	b axis	ferroelasticity ($\leqslant$293 K)
$[N(CH_3)_4]_2MnCl_4$	$\mathbf{q}=(1/2-\delta)\mathbf{a}^*$	293 K	292 K	b axis	ferroelasticity ($\leqslant$292 K)

(TMACC), and $[N(CH_3)_4]_2MnCl_4$ (TMAMC). Single crystals of A_2BX_4-type materials were grown at 310 K by the slow evaporation method from an aqueous solution of a stoichiometric mixture of $2A$Cl and BCl$_2$ with 10% excess of BCl$_2$. We obtained transparent and colorless single crystals. (TMACC and TMAMC have exceptionally light brown and red colors, respectively.)

The crystals were cut perpendicularly to the ferroelectric (or ferroelastic) axes using a wet string and polished carefully. The ferroelectric (or ferroelastic) axes for each crystal are given in Table I.

The temperature dependence of LB was measured at 632.8 nm wavelength of a He-Ne laser by the Senarmont method with an accuracy of at least 10^{-7}. The LB behavior was measured in the heating run. The temperature heating rate was 0.04 K/min near T_i, and about 0.4 K/min in the temperature region far from T_i. Only for KZC, the temperature heating rate was 0.4 K/min in all of the temperature range.

III. CRITICAL BEHAVIOR OF BIREFRINGENCE

The N-IC phase transition of dielectric materials was studied theoretically by Cowley and Bruce.[5] They proposed the Landau-Ginzburg-Wilson (LGW) Hamiltonian of the N-IC phase transition using the renormalization technique. A_2BX_4-type crystals belong to the case where the star of the soft mode consists of only two wave vectors $\mathbf{q}_s$ and $-\mathbf{q}_s$. When $\mathbf{q}_s$ is incommensurate, the obtained LGW Hamiltonian was of the effective Hamiltonian of the 3D XY model.[5] (The XY model means the isotropic $n=2$ Heisenberg ferromagnet.)

The critical behavior of specific heat (C_p), order parameter (Q), and dielectric susceptibility (ϵ) can be expressed as

$$C_p \sim |t|^{\alpha}, \quad Q \sim |t|^{\beta}, \quad \epsilon \sim |t|^{\gamma}, \tag{1}$$

with the reduced temperature $t \equiv [(T-T_i)/T_i]$ and the critical exponents α, β, and γ. The critical exponent β of the 3D XY model was predicted by Majkrzak *et al.* as $2\beta = 0.70 \pm 0.04$.[9] If we accept their prediction, the order parameter behavior of A_2BX_4-type crystals can be expected to be $Q^2 \sim |t|^{2\beta} \sim |t|^{0.70\pm 0.04}$ in the critical region of the N-IC phase transition.

In the N-IC phase transition of A_2BX_4-type crystals, the order parameter Q is the amplitude of the modulated polarization. The incommensurate modulation near T_i can be described by the plane wave approximation[1,17]

$$P_k(x) = 2Q \cos \mathbf{q}x. \tag{2}$$

In Eq. (2), P_k is the spontaneous polarization along the k axis and $\mathbf{q}$ is the modulation wave vector. The square of the order parameter Q^2 can be easily obtained from the spatial average of the modulated polarization $\langle P_k(x)^2 \rangle$ by

$$\langle P_k(x)^2 \rangle = Q^2. \tag{3}$$

A relation between the incommensurately modulated polarization and LB, $\Delta n_{ij} \equiv n_i - n_j$, is given by[14,17]

$$\Delta n_{ij} = \frac{1}{2}(n_j^3 g_{jk} - n_i^3 g_{ik}) \langle P_k(x)^2 \rangle + \Delta n_{ij0}, \tag{4}$$

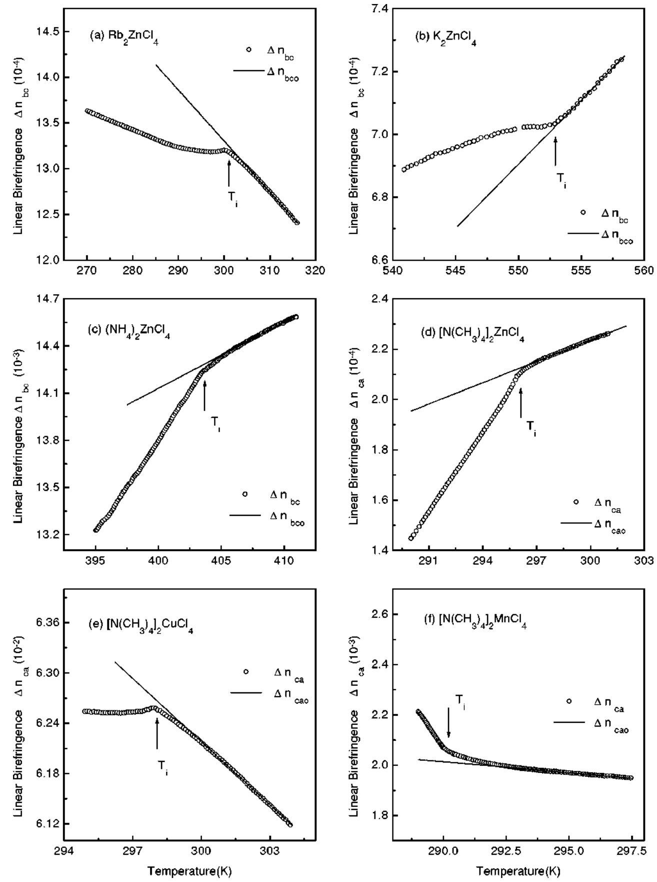

FIG. 1. The temperature dependence of Δn_{ij} (open circle) near T_i for (a) RZC, (b) KZC, (c) AZC, (d) TMAZC, (e) TMACC, and (f) TMAMC. T_i is indicated by an arrow. The solid line represents the base line which corresponds to Δn_{ij0}.

where i,j,k are crystal axes, n_i the refractive index along the i axis, g_{jk} and g_{ik} the electro-optic coefficients, and Δn_{ij0} the photoelastic and thermoelastic contributions.

From Eqs. (1)–(4), we can expect[15]

$$\Delta n \equiv |\Delta n_{ij} - \Delta n_{ij0}| \sim \langle P_k(x)^2 \rangle \sim Q^2 \sim |t|^{2\beta}. \tag{5}$$

Thus, the critical exponent β can be determined from the LB measurement.

IV. RESULTS AND DISCUSSION

The results of LB measurement given in Fig. 1 shows an obvious kink at T_i which is indicated by an arrow. These results agree well with the previous LB studies of A_2BX_4-type crystals.[22–27]

For the investigation of the critical behavior of Δn and Q^2, it is important to determine correctly the base line Δn_{ij0} of Eq. (4). Because $P_k(x)=0$ in the N phase, a base line Δn_{ij0} is expected to be obtained from linear fitting of LB behavior in the N phase (solid line in Fig. 1). It is assumed that the base line Δn_{ij0} can be extrapolated into the IC phase.

In addition, the fluctuation effect on the LB behavior is expected to be observed in the N phase. Accounting for the time-averaged static order parameter $\langle Q \rangle_t$, we can set $Q = \langle Q \rangle_t + \delta Q$, where δQ represents fluctuation amplitude.

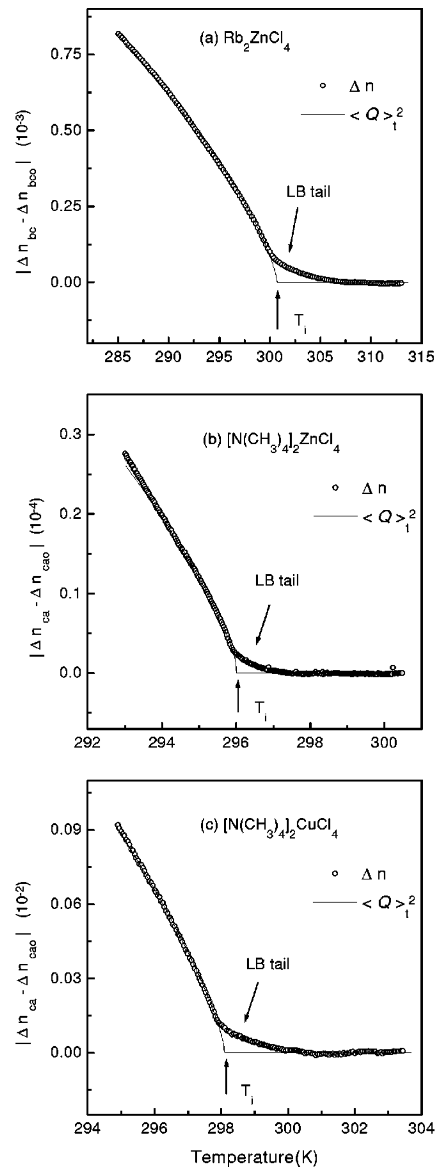

FIG. 2. The temperature dependence of $\Delta n \equiv \Delta n_{ij} - \Delta n_{ij0}$ (open circle) for (a) RZC, (b) TMAZC, and (c) TMACC. The solid line represents the temperature dependence of the static order parameter $\langle Q \rangle_t^2$.

As mentioned in Eq. (4), LB is proportional to the time-averaged square of the order parameter $\langle Q^2 \rangle_t$,[28]

$$\Delta n \sim \langle Q \rangle_t^2 + \langle \delta Q^2 \rangle_t . \tag{6}$$

The term $\langle Q \rangle_t$ must be zero in the N phase, and the effect of the order parameter fluctuation $\Delta n \sim \langle \delta Q^2 \rangle_t$ is also expected to be observed in the LB behavior of the N phase.

In Fig. 2, the existence of the LB tail can be easily observed just above T_i in the N phase. The relation between the LB tail and the fluctuation was discussed by Courtens and Kleemann, Schäfer, and Nouet who explained it in terms of anisotropic fluctuation.[29,30] The LB tail of A_2BX_4-type crystals can be similarly explained by the fluctuation effect. Microscopic origin of the anisotropic fluctuation in A_2BX_4-type crystals can be deduced from previous x-ray diffraction and neutron scattering experiments, the results of which indicate that the fluctuation consists of atomic displacements of the BX_4 tetrahedra.[1]

The LB tail must be correctly taken into account to determine the base line Δn_{ij0}. Because the LB tail is not a part of Δn_{ij0} but caused by the fluctuation effect, for obtaining a correct Δn_{ij0}, it was excluded from the linear fitting of the LB behavior in the N phase. In many previous LB studies on A_2BX_4-type crystals, the existence of the LB tail is overlooked in the linear fitting for Δn_{ij0}.[21] It is thought to be the reason why our LB studies are different than the previous ones.

An initial value of T_i was determined at which we can find the kink by a direct inspection of the LB behavior. Then, with a small variation of the initial value of T_i, we were able to get a precise T_i where the best linear fitting of a $\log_{10}\Delta n$-$\log_{10} t$ plot was obtained. The T_i's determined in this way are given in Table II, and were treated as a fixed parameter afterwards.

Figure 3 shows the results of the scaling analysis, i.e., the $\log_{10}\Delta n$-$\log_{10} t$ plot of LB behavior. The exponent 2β was obtained from the inclination of the log-log plot in a proper temperature region. The obtained value of 2β is also presented for each crystal in Table II. The 2β's estimated from LB measurement are distributed in the range $2\beta = 0.72$–0.75 (± 0.02). Within experimental error, these results are in good agreement with the theoretical prediction of the 3D *XY* model, $2\beta = 0.70 \pm 0.04$, and with some previous LB studies.[18,31] As mentioned in Sec. I, it has not yet been resolved whether the N-IC phase transition is *XY*-like or Ising-like.[13] It can be thought that our LB study strongly supports the 3D *XY* model. For the exponent α and γ, similar results supporting the 3D *XY* model were also obtained by recent specific heat and nuclear magnetic resonance (NMR) studies.[6,10]

TABLE II. The numerical values of the critical exponent 2β obtained from this LB study. T_i was determined from the scaling analysis as described in the text. The $\log_{10} t_{\mathrm{cr}}$ is the temperature where the 2β line and the $2\beta^*$ line meet in Fig. 3.

	β	T_i	β^*	$\log_{10} t_{\mathrm{cr}}$
Rb_2ZnCl_4	$2\beta = 0.72 \pm 0.02$	300.7 K	$2\beta^* = 0.99 \pm 0.02$	-0.71
K_2ZnCl_4	$2\beta = 0.75 \pm 0.02$	553.1 K	$2\beta^* = 1.02 \pm 0.02$	-0.92
$(NH_4)_2ZnCl_4$	$2\beta = 0.73 \pm 0.02$	403.6 K	$2\beta^* = 0.89 \pm 0.02$	-2.34
$[N(CH_3)_4]_2ZnCl_4$	$2\beta = 0.73 \pm 0.02$	296.1 K	$2\beta^* = 0.88 \pm 0.02$	-2.13
$[N(CH_3)_4]_2CuCl_4$	$2\beta = 0.75 \pm 0.02$	298.1 K		
$[N(CH_3)_4]_2MnCl_4$	$2\beta = 0.72 \pm 0.02$	290.3 K		

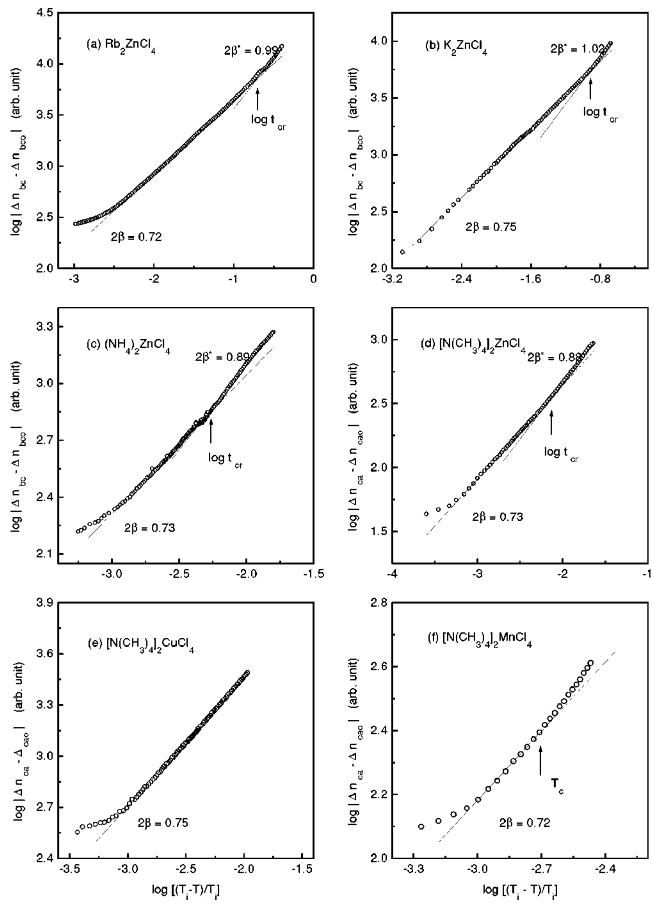

FIG. 3. The result of scaling analysis (open circle) of LB behavior for (a) RZC, (b) KZC, (c) AZC, (d) TMAZC, (e) TMACC, and (f) TMAMC. The solid line represents the slope of the exponent 2β.

There have been some reports that the exponent obtained from the LB behavior is not 2β but another exponent. Régis *et al.* reported that LB behaved as $\Delta n \sim |t|^{0.87\pm0.02}$ for TMAZC.[32] In order to explain the result, they introduced C_2 symmetry breaking in the N-IC phase transition of A_2BX_4-type crystals. The LB would not behave as $\Delta n \sim |t|^{2\beta} \sim |t|^{0.70}$, but as $\Delta n \sim |t|^{\tilde{\beta}} \sim |t|^{0.845}$ under the assumption of symmetry breaking.[15] However, their result is inconsistent with our results. In addition, the C_2 symmetry breaking was not observed in x-ray diffraction studies in the N-IC phase transition. A_2BX_4-type crystals have $Pmcn$ structure in the N phase, and still preserve the $Pmcn_{ss\bar{1}}$ or $Pmcn_{1s\bar{1}}$ structure in the IC phase.[3,4]

On the basis of a critical review of existing LB theory ($\Delta n \sim |t|^{2\beta}$), Ivanov *et al.* asserted that the existing LB theory was wrong and LB had to behave as $\Delta n \sim |t|^{1-\alpha}$.[33] If we accept their assertion, our results $\Delta n \sim |t|^{0.72\sim0.75}$ must result in $\alpha = 0.25$–0.28 (± 0.02), which does not agree with any α of specific heat studies on A_2BX_4-type crystals.[6,11]

Because of the inconsistency between these previous LB experiments, Levanyuk even suggested that 2β of A_2BX_4-type crystals can vary in the range $\frac{1}{2} < 2\beta < 1$ in the vicinity of the tricritical point.[1] But, the 2β's obtained from this LB study are the same for various A_2BX_4-type crystals within experimental error. (This means that 2β does not depend on each A_2BX_4-type crystals but has a regular value.)

It is inferred that the origins of the inconsistency were due to overlooking the LB tail and taking an inappropriate base line. The exponent obtained from the LB study is 2β as described in Eq. (5).

In Figs. 3(a) and 3(b), $\log_{10}\Delta n$ follows the $2\beta = 0.72\sim0.75$ line in a wide temperature region $\log_{10} t \leqslant -1$. This means that the critical region of RZC and KZC is on the order of tens of a degree, and very wide compared to KH_2PO_4 and Ising-type ferroelectric materials.[34] The temperature region where the classical Landau theory is valid can be determined by the Ginzburg criterion[34,35]

$$\frac{k_B}{16\pi e \Delta C}\xi(0)^{-3} \ll |t|^{1/2}. \quad (7)$$

In Eq. (7), k_B is the Boltzmann constant, ΔC the jump in heat capacity per unit volume at T_i, and $\xi(0)$ the zero temperature correlation length. In the view point of the Ginzburg criterion, the range of the critical region is mainly dependent on $\xi(0)^{-3}$ of Eq. (7).[35] The wide critical region of RZC and KZC indicates that $\xi(0)$ is very short compared to Ising-type ferroelectric materials. It is inferred that the characteristics of the N-IC phase transition of A_2BX_4-type crystals are dominated by short-range interactions.

Generally speaking, the factor $(n_j^3 g_{jk} - n_i^3 g_{ik})$ of Eq. (4) is also dependent on temperature. However, for RZC and KZC, the $\log_{10}\Delta n$ agrees very well with the 2β line in the wide temperature range, so that the factor $(n_j^3 g_{jk} - n_i^3 g_{ik})$ must have negligible temperature dependence in the logarithmic scale.

Because Cowley and Bruce's theoretical description is based on the renormalization technique, it is valid only in the critical region near T_i. The exponent of Q beyond the critical region β^* is expected to be $2\beta^* = 1$ as described by Landau theory (mean field theory).[34] In Figs. 3(a) and 3(b), it is observed that Δn deviates from the $2\beta = 0.72$–0.75 line, and asymptotically approaches the $2\beta^* = 0.99$–1.02 line. This is consistent with the expectation of the crossover behavior from the 3D XY model into Landau theory. A similar behavior of Q was observed for $SrTiO_3$ and $LaAlO_3$.[36]

Near the IC-C phase transition temperature (T_c), the plane wave approximation of Eq. (2) is not valid and high order hamonics and discommensurations appear, so that Δn is not proportional to Q^2 any more.[1,2,17] In Figs. 3(c) and 3(d), the LB behavior of AZC and TMAZC follows the $2\beta = 0.73$ line only in the region $\log_{10} t \leqslant -2$, and approaches the $2\beta^* = 0.88$–0.89 line in the region $\log_{10} t \geqslant -2$. As presented in Table I, the IC phase region of AZC and TMAZC is so narrow that T_c is close to T_i. Thus, the LB behavior cannot follow the $2\beta = 0.73$ line because Δn is not proportional to Q^2 in the region $\log_{10} t \geqslant -2$. The ambient exponent $2\beta^* = 0.88$–0.89 is thought to be not a crossover behavior of the order parameter but an effect of the high order harmonics and discommensuration near T_c.

A new phase transition of AZC was recently reported just below T_i under uniaxial stress.[37] This could be an explanation for the narrow critical region of AZC. The ambient exponent of TMAZC, $2\beta^* = 0.88$, is close to $2\beta = 0.87$, obtained by Régis *et al.*, and it might be an origin of their wrong results.[32]

For TMACC and TMAMC, the IC phase region is very narrow, and any crossover behavior was not observed as seen in Figs. 3(e) and 3(f). The IC phase region is only 5 K and even 0.6 K wide for TMACC and TMAMC, respectively. A more detailed description of the crossover phenomena is presented in Table II.

Whether the β^* of each crystal is a Landau-like one (such as RZC and KZC) or the ambient one (such as AZC and TMAZC) is thought to be dependent on the temperature range of the IC phase. The temperature range of the IC phase is mainly governed by the anisotropy of the thermodynamic potential of each crystal. In general, the stronger the anisotropy, the narrower the region of the IC phase.[2]

V. SUMMARY

The critical behavior of LB was studied near the N-IC phase transition of various A_2BX_4-type crystals. The existence of the LB tail above T_i was observed and is explained by the fluctuation effect. The values of 2β obtained from LB are distributed in the range 0.72–0.75 ($\pm$ 0.02). The 2β obtained in this study could be evidence supporting the prediction that the N-IC phase transition of A_2BX_4-type crystals belongs to the universality class of the 3D XY model. Our results also imply that LB behaves as $\Delta n \sim |t|^{2\beta}$, rather than $\Delta n \sim |t|^{\tilde{\beta}}$ or $\Delta n \sim |t|^{1-\alpha}$. The asymptotic approach to $2\beta^* = 1$ was observed beyond the critical region for RZC and KZC. It is thought to be the crossover from XY-like behavior into Landau-like behavior.

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