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## PHASE ORDERING KINETICS IN CHIRAL LIQUID CRYSTALS

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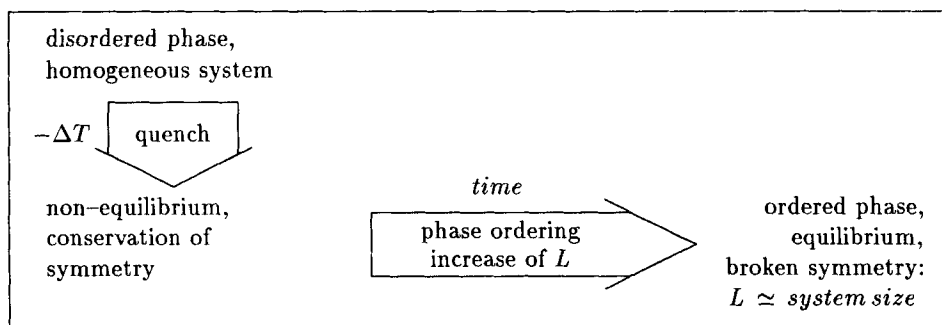
### Abstract

The phase ordering process has been investigated in the liquid crystalline blue phase II. The domain growth has been measured for the phase transitions BP<sub>II</sub>–BP<sub>III</sub> and BP<sub>II</sub>–isotropic by two different experimental methods: direct microscopic observations and visible light Bragg scattering measurements. The results for the BP<sub>II</sub>–isotropic transition with a non-conserving order parameter field are consistent with theoretical predictions. For BP<sub>II</sub>–BP<sub>III</sub> transition with a conserving order parameter field deviations from theoretical predictions are found.

### INTRODUCTION

Recently there has been a considerable interest in the phase ordering kinetics of systems with a non-scalar order parameter like liquid crystals. In this paper we present the first experimental study for blue phases (BP).

The process of phase ordering is investigated as the evolution of a large-scale structure following the quench of a system from its disordered to ordered phases. The following diagram illustrates this situation:



In our case the quench corresponds to an abrupt decrease of temperature, but it can be obtained by variation of other external parameters as well. The temperature is decreased below the interval of stability of the disordered phase by a definite supercooling depth  $\Delta T$ . After the quench we observe the relaxation of the system from non-equilibrium to an equilibrium state as a function of time. This process consists in the formation and growth of domains of the ordered phase characterized by the domain size  $L(t)$ .

To describe the early-stage of the phase separation one has to distinguish between two different types of instability of the quenched system [1]. In the first

case the system remains metastable and the formation of domains is regarded as *homogeneous nucleation*. In the second case the quenched system is unstable. The phase separation is than initiated by *spinodal decomposition*. The late-stage of the phase ordering is characterized by the *coarsening process* of domains.

The phase ordering kinetics depends on the number of components of the order parameter, the dimension of space and the type of the phase transition (phase transitions in "conserving" and "non-conserving" order parameter fields).

Theoretically, the domain growth can be described by

$$L(t) \sim t^n$$

with different values of  $n$  for different cases (cf. Table I).

Table I: Theoretical predicted values of the domain growth exponent  $n$

order parameter field	non-conserving	conserving
scalar	$n=0.5$ Allen-Cahn [2]	$n=0.33$ Lifshitz-Slyosov
multi- component	$n=0.5$	$n=0.25$ and $d(t) - d_0 = (\frac{t}{\tau})^{\frac{1}{4}}$ Coniglio-Zanetti [4]

In the case of a conserved multicomponent order parameter field the growth process is characterized by the time dependence of two lengths. In the theory of multiscaling behaviour the spatial periodicity  $d$  of the system must be taken into account additionally [4].

In this paper we describe growth processes in Blue Phase Systems of two different transitions:

- i) BP<sub>II</sub>←iso with a non-conserving order parameter field and
- ii) BP<sub>II</sub>←BP<sub>III</sub> with a conserving order parameter field because a helical structure is already developed in a short-range scale in the BP<sub>III</sub>.

The order parameter of blue phases is a tensorial quantity – the anisotropic part of the tensor of dielectric permittivity. Therefore these systems are described by multicomponent order parameter fields [5].

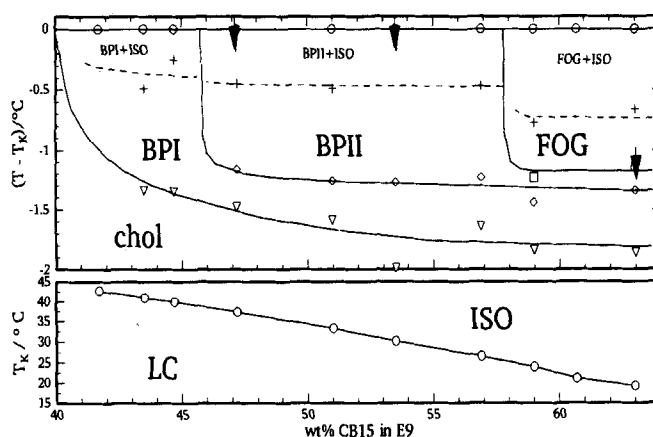


Figure 1: Phase diagram of the mixture CB15/E9

## EXPERIMENTAL

In Figure 1 the phase diagram of the mixture CB15/E9 is shown.

This liquid crystalline system exhibits the three blue phases at appropriated compositions. We were able to grow faceted single crystals of the BPI and BP11 in two-phase regions with the isotropic liquid or during the phase transition from the BP11. The investigated transitions are marked by arrows.

The growth process is examined by two different experimental methods (cf. Figure 1):

- i) Direct microscopic observations (polarizing microscope *Leitz Orthoplan*),
- ii) visible light Bragg scattering measurements (UV-VIS-spectrophotometer *Perkin Elmer Lambda 19*).

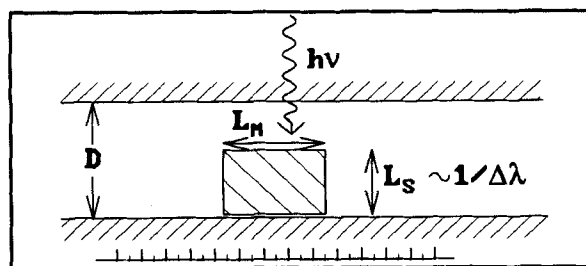


Figure 2: Principle of the measurement of the domain size  $L_M$  and  $L_S$  ( $D$ : sample thickness)

The latter method gives information about two characteristic lengths of the blue phase: The lattice constant  $d$  and in the thin samples the domain size  $L_S$  perpendicular to the substrate. From the kinematic theory of light diffraction it follows that  $L_S$  is proportional to the reciprocal half-width of the scattering curve [6]:

$$L_S \sim \frac{1}{\Delta\lambda}.$$

By microscopic observations the domain size parallel to the substrate  $L_M$  could be measured as a function of time. The growth process of the single crystals was recorded photographically. The samples were prepared in cells of  $D = 30 \mu\text{m}$  thickness. By application of a weak electric field of  $1\text{V}/\mu\text{m}$  we could obtain a definite orientation of the blue phase lattice.

## RESULTS AND DISCUSSION

### Microscopic observations

In Figure 3 the growth of BPII single crystals from the BPIII (59.0wt% CB15 in E9) is shown. The liquid single crystals are orientated with there (100) lattice planes parallel to the substrate and their habit is determined by (110) facets [7]. The function

$$L_M(t) = A(t - t_o)^n$$

is given in Figure 4 for different supercooling depths  $\Delta T$ . The exponent  $n$  of the fit curves increases with  $\Delta T$  resulting in a value of  $n > 1$  at  $\Delta T = 0.06\text{K}$ .

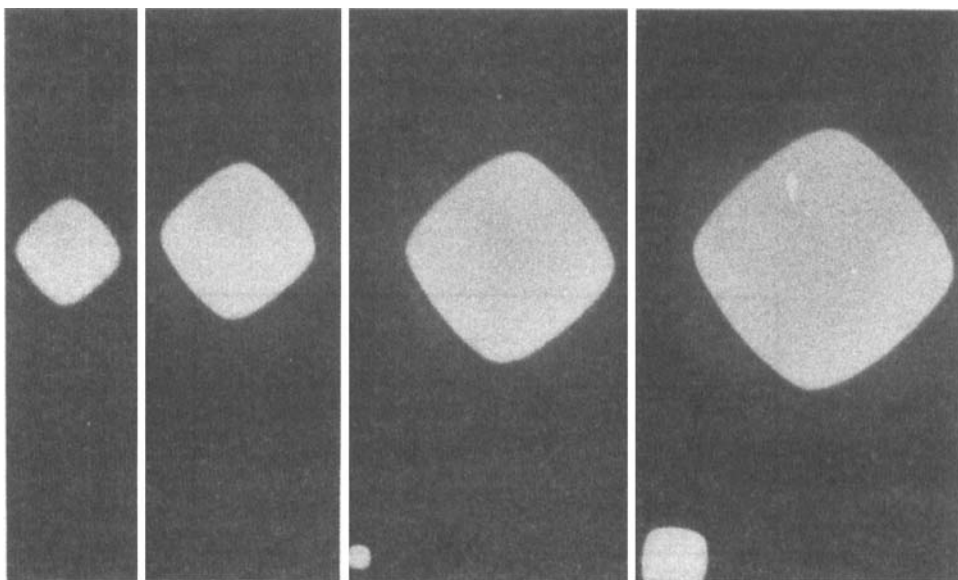


Figure 3: Domain growth process BPII←BPIII; time difference between the microphotographs:  $\Delta t = 0.5 \text{ min}$ ;  $\left| \frac{100\mu\text{m}}{\quad} \right|$ . See Color Plate XVI.

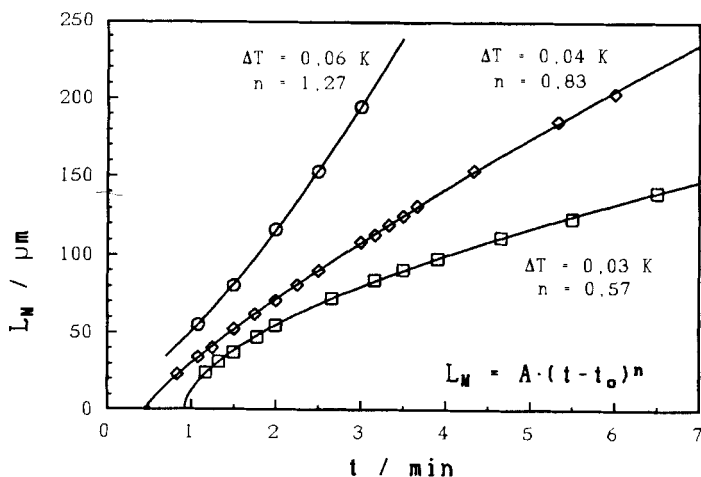


Figure 4: Time dependence of the domain size  $L_M$  for the transition BP II ← BP III.

In Figure 5 the growth of BP II single crystals in the [100] direction developing from the isotropic liquid in the two phase region BP II/iso is shown (47.2wt% CB15 in E9). The growth function could not be fitted by  $t^n$ ; obviously  $n$  decreases with  $t$ .

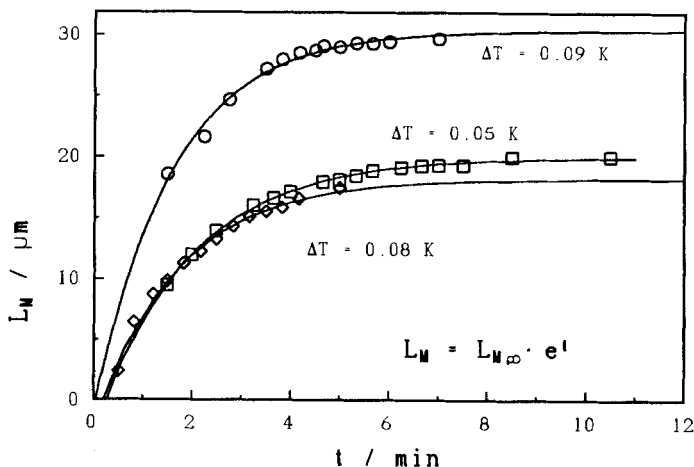


Figure 5: Time dependence of the domain size  $L_M$  for different liquid single crystals for the transition BP II ← iso.

### Bragg scattering

The growth process of BP II single crystals from the isotropic liquid in the system with 47.2wt% CB15 in E9 investigated by Bragg scattering is shown in Figure 6.

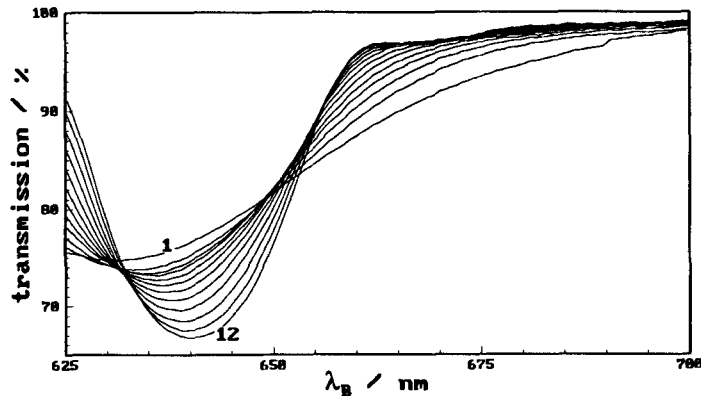


Figure 6: Transmission spectra of the Bragg reflection during the phase transition BP11←iso (time difference between two spectra: 0.35min).

The intensity of the reflexion band increases with  $t$  whereas the half width  $\Delta\lambda$  decreases as shown in Figure 7. The initial process can be described by the expression

$$L_S \sim \frac{1}{\Delta\lambda}(t) = A(t - t_o)^n$$

with an exponent  $n = 0.43...0.47$  at  $\Delta T = 0.22K$ . After about 5 min  $1/\Delta\lambda(t)$  levels out because the domain size  $L_S$  has reached the sample thickness  $D$ .

The growth process of the BP11 from the isotropic liquid in the two-phase region BP11/iso in a mixture of 53.5wt% CB15 in E9 has also been measured by Bragg scattering. The results are quite similar to that of the BP11 in the mixture of

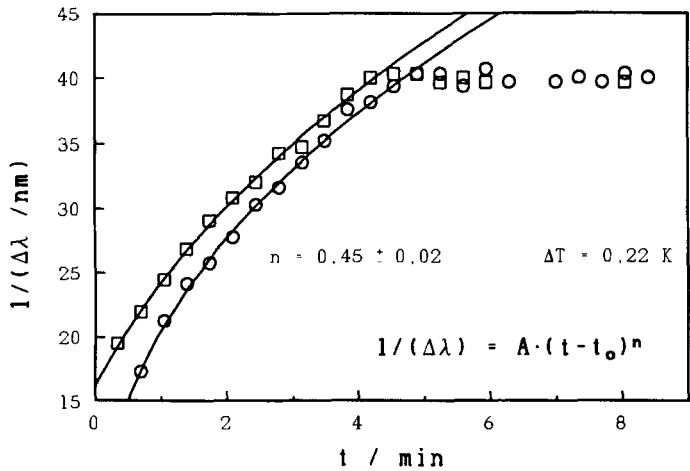


Figure 7: Time dependence of the domain size  $L_S \sim 1/\Delta\lambda$  for the transition BP11←iso.

47.2wt% CB15 in E9: We have measured exponents of  $n = 0.47 \dots 0.53$  at  $\Delta T = 0.16K$ .

## CONCLUSIONS

The phase ordering process for different types of phase transitions has been investigated in the system CB15/E9. The domain growth has been measured in the case of conserving order parameter field (BP11 $\leftarrow$ BP111) and of non-conserving conditions (BP11 $\leftarrow$ iso). The experimental data are summarized in Table II.

Table II:

order parameter field	system	wt%	$\Delta T/K$	$n$	method
conserving	BP11 $\leftarrow$ BP111	59.0	$\leq 0.06$	$\sim \Delta T$	microscopic
non conserving	BP11 $\leftarrow$ iso	47.2	$\leq 0.09$	$\sim t$	microscopic
	BP11 $\leftarrow$ iso	47.2	0.22	$\approx 0.45$	Bragg
	BP11 $\leftarrow$ iso	53.5	0.16	$\approx 0.5$	Bragg

From Bragg scattering measurements we have obtained exponents  $n$  in accordance with the theoretically predicted growth law  $L_S \sim t^n$  [4] for non-conserving order parameter fields for large values of the supercooling depth  $\Delta T$  and the initial process if  $L_S \leq D$ . For small values of  $\Delta T$ , however, the exponent  $n$  depends on time which cannot be explained by the existing theories [1].

In the case of the phase ordering in systems with conserving order parameter fields as given for the transition BP11 $\leftarrow$ BP111 the exponent  $n$  of the theoretical predicted growth law  $L_M \sim t^n$  was found to depend on the supercooling depth  $\Delta T$ . This behavior deviates from theoretical predictions [4] and deserves further experimental studies.

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