

Spatially resolved x-ray studies of liquid crystals with strongly developed bond-orientational order

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(Received 28 November 2014; revised manuscript received 19 January 2015; published 17 April 2015)

We present an x-ray study of freely suspended hexatic films of the liquid crystal 3(10)OBC. Our results reveal spatial inhomogeneities of the bond-orientational (BO) order in the vicinity of the hexatic-smectic phase transition and the formation of large-scale hexatic domains at lower temperatures. Deep in the hexatic phase up to 25 successive sixfold BO order parameters have been directly determined by means of angular x-ray cross-correlation analysis (XCCA). Such strongly developed hexatic order allowed us to determine higher order correction terms in the scaling relation predicted by the multicritical scaling theory over a full temperature range of the hexatic phase existence.

DOI: [10.1103/PhysRevE.91.042506](https://doi.org/10.1103/PhysRevE.91.042506)

PACS number(s): 64.70.mj, 61.05.C—, 61.30.Gd

I. INTRODUCTION

The influence of angular correlations on structural and physical properties of complex fluids, colloidal suspensions, and liquid crystals (LCs) remains one of fundamental and unresolved problems in modern condensed matter physics [1]. A prominent example of a system with angular correlations is the hexatic phase that combines the properties of both crystals and liquids [2]. The two-dimensional (2D) hexatic phase shows a sixfold quasi-long-range bond-orientational (BO) order, while the positional order is short range [3]. The hexatic phase is a general phenomenon that was observed in a number of systems of various physical nature, such as 2D colloids [4–6], electrons at the surface of helium [7], 2D superconducting vortices [8,9], and, particularly, in liquid crystals [10–13].

The hexatic phase was predicted by Halperin and Nelson [14] as an intermediate state in 2D crystal melting. According to their theory the hexatic phase arises as a consequence of the broken translational symmetry of a 2D crystal induced by dissociation of dislocation pairs. This mechanism does not work in three-dimensional (3D) crystals; however, the 3D hexatic phase was observed experimentally in LCs [10]. The multicritical scaling theory (MCST) developed by Aharony and coworkers [15] based on renormalization group approach to critical phenomena enabled quantitative characterization of the BO order in the hexatic phase. It allowed one to study a crossover from 2D to 3D behavior [13,16]. In spite of the extensive experimental and theoretical work the origin of the hexatic phase in LCs and the features of the hexatic-smectic phase transition remain puzzling and controversial.

The structure of hexatics is traditionally studied by means of x-ray or electron diffraction in a single-domain area of a hexatic film (see for reviews Refs. [17–19]). The quantitative

characteristics of the BO order, the so-called BO order parameters [11], are typically determined by fitting the measured azimuthal intensity distribution by the Fourier cosine series. In contrast to this approach in the present work we performed spatially resolved x-ray diffraction studies of free-standing LC films. Measured x-ray data were analyzed by means of direct Fourier transformation and by using angular x-ray cross-correlation analysis (XCCA) [20–23]. The latter method enabled a direct determination of sixfold BO order parameters from the ensemble of diffraction patterns without applying a fitting procedure [24].

II. EXPERIMENT

The coherent x-ray scattering experiment on smectic LC membranes was performed at the beamline P10 of the PETRA III facility at DESY in Hamburg. The incident photon energy was 13 keV with the flux 3×10^{10} photons/sec. The geometry of the experiment is schematically shown in Fig. 1. Pilatus 1M detector (981×1043 pixels of $172 \times 172 \mu\text{m}^2$ size) was positioned in transmission geometry at the distance of 263 mm from the sample. The beam at the sample plane was focused to $2 \times 3 \mu\text{m}^2$ (vertical vs horizontal) at full width at half maximum (FWHM) by a set of compound refractive lenses (CRLs) [25]. A specially designed sample stage FS1 and a mK1000 temperature controller from INSTEC was used to prepare LC films *in situ* and maintain the temperature with an accuracy of 0.005 °C.

In our experiment we have used the LC compound 3(10)OBC (*n*-propyl-4'-*n*-decyloxybiphenyl-4-carboxylate) [12,26]. The hexatic-smectic phase transition was found at $T \approx 66.3 \pm 0.1$ °C, and the material crystallizes below $T \approx 54$ °C. The films of 3(10)OBC sample were drawn across a small circular glass aperture of 2 mm in diameter inside the chamber at 10 °C above the temperature of the hexatic-smectic phase transition. The thickness of the films was determined to be in the range of 5–7 μm with the AVANTES fiber optical spectrometer. Films were slowly cooled to observe the

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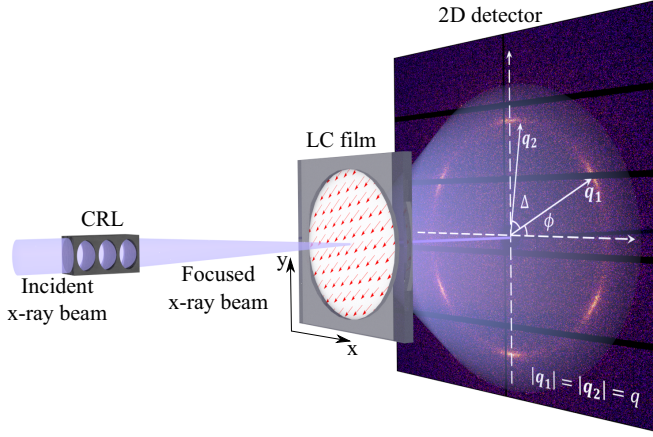


FIG. 1. (Color online) The scheme of the diffraction experiment showing the focusing system, glass sample stage with drawn LC membrane, and 2D detector.

formation and development of the hexatic phase. For spatially resolved studies at each temperature the sample was scanned in the plane perpendicular to the incident beam direction in the region of $100 \times 100 \mu\text{m}^2$ with $11 \mu\text{m}$ step size. Exposure time of 0.6 sec was chosen to sustain the nondestructive regime of measurements.

Typical diffraction patterns in the smectic and hexatic phases corrected for background scattering and horizontal synchrotron polarization are shown in Figs. 2(a)–2(c). At high temperatures in the smectic phase the diffraction pattern has a form of a broad uniform ring [Fig. 2(a)] due to the absence of angular correlations and short-range positional order. During

the hexatic-smectic phase transition this scattering ring splits into six arcs [Fig. 2(b)], revealing the sixfold rotational symmetry of the in-plane molecular arrangement. While the temperature decreases all six arcs become narrower in both azimuthal and radial directions, indicating the simultaneous development of the BO and positional order [Fig. 2(c)].

III. RESULTS AND DISCUSSION

The in-plane structure of smectic films is characterized by the positional correlation length ξ , which is commonly defined as $\xi = 1/\Delta q$, where Δq is the half width at half maximum (HWHM) of a radial scan through the center of a diffraction peak [17]. Typically in the systems with short-range positional order the shape of the diffraction peak can be described by Lorentzian function. However, according to the phenomenological theory of Aeppli and Bruinsma [27] the radial scan through the scattering peak maximum position should have a form of a square root Lorentzian (SRL) function in the vicinity of the hexatic-smectic phase transition. Our studies revealed that for our system the square root Lorentzian function described the experimental data better only within the small temperature region $65.5^\circ\text{C} \leq T \leq 66.1^\circ\text{C}$. Out of this range the experimental data were better fitted with the Lorentzian function. We would like to point out that the values of the positional correlation length ξ obtained by the fitting with these two functions do not differ significantly close to the phase transition point.

The value of ξ determined by fitting the radial intensity profiles with the Lorentzian function at each position in the sample and averaged over a set of $N = 100$ diffraction patterns at each temperature is shown in Fig. 2(d). In the smectic phase the positional correlation length ξ was determined to be about 1.5 nm. Close to the hexatic phase transition point at $T = 66.3^\circ\text{C}$ the correlation length starts to increase, and upon further cooling it reaches the value of 24 nm in the hexatic phase. The simultaneous rise of the peak's maximum position q_0 and the presence of the inflection point on the q_0 temperature dependence [see inset in Fig. 2(d)] indicates the growing fluctuations of the BO order parameter in the vicinity of the hexatic-smectic phase transition [27,28].

To determine the details of the BO order formation and especially its spatial distribution in the vicinity of the hexatic-smectic phase transition region we performed an expansion of the scattered intensity measured at each position in the sample into a Fourier cosine series,

$$I(q, \phi) = I_0(q) + 2 \sum_{n=1}^{n=+\infty} |I_n(q)| \cos[n\phi + \psi_n(q)], \quad (1)$$

where

$$|I_n(q)| \exp[i\psi_n(q)] = \frac{1}{2\pi} \int_0^{2\pi} I(q, \phi) \exp(-in\phi) d\phi, \quad (2)$$

In this Fourier decomposition (q, ϕ) are the polar coordinates in the detector plane, $I_0(q)$ is the scattered intensity averaged over a scattering ring of a radius q , and $|I_n(q)|$ and $\psi_n(q)$ are the magnitude and phase of the n th Fourier component (FC).

The in-plane orientation of the molecular bonds was determined at each spatial position using the phase $\psi_6(q_0)$

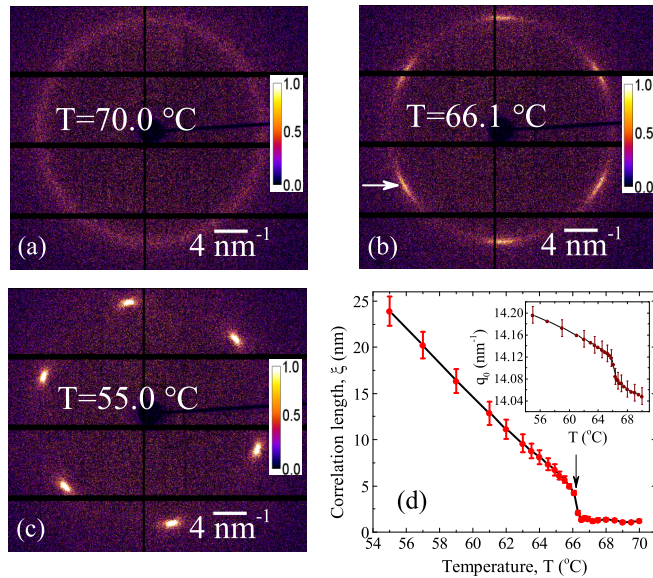


FIG. 2. (Color online) (a–c) Diffraction patterns from a LC film in the smectic (a) and hexatic phase (b, c). (d) Temperature dependence of the positional correlation length ξ determined from the radial profile of the scattering peak marked by an arrow in (b). The temperature of the hexatic-smectic phase transition is specified by an arrow. In the inset the temperature dependence of the peak maximum position q_0 is shown.

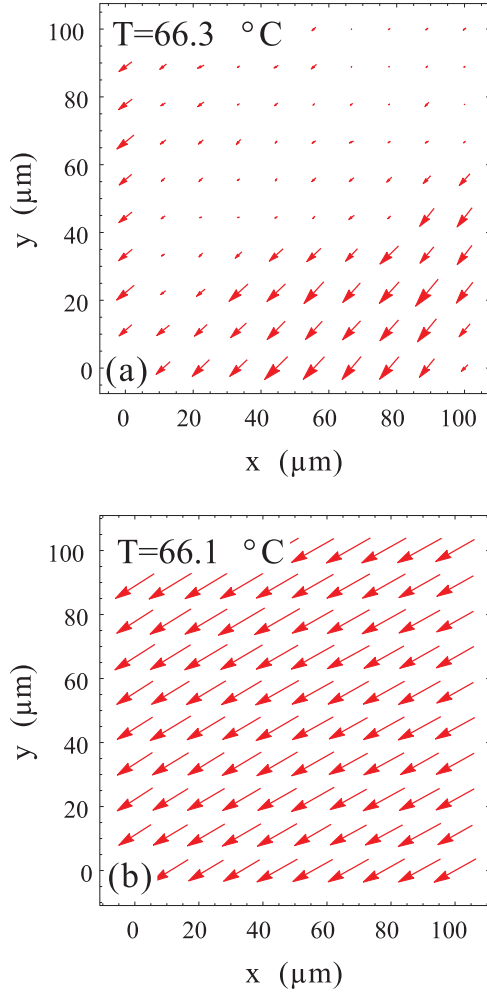


FIG. 3. (Color online) Spatially resolved 2D maps of the LC film structure at $T = 66.3^\circ\text{C}$ (a) and $T = 66.1^\circ\text{C}$ (b). Each vector in the plot corresponds to a certain position in the sample. Direction of a vector is associated with the angular position of the center of the peak marked with an arrow in Fig. 2(b), and the length of a vector is proportional to the magnitude $|I_6(q_0)|$. The length of the vectors in (a) is magnified five times as compared to the vectors in (b).

and magnitude $|I_6(q_0)|$ of the FC of the dominant sixth order, calculated at the scattering peak maximum position $q = q_0$ (see for details Appendix A). In the smectic phase $|I_6(q_0)|$ was found to be below the noise threshold due to the absence of angular correlations. As soon as the temperature was decreased to the phase transition region the situation dramatically changed [see Figs. 3(a) and 3(b)]. Remarkably, the 2D map shown in Fig. 3(a) reveals inhomogeneity in the spatial distribution of the hexatic phase at the temperature $T = 66.3^\circ\text{C}$. This is clearly reflected in variation of the magnitude $|I_6(q_0)|$ across the sample. Areas with larger magnitudes of the vectors correspond to the regions with higher degree of the BO order. While cooling the sample to the temperature $T = 66.1^\circ\text{C}$ a single domain with identical orientation of molecular bonds over the entire region of scanning was formed [Fig. 3(b)]. Such uniform BO order with the same orientation of molecular bonds persists upon cooling into the crystalline phase.

Next, we applied XCCA [21] to determine variation of the BO order parameters in the hexatic phase as a function of temperature. Such approach is not sensitive to small spatial variations of the molecular bonds' orientation and gives more reliable information on BO order parameters than fitting of azimuthal intensity distribution. The basic element of this approach is the two-point cross-correlation function (CCF) [20,21,29]

$$G(q, \Delta) = \langle I(q, \phi) I(q, \phi + \Delta) \rangle_\phi, \quad (3)$$

where $\langle \dots \rangle_\phi$ is the angular average over a ring of radius q and $0 \leq \Delta < 2\pi$ is the angular coordinate. Since $G(q, \Delta)$ is a real even function, it can be decomposed into the Fourier cosine series,

$$G(q, \Delta) = G_0(q, \Delta) + 2 \sum_{n=1}^{n=+\infty} G_n(q) \cos(n\Delta), \quad (4)$$

where $G_n(q)$ are the FCs of the CCF. In the present case of a single domain the FCs $G_n(q)$ averaged over a set of N diffraction patterns are related to the FCs of intensity $I_n(q)$ by a simple relation $\langle G_n(q) \rangle_N = |I_n(q)|^2$ [23].

The FCs of intensity $|I_n(q)| = \sqrt{\langle G_n(q) \rangle_N}$, where $N = 100$ in our case, as a function of q measured at the temperature $T = 61.0^\circ\text{C}$ are presented in Fig. 4(a). As one can see, we determined an unprecedented number of harmonics in the hexatic phase of the 3(10)OBC film. To characterize the BO order parameters we used the maximum values of the magnitudes of the FCs $|I_n(q_0)|$ ($q_0 = 14.16 \text{ nm}^{-1}$) that are shown in the inset of Fig. 4(a). One can clearly see that the magnitudes of the FCs systematically decrease as a function of the harmonic number n (see discussion below), in agreement with MCST [15] (see red line). In the ideal hexatic phase only FCs $|I_n(q_0)|$ of the orders $n = 6m$, where $m = 1, 2, 3, \dots$, contribute to the Fourier decomposition [11]. A finite nonzero contribution of other FCs appear due to small background contribution and other uncompensated experimental factors. A threshold was defined as the average value of these components, and only FCs with the magnitudes above this threshold were considered in the further analysis. This averaged background was subtracted from the values of FCs before further analysis.

In the previous studies of the BO order in the hexatic phase of LCs the q dependence of the FCs $I_n(q)$ was not considered. Presently none of the theories describing the hexatic ordering give predictions for the general expression of the harmonics $I_n(q)$. This question will require further theoretical consideration. Analysis of our data has shown that it is not possible to characterize the FCs of intensity as a function of q presented in Fig. 4(a) by single functional form. In our experiment the FCs of lower orders ($n = 6, 12, 18$) were better described by the SRL function. For larger values of n the tails of the curves become steeper and harmonics $I_n(q)$ are better fitted by the Lorentzian functions [see Fig. 4(a)]. The temperature dependence of the HWHM γ_n of the few first FCs is presented in Fig. 4(b). We can observe a gradual decrease of these values with the harmonic number n and upon cooling [24].

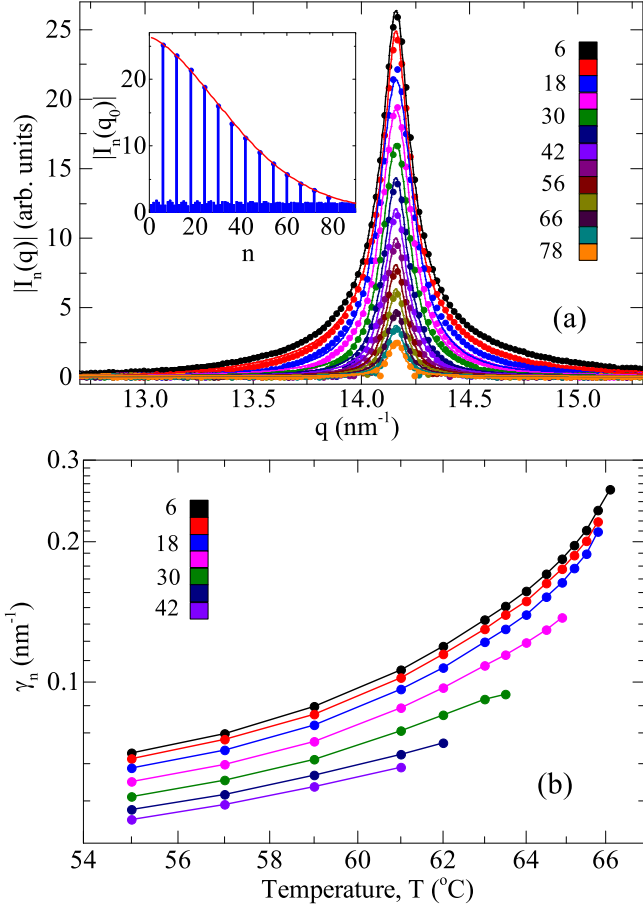


FIG. 4. (Color) (a) Magnitudes of the FCs of intensity $|I_n(q)|$ with $n = 6, 12, \dots, 78$ as a function of q at the temperature $T = 61.0^\circ\text{C}$. Solid lines are SRL and Lorentzian fits to the experimental data (points) (see text for details). (inset) Magnitudes of the FCs $|I_n(q_0)|$ at $q_0 = 14.16 \text{ nm}^{-1}$ as a function of the order n . Red line represents the fitting using the MCST. (b) Temperature dependence of the HWHM γ_n of the FCs of intensity.

The evolution of the BO order as a function of temperature can be described by the set of independent normalized BO order parameters, $C_{6m} = |I_{6m}(q_0)|/|I_0(q_0)|$ defined in Ref. [11]. The number of nonzero coefficients C_{6m} and their magnitude characterize a degree of the BO order development. The temperature dependence of the BO order parameters C_{6m} is presented in Fig. 5(a). In the smectic phase all parameters C_{6m} have values below the threshold level. According to our results only the FCs of the 6th and 12th order appear at the phase transition temperature $T = 66.3^\circ\text{C}$, and further components exceed the threshold level one after another while cooling below the phase transition temperature [see Fig. 5(a)]. The possible explanation of this behavior could be nonlinear interactions between parameters C_{6m} of different order. Finally, we observed an unusually large number $m = 25$ of the successive BO order parameters at the temperature $T = 55.0^\circ\text{C}$. Interestingly, we also observed a subtle nonmonotonic behavior of the FCs of the orders $m = 4, \dots, 10$ in the temperature range $63^\circ\text{C} < T < 66^\circ\text{C}$ [see inset in Fig. 5(a)]. The temperature dependence of the C_{6m} parameters confirms a general trend described earlier for 2D [13] and

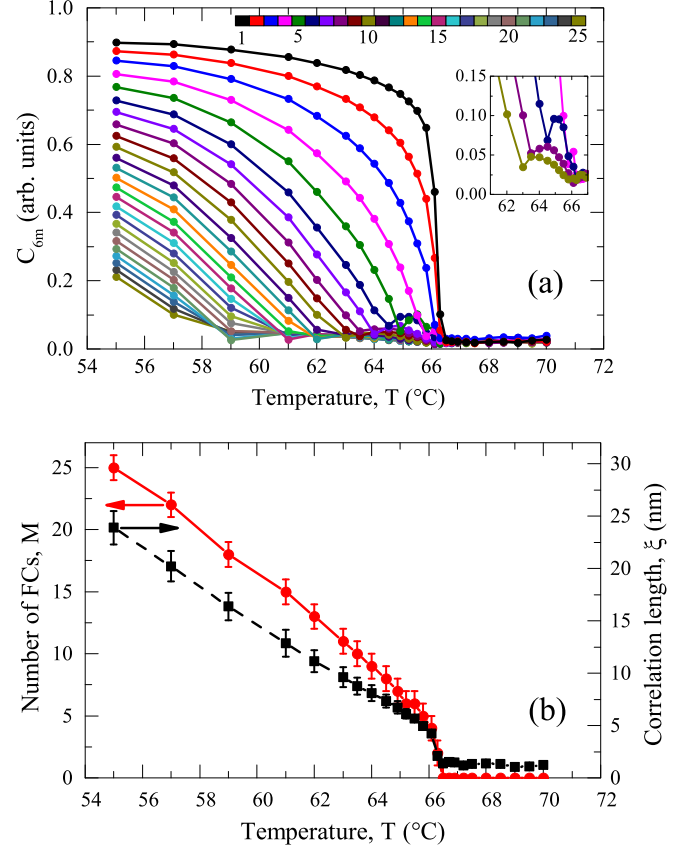


FIG. 5. (Color) (a) Temperature dependence of the normalized BO order parameters C_{6m} . In the inset the nonmonotonic behavior of the BO order parameters of the orders $m = 4, \dots, 10$ is shown. (b) Temperature dependence of the total number of FCs M and positional correlation length ξ [see Fig. 2(d)].

3D [11,15,19] hexatics. However, the maximum number of the registered BO order parameters in these experiments was significantly smaller, typically less than 10. The total number of the determined FCs, M , as a function of temperature is shown in Fig 5(b). The correlation between this number and the positional correlation length ξ directly indicates strong coupling between the BO and positional order in the hexatic phase of 3(10)OBC film.

A quantitative comparison of the BO parameters of different orders can be made on the basis of the MCST [15]. This theory predicts the following scaling relation for the BO order parameters,

$$C_{6m} = (C_6)^{\sigma_m}, \quad (5)$$

with the exponent σ_m of the form

$$\sigma_m = m + x_m \cdot m \cdot (m - 1), \quad x_m^{(1)} = \lambda(T) - \mu(T)m. \quad (6)$$

Here $x_m^{(1)}$ is the expansion of parameter x_m up to the first order correction in number m . The values of the parameters $\lambda(T) \cong 0.3$ and $\mu(T) \cong 8 \times 10^{-3}$ are given by the MCST for the 3D hexatic phase [18,19]. At each temperature the experimentally determined BO order parameters C_{6m} were fitted by the scaling relation (5,6) [an example of such fit for the temperature $T = 61.0^\circ\text{C}$ is shown in the inset in Fig. 4(a)]. The temperature dependence of the determined

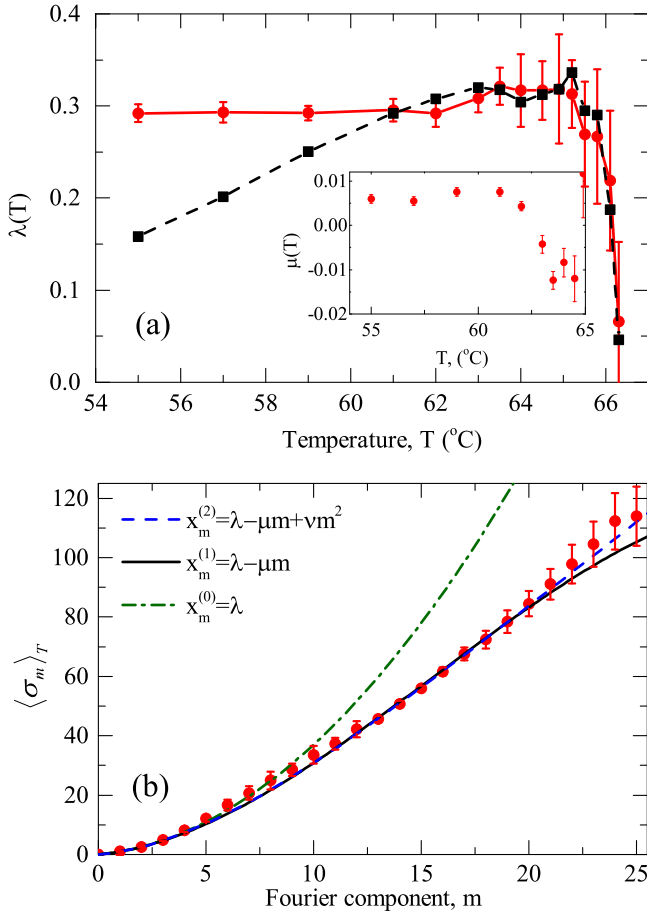


FIG. 6. (Color online) (a) Temperature dependence of the parameter $\lambda(T)$ (red circles) in the scaling relation (6) from the MCST. Compare with the temperature dependence of the same parameter (black squares) when the linear correction term in the scaling relation (6) is neglected. (b) Temperature-averaged values of $\langle\sigma_m\rangle_T$ (red circles) and their fit with the scaling relation (6) with the first order correction term (black line). Results of fitting without the correction terms (green dash-dot line) and with the 2nd order correction term (blue dash line) are also shown.

parameters $\lambda(T)$ and $\mu(T)$ is presented in Fig. 6(a) by red circles (see for details Appendix B). As we can see, the scaling relation (5) accurately describes our experimental data in the entire temperature range of the hexatic phase existence. At low temperatures the parameters of the scaling law reach practically the constant values, $\lambda(T) \approx 0.29 \pm 0.01$ and $\mu(T) \approx 0.007 \pm 0.001$, that are in excellent agreement with the theoretical predictions of the MCST for 3D hexatics [15]. In the previous experiments [11,15] only the parameter λ has been determined. Here, due to the presence of the large number of harmonics, we were able to deduce also the value of the first order correction term μ . Our results also show that if the first order correction term in $x_m^{(1)}$ is neglected [shown by black squares in Fig. 6(a)], then we get a strong deviation from the theoretically predicted value $\lambda(T) = 0.3$ at low temperatures, where many FCs are present. We also found that in the vicinity of the hexatic-smectic phase transition the parameters $\lambda(T)$ and $\mu(T)$ rapidly decrease in a

narrow temperature range of $\Delta T \approx 1^\circ\text{C}$ [Fig. 6(a)] (a similar observation was made for the parameter $\lambda(T)$ in Ref. [15]).

Since the exponents σ_m in the scaling relation (5) appear to be almost independent of temperature, one can consider the temperature averaged values $\langle\sigma_m\rangle_T$ to determine the parameters λ and μ by applying Eqs. (6) [15]. The experimentally obtained values of $\langle\sigma_m\rangle_T$ are shown in Fig. 6(b). We clearly see that the term $x_m^{(1)}$ with $\lambda = 0.31 \pm 0.015$ and $\mu = 0.009 \pm 0.001$ determined by fitting $\langle\sigma_m\rangle_T$ data points with Eqs. (6) describes very well all orders m measured in the experiment [30]. At the same time if the first order correction term in $x_m^{(1)}$ is neglected, then the scaling relation (6) correctly describes the behavior of the parameter $\langle\sigma_m\rangle_T$ only up to the 7th order but fails to characterize accurately higher orders m [see Fig. 6(b)]. We also observed that at high values of $m \geq 20$ there is a small but constant deviation of the measured and fitted values. Statistical analysis based on the F-test [31] revealed that the scaling relation term in the form $x_m^{(2)} = \lambda - \mu m + \nu m^2$ is statistically significant with the significance level of 0.02. The fitting of the experimentally determined values of $\langle\sigma_m\rangle_T$ with this scaling relation [shown by dash blue line in Fig. 6(b)] gave for the parameter ν a small value of $\nu \approx 1 \times 10^{-4}$. Determining this second order correction value theoretically using renormalization theory would require one to perform ϵ expansion of the order higher than ϵ^2 ($\epsilon = 4 - d$, where d is the space dimension) [15].

IV. CONCLUSION

In summary, we investigated the in-plane structure of free-standing hexatic films of 3(10)OBC LC by means of x-ray diffraction technique. A micron-sized spatially resolved measurements allowed us to construct 2D maps of the in-plane molecular bonds orientations that revealed inhomogeneity of the BO order at the hexatic-smectic phase transition region. An exceptional high number ($m = 25$) of the successive BO order parameters have been directly determined at low temperatures in the hexatic phase by means of XCCA. Our results support the validity of the MCST in the whole temperature range of existence of the 3D hexatic phase. We have confirmed experimentally the value of the first order correction term in the scaling relation of the BO order parameters that was predicted earlier by the MCST. Moreover, we also revealed a small second order correction term that was not yet determined theoretically.

Many questions on the hexatic phase are still to be answered. Among them are the spatial distribution of the BO order parameters close to defects and domain boundaries, which can be studied by an extremely focused x-ray beam; the high precision measurements of the BO order parameters' temperature dependence in the vicinity of the phase transition point; the time-resolved measurement of the BO order fluctuations; and many others. We expect that our work will stimulate both theoretical studies and development of experimental techniques for quantitative investigation of the effects of large-scale angular correlations on the structure and physical properties of diverse LC phases and complex fluids.

ACKNOWLEDGMENTS

We acknowledge support of this project and discussions with E. Weckert. We are thankful to C. C. Huang, W. H. de Jeu, E. I. Kats, V. V. Lebedev, A. R. Muratov, and V. M. Kaganer for fruitful discussions and R. Gehrke for careful reading of the paper. This work was partially supported by the Virtual Institute VH-VI-403 of the Helmholtz Association. The work of I.A.Z. and B.I.O. was partially supported by the Russian Science Foundation (Grant No. 14-12-00475).

APPENDIX A: DETERMINATION OF A HEXATIC DOMAIN ORIENTATION

The scattered intensity of each diffraction pattern (Fig. 7) was transformed from Cartesian (x, y) to polar (q, ϕ) coordinate system using bilinear interpolation. The obtained intensities $I(q, \phi)$ were decomposed into cosine Fourier series according to Eqs. (1)–(2). To determinate the intermolecular bond orientations in the hexatic phase we considered the dominant FC of the order $n = 6$, calculated at the value $q = q_0$. Since the term $|I_6(q)| \cos[6\phi + \psi_6(q)]$ in Eqs. (1)–(2) is a periodic function of an angle ϕ with a period of $2\pi/6$, the values of the phase $\psi_6(q_0)$ for each diffraction pattern can be chosen uniquely to be within the range $\psi_6(q_0) \in [\psi_6(q_0) - \pi/6, \psi_6(q_0) + \pi/6]$. The value $\psi_6(q_0)$ was chosen in such way, that the angular position $\psi_6(q_0)$ of the diffraction peak in the hexatic phase [specified by an arrow in Fig. 2(b) in the main text] was always within the forgoing range.

APPENDIX B: FITTING WITH THE MULTISCALING THEORY

In order to compensate the contribution of the noise in $I_0(q_0)$ and consequently in $C_{6m} = |(I_{6m}(q_0))/(I_0(q_0))|$ we introduced the correction coefficient $S(T)$ into the scaling relation:

$$S(T)C_{6m} = (S(T)C_6)^{\sigma_m}. \quad (\text{B1})$$

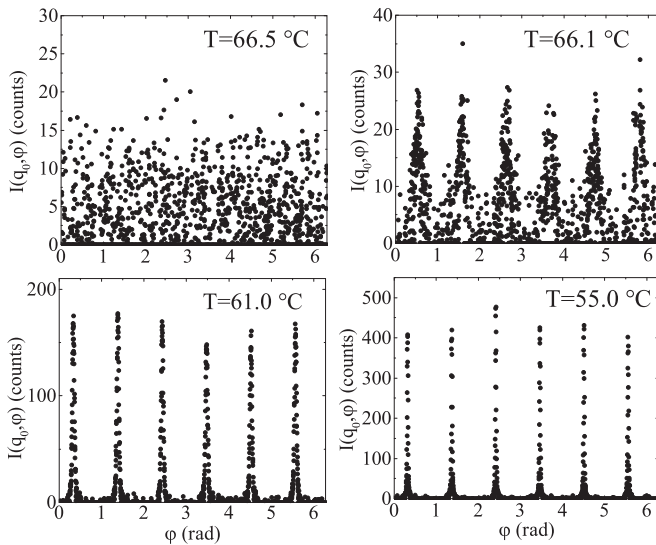


FIG. 7. The scattered intensity $I(q_0, \phi)$ as a function of azimuthal angle at the temperatures $T = 66.5^\circ\text{C}$, $T = 66.1^\circ\text{C}$, $T = 61.0^\circ\text{C}$, and $T = 55.0^\circ\text{C}$.

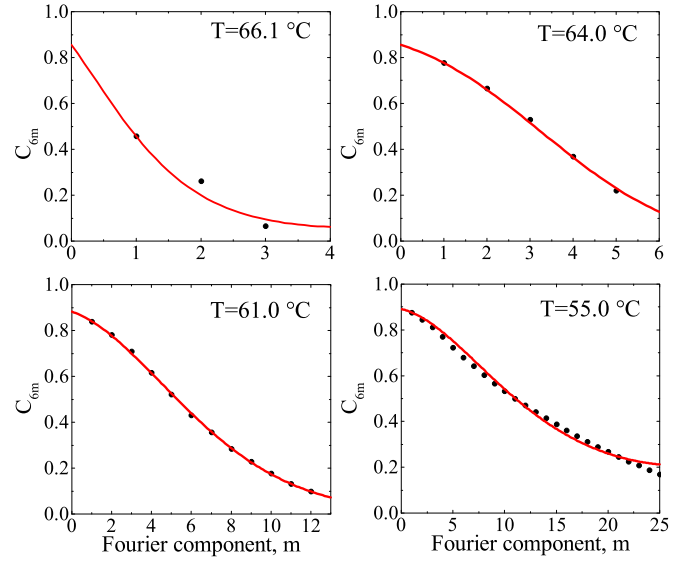


FIG. 8. (Color online) The fitting of C_{6m} with function (B2) at the temperatures $T = 66.1^\circ\text{C}$, $T = 64.0^\circ\text{C}$, $T = 61.0^\circ\text{C}$, and $T = 55.0^\circ\text{C}$.

At each temperature point the experimentally obtained values C_{6m} ($m = 1, 2, 3, \dots$) were fitted with the following function:

$$C_{6m} = \frac{1}{S(T)} [S(T)C_6]^{\sigma_m} \quad (\text{B2})$$

with three free parameters $S(T)$, $\lambda(T)$, and $\mu(T)$. The examples of the fitting for different temperatures are shown in Fig. 8. The value of $S(T)$ slightly decreases upon cooling from 1.15 at $T = 66.1^\circ\text{C}$ to 1.13 at $T = 55.0^\circ\text{C}$. When the values of the correction coefficient $S(T)$ are known at each temperature point one can directly calculate values of $\sigma_m(T)$. The exponents $\sigma_m(T)$ are shown in Fig. 9 for all temperatures. One can clearly see that the values of $\sigma_m(T)$ are practically independent of temperature for lower harmonics and change within the error for higher harmonics. This allows us to calculate the temperature-averaged values $\langle \sigma_m \rangle_T$.

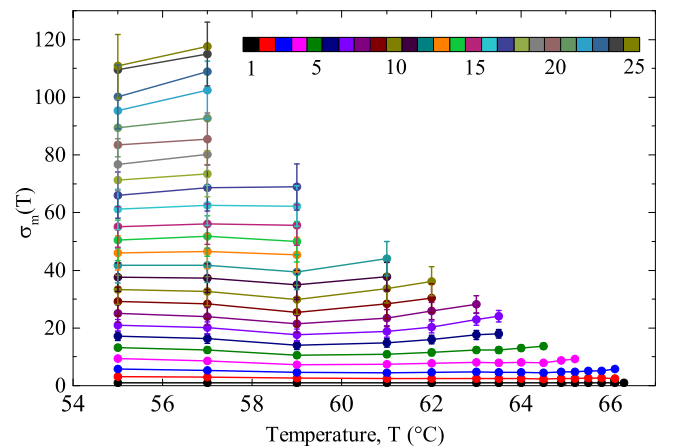


FIG. 9. (Color) Temperature dependence of the exponents σ_m for all harmonics.

- [1] P. Chaikin and T. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, 1995).
- [2] K. J. Strandburg, in *Bond-Orintational Order in Condensed Matter Systems*, edited by K. J. Strandburg (Springer-Verlag, New York, 1992).
- [3] D. R. Nelson, *Defects and Geometry in Condensed Matter Physics* (Cambridge University Press, Cambridge, 2002).
- [4] C. A. Murray and D. H. Van Winkle, *Phys. Rev. Lett.* **58**, 1200 (1987).
- [5] R. E. Kusner, J. A. Mann, J. Kerins, and A. J. Dahm, *Phys. Rev. Lett.* **73**, 3113 (1994).
- [6] P. Keim, G. Maret, and H. H. von Grünberg, *Phys. Rev. E* **75**, 031402 (2007).
- [7] D. C. Glatli, E. Y. Andrei, and F. I. B. Williams, *Phys. Rev. Lett.* **60**, 420 (1988).
- [8] C. A. Murray, P. L. Gammel, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, *Phys. Rev. Lett.* **64**, 2312 (1990).
- [9] I. Guillaumon, H. Suderow, A. Fernandez-Pacheco, J. Sese, R. Cordoba, J. M. De Teresa, M. R. Ibarra, and S. Vieira, *Nat. Phys.* **5**, 651 (2009).
- [10] R. Pindak, D. E. Moncton, S. C. Davey, and J. W. Goodby, *Phys. Rev. Lett.* **46**, 1135 (1981).
- [11] J. D. Brock, A. Aharony, R. J. Birgeneau, K. W. Evans-Lutterodt, J. D. Litster, P. M. Horn, G. B. Stephenson, and A. R. Tajbakhsh, *Phys. Rev. Lett.* **57**, 98 (1986).
- [12] T. Stoebe, A. J. Jin, P. Mach, and C. Huang, *Int. J. Thermophys.* **16**, 33 (1995).
- [13] C.-F. Chou, J. T. Ho, and S. W. Hui, *Phys. Rev. E* **56**, 592 (1997).
- [14] B. I. Halperin and D. R. Nelson, *Phys. Rev. Lett.* **41**, 121 (1978).
- [15] A. Aharony, R. J. Birgeneau, J. D. Brock, and J. D. Litster, *Phys. Rev. Lett.* **57**, 1012 (1986).
- [16] T. Stoebe, R. Geer, C. C. Huang, and J. W. Goodby, *Phys. Rev. Lett.* **69**, 2090 (1992).
- [17] W. H. de Jeu, B. I. Ostrovskii, and A. N. Shalaginov, *Rev. Mod. Phys.* **75**, 181 (2003).
- [18] J. Brock, D. Noh, B. McClain, J. Litster, R. Birgeneau, A. Aharony, P. Horn, and J. Liang, *Z. Phys. B* **74**, 197 (1989).
- [19] J. D. Brock, R. J. Birgeneau, D. Litster, and A. Aharony, *Contemp. Phys.* **30**, 321 (1989).
- [20] P. Wochner, C. Gutt, T. Autenrieth, T. Demmer, V. Bugaev, A. D. Ortiz, A. Duri, F. Zontone, G. Grübel, and H. Dosch, *Proc. Natl. Acad. Sci. USA* **106**, 11511 (2009).
- [21] M. Altarelli, R. P. Kurta, and I. A. Vartanyants, *Phys. Rev. B* **82**, 104207 (2010).
- [22] R. P. Kurta, M. Altarelli, E. Weckert, and I. A. Vartanyants, *Phys. Rev. B* **85**, 184204 (2012).
- [23] R. P. Kurta, M. Altarelli, and I. A. Vartanyants, *Adv. Condens. Matter Phys.* **2013**, 959835 (2013).
- [24] R. P. Kurta, B. I. Ostrovskii, A. Singer, O. Y. Gorobtsov, A. Shabalin, D. Dzhigaev, O. M. Yefanov, A. V. Zozulya, M. Sprung, and I. A. Vartanyants, *Phys. Rev. E* **88**, 044501 (2013).
- [25] A. V. Zozulya, S. Bondarenko, A. Schavkan, F. Westermeier, G. Grübel, and M. Sprung, *Opt. Express* **20**, 18967 (2012).
- [26] C. C. Huang, G. Nounesis, R. Geer, J. W. Goodby, and D. Guillon, *Phys. Rev. A* **39**, 3741 (1989).
- [27] G. Aeppli and R. Bruinsma, *Phys. Rev. Lett.* **53**, 2133 (1984).
- [28] S. C. Davey, J. Budai, J. W. Goodby, R. Pindak, and D. E. Moncton, *Phys. Rev. Lett.* **53**, 2129 (1984).
- [29] N. A. Clark, B. J. Ackerson, and A. J. Hurd, *Phys. Rev. Lett.* **50**, 1459 (1983).
- [30] We note that the values of λ and μ are similar to the values determined in Fig. 6(a).
- [31] P. R. Bevington and R. D. Keith, *Data Reduction and Error Analysis for the Physical Sciences* (McGraw-Hill, New York, 2003).