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Supplementary information for

Angular structure factor of the hexatic-B liquid crystals: bridging theory and experiment

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1 Coupling between the bond-orientational order and positional correlations in the hexatic phase

Below we reproduce the arguments presented in the paper by G. Aeppli and R. Bruinsma[1] in order to show that the coupling between the density fluctuations and the bond-orientational (BO) order leads to the angular profile of the scattering peaks in the Hex-B phase described by the Voigt function (see also[2]).
At the temperatures below the Sm-A – Hex-B phase transition, the positional correlations between the molecules in the Hex-B layers decay over a short distance $\xi \sim q_0^{-1}$, while the BO order persists over much larger distances within single hexatic domains, $\Lambda_0^{-1} \gg q_0^{-1}$. Here q_0 is a reciprocal vector corresponding to the average distance between the moleccules. This allows us to divide the Hex-B into the large cells located at point $\boldsymbol{r}$ with a typical size of Λ_0^{-1}, in which the two-component BO order parameter $\Psi(\boldsymbol{r}) = |\Psi(\boldsymbol{r})|e^{i6\psi(\boldsymbol{r})}$ has approximately constant amplitude $|\Psi(\boldsymbol{r})|$ and phase $\psi(\boldsymbol{r})$[1]. We will also assume that the fluctuations of the amplitude $|\Psi|$ of the BO order parameter are relatively small and within the mean-field approximation it can replaced by its root-mean-square value $\langle|\Psi|^2\rangle^{1/2}$. This means that only the phase $\psi(\boldsymbol{r})$ fluctuates between the cells.
The free-energy functional of the Hex-B phase has three contributions, describing the positional order, the bond-orientational order and the coupling between them:

$$F = F_{\Delta\rho}[\Delta\rho] + F_{\Psi}[\Psi] + F_{\Delta\rho-\Psi}[\Delta\rho, \Psi]. \tag{S1}$$

Let us consider only the in-plane fluctuations of the density, and assume that the molecular layers are perfectly flat. In this case, the first term describing the short-range positional order within a molecular layer can be represented as an integral over the Fourier components of the in-plane density fluctuations $\delta\rho(\boldsymbol{q}_\perp)$

$$F_{\delta\rho}[\delta\rho] = \int d\boldsymbol{q}_\perp \{a + b(q_\perp - q_{\perp 0})^2\}(\delta\rho(\boldsymbol{q}_\perp))^2. \tag{S2}$$

where the integration is performed in the vicinity of the in-plane peak $q_\perp \sim q_{\perp 0}$[3]. Here we used polar coordinates for the scattering vector $\boldsymbol{q} = (q_z, \boldsymbol{q}_\perp) = (q_z, q_\perp, \varphi)$. Coefficients a and b are the coefficients of Landau expansion of the free energy.

Within an individual cell, the two-component BO order parameter $\Psi(\boldsymbol{r}) = |\Psi(\boldsymbol{r})| e^{i6\psi(\boldsymbol{r})}$ is approximately a constant, so the contribution from the second term $F_\Psi[\Psi]$ in eqn (S1) is zero. We will take the fluctuations of the phase $\psi(\boldsymbol{r})$ between the cells into account later (see eqn (S7)).

The third term can be written as a functional of a real-valued function f of three variables[1]

$$F_{\delta\rho-\Psi}[\Delta\rho, \Psi] = \int d\boldsymbol{q}_\perp f\left(q, \langle|\Psi|^2\rangle^{1/2}, \cos[6\varphi - 6\psi(\boldsymbol{r})]\right)(\delta\rho(\boldsymbol{q}_\perp))^2. \tag{S3}$$

From the Fourier representation of the free energy (eqns (S1-3)), we can find the a certain component $\delta\rho(\boldsymbol{q}_\perp)$ of the density fluctuations to the free energy and evaluate the mean squared amplitude of the density fluctuation within a single cell as

$$\langle|\delta\rho(\boldsymbol{q}_\perp)|^2\rangle = \frac{\int d(\delta\rho)\exp\left[-\frac{F_q}{k_B T}\right]|\delta\rho|^2}{\int d(\delta\rho)\exp\left[-\frac{F_q}{k_B T}\right]} = \frac{k_B T}{a + b(q_\perp - q_{\perp 0})^2 + f}. \tag{S4}$$

The same result can be obtained without direct evaluation of the integrals in eqn (S4), but noticing that both contributions to the free energy in eqns (S2) and (S3) are quadratic with respect to the fluctuations $\delta\rho(\boldsymbol{q}_\perp)$, which allows one to use classical equipartition theorem.

To obtain the total structure factor of the hexatic films, the result (S4) has to be averaged over the fluctuations of the BO order parameter $\Psi(\boldsymbol{r})$, i.e. over the hexatic degree of freedom. In the absence of the coupling term ($f \equiv 0$), the resulting structure factor will be described by a uniform scattering ring with a Lorentzian radial cross section, similar to the Sm-A phase. In the Hex-B phase, $f \neq 0$ and to describe the corresponding structure factor, the small fluctuations of the phase $\psi(\boldsymbol{r})$ should be taken into account. To do this it is convenient to use an angular Fourier expansion of $\langle|\delta\rho(\boldsymbol{q}_\perp)|^2\rangle$ over $\left(\varphi - \psi(\boldsymbol{r})\right)$

$$\langle|\delta\rho(\boldsymbol{q}_\perp)|^2\rangle = \sum_{p=-\infty}^{+\infty} S_{6p} e^{6ip(\varphi - \psi(\boldsymbol{r}))}, \tag{S5}$$

with the Fourier coefficients

$$S_{6p} = \frac{1}{\pi/3} \int_{-\pi/6}^{\pi/6} d\theta e^{-6ip\theta} \frac{k_B T}{a + b(q_\perp - q_{\perp 0})^2 + f\left(q, \langle|\Psi|^2\rangle^{1/2}, \cos 6\theta\right)}. \tag{S6}$$

Let us represent the phase $\psi(\boldsymbol{r})$ of the BO order parameter fluctuating around the mean value ψ_0 as $\psi(\boldsymbol{r}) = \psi_0 + \delta\psi(\boldsymbol{r})$, and perform averaging over the fluctuations $\delta\psi(\boldsymbol{r})$ independently for each term in the angular Fourier expansion

$$S(\boldsymbol{q}_\perp) = \sum_{p=-\infty}^{+\infty} S_{6p} e^{6ip(\varphi - \psi_0)} \langle e^{-6ip\delta\psi(\boldsymbol{r})}\rangle. \tag{S7}$$

We will consider the case of low temperature hexatic phase, when the fluctuations are small, $\delta\psi(\boldsymbol{r}) \approx 0$. Thus, we can use the Taylor expansion and keep only the first non-zero term

$$\langle e^{-6ip\delta\psi(\boldsymbol{r})}\rangle \approx \langle 1 - 6pi\delta\psi(\boldsymbol{r}) - \frac{\left(6p\delta\psi(\boldsymbol{r})\right)^2}{2}\rangle = 1 - 18p^2\langle\delta\psi^2\rangle \approx e^{-18p^2\langle\delta\psi^2\rangle}. \tag{S8}$$

Here we assumed that the mean value $\langle\delta\psi(\boldsymbol{r})\rangle = 0$ because the fluctuations $\delta\psi(\boldsymbol{r})$ and $-\delta\psi(\boldsymbol{r})$ have the same contribution to the free-energy and therefore are equally probable. The root-mean-square value $\langle\delta\psi^2\rangle = \langle(\psi - \psi_0)^2\rangle$ can be estimated in the frames of the $x - y$ model in 2D and 3D[4,5].

Combining eqns (S6) and (S7), the structure factor can be rearranged as

$$\begin{aligned} S(\boldsymbol{q}_\perp) &= \sum_{p=-\infty}^{+\infty} S_{6p} e^{6ip(\varphi-\psi_0)} e^{-18p^2\langle\delta\psi^2\rangle} \\ &= \frac{3}{\pi}\int_{-\pi/6}^{\pi/6} d\theta \left[\frac{k_B T}{a + b(q_\perp - q_{\perp 0})^2 + f\left(q, \langle|\Psi|^2\rangle^{1/2}, \cos 6\theta\right)} \sum_{p=-\infty}^{+\infty} e^{-6ip(\varphi-\psi_0-\theta)} e^{-18p^2\langle\delta\psi^2\rangle}\right] \end{aligned} \tag{S9}$$

It is easy to check that the sum under the integral converges to a Gaussian function (compare with eqns (S22-S27) in the next section):

$$\frac{3}{\pi}\sum_{p=-\infty}^{+\infty} e^{-6ip(\varphi-\psi_0-\theta)} e^{-18p^2\langle\delta\psi^2\rangle} = \frac{1}{\sqrt{2\pi\langle\delta\psi^2\rangle}} \sum_{n=-\infty}^{\infty} \exp\left[-\frac{\left(\theta - (\varphi - \psi_0) - \frac{\pi}{3}n\right)^2}{2\langle\delta\psi^2\rangle}\right]. \tag{S10}$$

Since integration over the angular variable θ in eqn (S9) is performed over a limited range from $-\frac{\pi}{6}$ to $\frac{\pi}{6}$, only one term in the sum over n will contribute to the hexatic structure factor (because $\langle\delta\psi^2\rangle$ is small and there is no overlap between various terms). This can be taken into account by selecting the reference axis in such a way that $\psi_0 = 0$ and considering the scattering in the same direction, i.e. $|\varphi| < \pi/6$. In this case, we can rewrite the hexatic structure factor as

$$S(\boldsymbol{q}_\perp) = \frac{k_B T}{\sqrt{2\pi\langle\delta\psi^2\rangle}} \int_{-\pi/6}^{\pi/6} d\theta \frac{e^{-\frac{(\theta-\varphi)^2}{2\langle\delta\psi^2\rangle}}}{a + b(q_\perp - q_{\perp 0})^2 + f\left(q, \langle|\Psi|^2\rangle^{1/2}, \cos 6\theta\right)}. \tag{S11}$$

Due to the sharp Gaussian peak in the nominator, the main contribution to $S(\boldsymbol{q}_\perp)$ comes from the region around $\theta \approx 0$. This allows us to expand the coupling function f into the Taylor series

$$f\left(q, \langle|\Psi|^2\rangle^{1/2}, \cos 6\theta\right) \approx f_0 + f_2\theta^2 \tag{S12}$$

The integration region in (S11) can be formally extended to infinity, because contribution from the large angles ($|\theta| > \pi/6 \gg \sqrt{\langle\delta\psi^2\rangle}$) is negligibly small:

$$S(\boldsymbol{q}_\perp) = \frac{k_B T}{\sqrt{2\pi\langle\delta\psi^2\rangle}} \int_{-\infty}^{+\infty} d\theta \frac{e^{-\frac{(\theta-\varphi)^2}{2\langle\delta\psi^2\rangle}}}{a + f_0 + b(q_\perp - q_{\perp 0})^2 + f_2\theta^2}. \tag{S13}$$

If $\langle\delta\psi^2\rangle \ll (a+f_0)/f_2$, one can expand the denominator into the Taylor series and keep only the first term

$$\frac{1}{a+f_0+b(q_\perp-q_{\perp 0})^2+f_2\theta^2} \approx \frac{1}{a+f_0+b(q_\perp-q_{\perp 0})^2}\left[1-\frac{f_2\theta^2}{a+f_0+b(q_\perp-q_{\perp 0})^2}\right] \quad \text{(S14)}$$

After this simplification, the integral (S13) can be easily evaluated, the radial cross section ($\varphi = 0$) of the diffraction peak from the hexatic phase can be described by a Lorentzian function

$$\begin{aligned} S(q_\perp) &= \frac{k_B T}{a+f_0+b(q_\perp-q_{\perp 0})^2}\left[1-\frac{f_2\langle\delta\psi^2\rangle}{a+f_0+b(q_\perp-q_{\perp 0})^2}\right] \\ &\approx \frac{k_B T}{a+f_0+f_2\langle\delta\psi^2\rangle+b(q_\perp-q_{\perp 0})^2} \propto \frac{1}{\gamma^2+(q_\perp-q_{\perp 0})^2} \end{aligned} \quad \text{(S15)}$$

with the half width at half maximum $\gamma = \sqrt{(a+f_0+f_2\langle\delta\psi^2\rangle)/b}$.
The azimuthal profile through the maximum of the diffraction peak ($q_\perp = q_{\perp 0}$) is given by the Voigt function which is a convolution of the Gaussian and Lorentzian functions

$$\begin{aligned} S(\varphi) &= \frac{k_B T}{\sqrt{2\pi\langle\delta\psi^2\rangle}}\int_{-\infty}^{+\infty} d\theta \frac{e^{-\frac{(\theta-\varphi)^2}{2\langle\delta\psi^2\rangle}}}{a+f_0+f_2\theta^2} = \pi\frac{k_B T}{\sqrt{f_2(a+f_0)}}G(\varphi;\sigma) * L(\varphi;\kappa) \\ &= \frac{k_B T\pi}{\sqrt{f_2(a+f_0)}}V(\varphi;\sigma,\kappa). \end{aligned} \quad \text{(S16)}$$

Here the Gaussian function

$$G(\varphi;\sigma) = \frac{1}{\sqrt{2\pi\sigma^2}}e^{-\frac{\varphi^2}{2\sigma^2}} \quad \text{(S17)}$$

is defined by the parameter $\sigma = \sqrt{\langle\delta\psi^2\rangle}$ which depends only on the mean squared fluctuations of the phase $\psi(\boldsymbol{r})$. The Lorentzian function

$$L(\varphi;\kappa) = \frac{\kappa}{\pi(\varphi^2+\kappa^2)} \quad \text{(S18)}$$

has the half width at half maximum $\kappa = \sqrt{(a+f_0)/f_2}$ which has contribution from the short-range position order (coefficient a in the Landau expansion of the free energy (2)) and the coupling between the positional and BO order (coefficients f_0 and f_2 in eqn (S12)).

2 Angular profile of the hexatic peak in the multicritical scaling theory

A natural way to describe the BO order in the hexatic phase is to expand the azimuthal dependence of the structure factor into the Fourier series[6]

$$I(q,\varphi) = I_0(q)\left[1 + 2\sum_{m=1}^{\infty} C_{6m}(q)\cos\big(6m(\varphi-\varphi_0)\big)\right], \tag{S19}$$

where the coefficients $0 \le C_{6m} \le 1$ depend on the degree of the orientational order. The multicritical scaling theory predicts the following relation between the coefficients C_{6m} of different order

$$C_{6m} = (C_6)^{m+\lambda m(m-1)}, \tag{S20}$$

where $\lambda \approx 0.3$ for 3D hexatic phase,[6,7] and $\lambda \approx 1$ for 2D hexatic phase.[4]
First, let us show that the series (S19) converges to a Gaussian function in the 2D case ($\lambda = 1$). Indeed,

$$\begin{aligned} 1 + 2\sum_{m=1}^{\infty} C_{6m}\cos\big(6m(\varphi-\varphi_0)\big) &= 1 + 2\sum_{m=1}^{\infty} (C_6)^{m^2}\cos\big(6m(\varphi-\varphi_0)\big) \\ &= \sum_{m=-\infty}^{\infty} (C_6)^{m^2} e^{i6m(\varphi-\varphi_0)} = \sum_{m=-\infty}^{\infty} \exp\left[-m^2 \ln\frac{1}{C_6} + i\cdot 6m(\varphi-\varphi_0)\right]. \end{aligned} \tag{S21}$$

At the same time, the azimuthal profile of the six hexatic peaks, each described by a Gaussian function, can be written as

$$I_G(\varphi) = \frac{I_0}{\sqrt{2\pi\sigma^2}}\sum_{n=-\infty}^{\infty}\exp\left[-\frac{\left(\varphi-\varphi_0-\frac{2\pi}{6}n\right)^2}{2\sigma^2}\right], \tag{S22}$$

where φ_0 defines the angular position of the peaks with respect to some reference axis, and the term $\frac{2\pi}{6}n$ appears due to the sixfold symmetry of the hexatic structure factor. The periodic function (S22) can be expanded into the Fourie series

$$I_G(\varphi) = \sum_{m=-\infty}^{\infty} C_{6m} e^{i6m\varphi} \tag{S23}$$

with the Fourier coefficients

$$\begin{aligned} C_{6m} &= \frac{1}{2\pi/6}\int_{\varphi_0-\frac{\pi}{6}}^{\varphi_0+\frac{\pi}{6}} I_G(\varphi)e^{-i6m\varphi}d\varphi \\ &= \frac{3I_0}{\pi\sqrt{2\pi\sigma^2}}\sum_{n=-\infty}^{\infty}\int_{\varphi_0-\frac{\pi}{6}}^{\varphi_0+\frac{\pi}{6}}\exp\left[-\frac{\left(\varphi-\varphi_0-\frac{2\pi}{6}n\right)^2}{2\sigma^2} - i6m\varphi\right]d\varphi. \end{aligned} \tag{S24}$$

Assuming that the hexatic peaks are sharp and do not overlap, we can neglect all terms with $n \neq 0$ (they correspond to the peaks of $I_G(\varphi)$ outside of the integration region). Then we can extend the region of integration to infinity

$$C_{6m} = \frac{3I_0}{\pi\sqrt{2\pi\sigma^2}} \int_{-\infty}^{+\infty} \exp\left[-\frac{(\varphi-\varphi_0)^2}{2\sigma^2} - i6m\varphi\right] d\varphi, \tag{S25}$$

since the main contribution to the integral (S24) comes from the vicinity of the peak at $\varphi = \varphi_0$.
Now substituting $\varphi = t \cdot \sigma\sqrt{2} + \varphi_0$, we can evaluate the coefficients

$$C_{6m} = \frac{3I_0\sigma\sqrt{2}}{\pi\sqrt{2\pi\sigma^2}} e^{-i6m\varphi_0} e^{-18\sigma^2 m^2} \int_{-\infty}^{+\infty} e^{-\left(t+i3m\sigma\sqrt{2}\right)^2} dt = \frac{3I_0}{\pi} e^{-i6m\varphi_0} e^{-18\sigma^2 m^2}. \tag{S26}$$

Thus, the Fourier series (S23) can be written as

$$I_G(\varphi) = \frac{3I_0}{\pi} \sum_{m=-\infty}^{\infty} \exp[-18m^2\sigma^2 + i \cdot 6m(\varphi - \varphi_0)], \tag{S27}$$

which coincides with the multicritical theory expansion (S21) up to the prefactor, assuming $C_6 = e^{-18\sigma^2}$. Since the Fourier expansion is unique, we can conclude that the scaling law (S20) with $\lambda = 1$ (2D case) corresponds to the Gaussian shape of the hexatic peaks in the azimuthal direction.
In the 3D case ($\lambda \approx 0.3$), the angular Fourier series in eqn (S19) can be written as

$$\begin{aligned} 1 + 2\sum_{m=1}^{\infty} C_{6m} \cos\big(6m(\varphi-\varphi_0)\big) &= \sum_{m=-\infty}^{\infty} (C_6)^{\lambda m^2 + (1-\lambda)|m|} e^{i6m(\varphi-\varphi_0)} \\ &= \sum_{m=-\infty}^{\infty} \exp\left[-m^2\lambda \ln\frac{1}{C_6} - |m|(1-\lambda)\ln\frac{1}{C_6} + i\cdot 6m(\varphi-\varphi_0)\right] \\ &= \sum_{m=-\infty}^{\infty} \exp[-18m^2\sigma^2 - 6\kappa|m| + i\cdot 6m(\varphi-\varphi_0)], \end{aligned} \tag{S28}$$

where the parameters σ and κ can be expressed through the parameters of the multicritical scaling theory:

$$\sigma = \sqrt{\frac{\lambda}{18}\ln\frac{1}{C_6}}, \qquad \kappa = \frac{1-\lambda}{6}\ln\frac{1}{C_6}. \tag{S29}$$

Now the angular Fourier coefficients C_{6m} are a product of $\exp[-m^2\sigma^2]$, which corresponds to the Fourier coefficients of a Gaussian function (see the derivation above), and $\exp[-6\kappa|m|]$, which corresponds to the Fourier coefficients of a Lorentzian function. Let us prove this by expanding the periodic Lorentzian function into the Fourier series

$$I_L(\varphi) = I_0\frac{\kappa}{\pi}\sum_{n=-\infty}^{\infty} \frac{1}{\left(\varphi-\varphi_0-\frac{2\pi}{6}n\right)^2 + \kappa^2} = \sum_{m=-\infty}^{\infty} C_{6m} e^{i6m\varphi}, \tag{S30}$$

and evaluating the Fourier coefficients in a similar way, as it was done for the Gaussian function in eqns (S24-S26):

$$C_{6m} = \frac{1}{2\pi/6} \int_{\varphi_0-\frac{\pi}{6}}^{\varphi_0+\frac{\pi}{6}} I_L(\varphi) e^{-i6m\varphi} d\varphi = \frac{3\kappa I_0}{\pi^2} \sum_{n=-\infty}^{\infty} \int_{\varphi_0-\frac{\pi}{6}}^{\varphi_0+\frac{\pi}{6}} \frac{e^{-i6m\varphi}}{\left(\varphi - \varphi_0 - \frac{2\pi}{6} n\right)^2 + \kappa^2} d\varphi$$

$$= \frac{3\kappa I_0}{\pi^2} \int_{-\infty}^{+\infty} \frac{e^{-i6m\varphi}}{(\varphi - \varphi_0)^2 + \kappa^2} d\varphi = \frac{3\kappa I_0}{\pi^2} e^{-i6m\varphi_0} \int_{-\infty}^{+\infty} \frac{e^{-i6m\varphi}}{\varphi^2 + \kappa^2} d\varphi \tag{S31}$$

$$= \frac{3\kappa I_0}{\pi^2} e^{-i6m\varphi_0} \frac{\pi}{\kappa} e^{-\kappa|6m|} = \frac{3I_0}{\pi} e^{-6\kappa|m| - i6m\varphi_0}.$$

Therefore, the Fourier expansion of the Lorentzian function (S30) can be written as

$$I_L(\varphi) = \frac{3I_0}{\pi} \sum_{m=-\infty}^{\infty} \exp[-6\kappa|m| + i \cdot 6m(\varphi - \varphi_0)], \tag{S32}$$

proving that the term $\exp[-6\kappa|m|]$ in eqn (S28) indeed corresponds to the Lorentzian function.
Using the convolution theorem, we can expect that the Fourier series (S28), in which each term is equal to the product of the Fourier components of the Gaussian and Lorentzian functions, will converge to a convolution of these two functions.
Therefore, from eqns (S19) and (S28) it follows that for the 3D hexatics the angular distribution of intensity can be written as

$$I_V(\varphi) = I_0 \sum_{n=-\infty}^{\infty} V\left(\varphi - \varphi_0 - \frac{2\pi}{6} n; \sigma, \gamma\right), \tag{S33}$$

where $V(\varphi; \sigma, \kappa)$ is the Voigt function

$$V(\varphi; \sigma, \gamma) = G(\varphi; \sigma) * L(\varphi; \kappa) = \int_{-\infty}^{+\infty} d\theta \frac{e^{-\frac{\theta^2}{2\sigma^2}}}{\sqrt{2\pi\sigma^2}} \cdot \frac{\frac{\kappa}{\pi}}{(\varphi - \theta)^2 + \kappa^2}, \tag{S34}$$

which is a convolution of a Gaussian $G(\varphi; \sigma)$ and a Lorentzian $L(\varphi; \kappa)$ functions defined in eqns (S17-S18).

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