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Correlated and decorrelated positional and orientational order in the nucleosomal core particle mesophases

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Abstract. – We investigate the orientational order of transverse-polarization vectors of long columns of nucleosomal core particles and their coupling to positional order in high-density mesophases discovered recently. Inhomogeneous polar ordering of these columns precipitates crystallization of the 2D sections with different orientations of the transverse-polarization vector on each column in the unit cell. We propose possible scenarios for going from the 2D hexagonal phase into distorted lamellar and related phases observed experimentally.

The problem of efficient genome compaction in the evolution of eucaryotic organisms was solved on the smallest scale via the formation of DNA-histone protein complexes, that make a fundamental unit of chromatin: the nucleosome [1]. A nucleosome is composed of a 146 bp long DNA fragment, wrapped 1.75 times around the histone octamer core composed of four different histone proteins and a DNA linker that connects two consecutive nucleosomes [2]. Digestion of the linker DNA between different nucleosomes along the eucaryotic genome gives rise to the *nucleosome core particle* (NCP). NCP has an approximately cylindrical shape (fig. 1a) of height and radius ~ 55 Å, and a structural charge of ~ -165. This complex is stable in aqueous solutions from mM to 750 mM monovalent salt ionic strength [3].

At high enough concentration the solution of NCPs ceases to be isotropic. Competing, as yet poorly understood [4,5], microscopic interactions between NCPs in concentrated solutions give rise to a slew of liquid-crystalline phases of fascinating complexity [6]. At low salt concentrations and sufficiently high osmotic pressure NCPs aggregate into columns of almost ideal cylindrical shape which contain dozens of individual particles (fig. 1c). These columns then aggregate to form a lamellar phase with each lamella being formed by a bilayer of parallel columns of NCPs [7]. In each bilayer the columns are arranged in slightly distorted close packing of two rows of cylinders (fig. 2c). These bilayers alternate with layers of solvent to

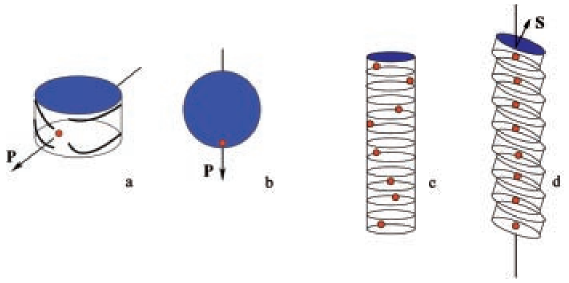

Fig. 1 – Nucleosomal core particles and their columnar aggregates. (a) DNA (solid curve) wrapped around the histone core; the front side with free DNA ends is different with respect to the back side and defines the orientation of the NCP dyadic axis [6]. (b) NCP top view: the small circle denotes the entry of the dyadic axis. (c) Column of aggregated NCP with disordered dyadic axes. (d) Column of aggregated NCP with ordered dyadic axes and tilted S-axes.

form a long-range lamellar structure with period varying from 27 to 42 nm, depending on NCP content in the solution. At higher salt concentrations this phase gives way to a 2D hexagonal columnar liquid crystalline phase (fig. 2a) leading at still higher salt concentrations to a 3D hexagonal crystal [8].

The NCP particle is characterized by a high shape and charge asymmetry due to the DNA wrapping around the histone core. The entry-exit point of the DNA free ends marks a s.c. dyadic axis (fig. 1a and b). Due to different densities of the DNA crown and the histone core, the dyadic axis is readily visible on the cryoelectron micrographs [7]. This allows one to actually observe the local orientational order of the NCPs, described by its in-plane dyadic axis vector, conventionally called the transverse polarization [9]. In the lamellar phase the dyadic axes of individual NCPs are strongly correlated along the column leading to a non-zero average transverse-polarization vector $\boldsymbol{P}$ of a column (fig. 1d). Cryomicroscopy studies [7] give strong evidence that the dyadic axes, and consequently the vectors $\boldsymbol{P}$, of columns in the two layers of a lamella are on the average in an antiparallel direction, normal to the bilayer, with the DNA exit point facing the solvent layer. In addition, due to nearly close packing in a bilayer, the columns in the two layers are shifted by a small but constant distance in such a way that the antiparallel dyadic axes form a periodic polar vector field (fig. 2c).

The 2D hexagonal phase is characterized, in turn, by a positional order in the plane perpendicular to the columns (fig. 2a), but dyadic axes of NCPs are orientationally disordered along the columns (fig. 1c). The corresponding average polar vector of the column is zero. The

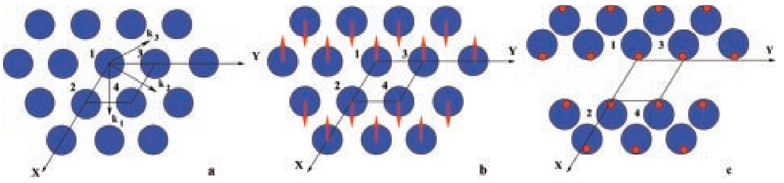

Fig. 2 – Hexagonal (a) and lamellar (c) phases. Antiparallel displacement field (b) for the transition from the hexagonal to lamellar phase. 2D sections perpendicular to the columns are presented. The hexagonal unit cell formed by columns 1, 2, 3 and 4 is distinguished. The directions of the wave vectors of the polar vector field are indicated by arrows in (a). Small circles indicate the orientations of the ordered columns dyadic axes (average polarization of the columns) in (c).

main task of the present letter is to describe the transition between these two phases which consists in simultaneous ordering of the NCPs dyadic axes along an individual column and their periodic orientational correlation between the columns, accompanied by collective columns' displacements. Detailed analysis of this mechanism permits to predict other ordered phases (including the inverted hexagonal phase observed recently in condensed NCP solutions [6]) induced by the dyadic axes ordering and related columns' displacements.

We also briefly mention several fine peculiar features of the NCP lamellar phase structure, namely, i) decorrelation of columns positions from one bilayer to another, ii) orientation of NCPs with respect to the column axis and its relation to NCP chirality, which will be thoroughly discussed in a separate publication [10]. Both refinements of the lamellar phase structure come from detailed SAXS measurements [8] indicating that the lamellar phase in the NCP-solvent system has some features apparently similar to those of sliding phases in lipid-DNA complexes [11–13], but in contrast to the sliding phase in lipid-DNA complexes it has no cross-correlation peaks and indicates classical liquid-crystalline behavior. SAXS also clearly indicates that in individual columns in the lamellar phase the axis of the flattened NCP cylinder is tilted with respect to the column axis (fig. 1d), while both axes are parallel in the 2D hexagonal phase (fig. 1c).

In what follows we will investigate the nature of positional and orientational order in phases that are induced by an ordering of the NCPs dyadic axes. We adopt a Landau theory approach based on symmetry analysis of the structural changes from 2D hexagonal to lamellar. This approach is standardly used in various fields of condensed-matter physics [14]; its basic notions and numerous examples of applications can be found in [15]. In a complex system with several competing interactions, this approach permits to obtain results that are not very sensible to model assumptions. In the framework of this theory, phase transition is considered to be driven by a critical mode condensation. A physical quantity (order parameter) corresponding to the mode has non-zero average value in the ordered phase and vanishes in the disordered one. It spans an irreducible representation of the symmetry group of the disordered structure.

The hexagonal-to-lamellar transition in NCP columnar aggregates is driven by the condensation of the periodic antiparallel transverse-polarization field mode resulting in the dyadic axes orientation as given in fig. 2c. The amplitude of this mode vanishes in the 2D hexagonal phase rendering the columns non-polar (fig. 1c) and the phase structure disordered. Basing our assumptions on experiments, we introduce the transverse-polarization order in a lamella described with $\boldsymbol{P}(\boldsymbol{\rho})$, where $\boldsymbol{\rho} = (x, y)$, which is periodic and highly anisotropic. We establish the representation spanned by this critical mode and corresponding Landau free energy which helps us to study relative thermodynamic stability of the phases.

Let us also stress an evident duality between the polar vector carried by a NCP column (average orientation of dyadic axes) and the polar vector of column's displacement in the solvent which has important consequences for the structure of the ordered phases. Since the displacements $\boldsymbol{u}_i$ of NCP columns and the polar vectors $\boldsymbol{P}_i$ of the dyadic axes span the same irreducible representation of the symmetry group of the 2D hexagonal phase, the components of the order parameter of the hexagonal-to-lamellar transition can be expressed in the same form either in terms of columns' polarizations $\boldsymbol{P}_i$ or in terms of the columns' displacements $\boldsymbol{u}_i$. In condensed NCP solutions, this general rule leads to the proportionality between $\boldsymbol{P}_i$ responsible for the *orientational* order and $\boldsymbol{u}_i$ expressing the *positional* one. Interactions between individual NCPs thus lead to the *simultaneous* antiparallel dyadic axes orientation (fig. 2c) and antiparallel columns displacements (fig. 2b). This fact permits one to obtain the final structures of the ordered phases. In the following quantitative study of the periodic transverse polar vector field we will follow orientational degrees of freedom $\boldsymbol{P}_i$, the final results having exactly the same form for the positional order $\boldsymbol{u}_i$.

If the transverse-polarization vector field condensation starts from a 2D hexagonal phase, then in general one would have several different choices for the direction of the wave vector $\boldsymbol{k}$ with respect to the symmetry planes of the 2D hexagonal phase. In the hexagonal coordinate system the wave vector of the condensation into the lamellar phase has to be described in Kovalev's notation [16] by $\boldsymbol{k}_{12} = \frac{1}{2}\boldsymbol{b}_1$, where $\boldsymbol{b}_1$ is one of the reciprocal basis vectors. The irreducible star of the $\boldsymbol{k}_{12}$ vector has three arms given by $\boldsymbol{k}_1 = \boldsymbol{k}_{12}^{(1)} = \frac{1}{2}\boldsymbol{b}_1$, $\boldsymbol{k}_2 = \boldsymbol{k}_{12}^{(2)} = \frac{1}{2}\boldsymbol{b}_2$ and $\boldsymbol{k}_3 = \boldsymbol{k}_{12}^{(3)} = \frac{1}{2}(-\boldsymbol{b}_1 + \boldsymbol{b}_2)$. They indicate three equivalent directions (fig. 2a) in the hexagonal structure for which the transverse-polarization field can change the translational symmetry of the structure in the same way. The basis functions of a polar vector field with this periodicity are given by $\psi_1 = (2\hat{e}_x + \hat{e}_y)\ e^{i\boldsymbol{k}_1\cdot\boldsymbol{r}}$, $\psi_2 = (\hat{e}_x + 2\hat{e}_y)\ e^{i\boldsymbol{k}_2\cdot\boldsymbol{r}}$ and $\psi_3 = (-\hat{e}_x + \hat{e}_y)\ e^{i\boldsymbol{k}_3\cdot\boldsymbol{r}}$. The physical realization of the order parameter is given by the symmetry coordinates [15] composed of the column's polar vectors $\boldsymbol{P}_i$. Denoting by i the index of the column, in such a way that its (x, y) coordinates are $i = 1 \to (0, 0)$, $i = 2 \to (1, 0)$, $i = 3 \to (0, 1)$ and $i = 4 \to (1, 1)$ in the 2D hexagonal unit cell (see fig. 2), and the 2D polar vector components by (P_x, P_y), one gets the following components of the order parameter:

$$\begin{aligned}
\eta_1 &= P_{x1} - P_{x2} + P_{x3} - P_{x4} + \frac{1}{2}\left(P_{y1} - P_{y2} + P_{y3} - P_{y4}\right),\\
\eta_2 &= \frac{1}{2}\left(P_{x1} + P_{x2} - P_{x3} - P_{x4}\right) + P_{y1} + P_{y2} - P_{y3} - P_{y4},\\
\eta_3 &= \frac{1}{2}\left(-P_{x1} + P_{x2} + P_{x3} - P_{x4} + P_{y1} - P_{y2} - P_{y3} + P_{y4}\right).
\end{aligned} \tag{1}$$

The Landau free energy of this system is now given via the orientation probability density function [14,15] $\rho(x, y, z) = \rho_0 + \sum_{i=1}^{3} \eta_i\psi_i(x, y, z)$, in the form $\mathcal{F} = \int f(\eta_i\psi_i(x, y, z))\,\mathrm{d}x\,\mathrm{d}y\,\mathrm{d}z = F(\eta_i)$. For the sake of clarity, we omit the terms stemming from the large-scale elastic deformations of this order. Consequently, the free energy of the system can be considered as a simple function of the order parameter components.

To establish the exaustive list of the phases induced by the considered condensation mechanism we use the fact that $F(\eta_i)$ has to be an invariant function of the irreducible representation spanned by the periodic transverse-polarization vector field. Since the basis invariants of this representation are $I_1 = \eta_1^2 + \eta_2^2 + \eta_3^2$, $I_2 = \eta_1^2\eta_2^2 + \eta_2^2\eta_3^2 + \eta_3^2\eta_1^2$ and $I_3 = \eta_1^2\eta_2^2\eta_3^2$, the Landau free energy has to have the form $\mathcal{F} = F(I_1, I_2, I_3)$. The minimization of this free energy with respect to η_i gives the equation of state. There are six different solutions of this equation of state which we list below in terms of η_i, the unit cell multiplication in the ordered state with respect to the 2D hexagonal phase, *i.e.* $V_{\mathrm{ord}}/V_{\mathrm{hex}}$ and their space groups. Let us show that the first solution, *i.e.* $\eta_1 \neq 0$, $\eta_{2,3} = 0$, describes the lamellar state. From eq. (1) it follows that in this case $\boldsymbol{P}_1 = \boldsymbol{P}_3$ and $\boldsymbol{P}_2 = \boldsymbol{P}_4$, or, in other words, that the orientations of dyadic axes carried by columns 1 and 3 or columns 2 and 4 are parellel and have the same degree of correlation along the column. Thus, due to the duality, the displacements of the columns 1 and 3 as well as 2 and 4 are also identical (fig. 2b). This means that the periodic transverse-polarization field induces a lamellar structure, where a single lamella is defined by the columns 1, 3, 1, 3... for the bottom part and 2, 4, 2, 4... for the top part of a single lamella (fig. 2c). The order parameter in this lamellar phase induced by the periodic transverse-polarization vector ordering is given by

$$\eta_1 = (2P_{x1} + P_{y1}) - (2P_{x2} + P_{y2}). \tag{2}$$

The simplest phenomenological Landau free-energy expansion of an isolated transition from a hexagonal 2D crystal $\eta_i = (0, 0, 0)$ to a lamellar 2D crystal with $\eta_i = (\eta, 0, 0)$, with basis

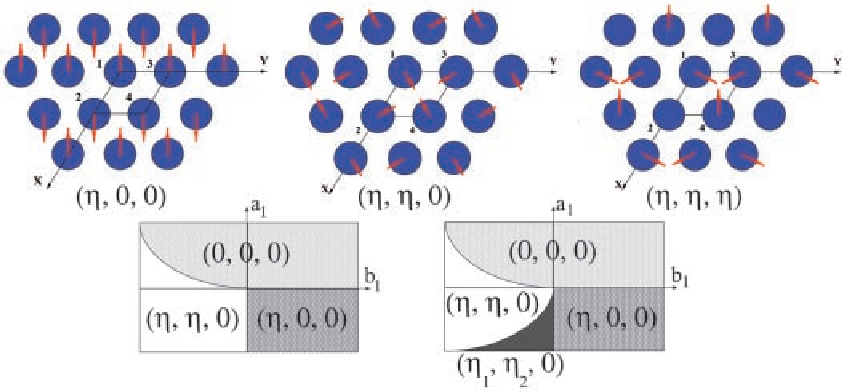

Fig. 3 – Displacement fields for the transition from the hexagonal to lamellar $(\eta, 0, 0)$ (columns forming a single lamella are enclosed within a gray rectangle), orthorhombic $(\eta, \eta, 0)$ and inverted hexagonal (η, η, η) phases. Below, the phase diagram stemming from eqs. (4), (5).

invariants $I_1 = \eta^2$, $I_2 = \eta^4$ and $I_3 = \eta^6$ can be written as

$$\mathcal{F} = a_1 I_1 + b_1 I_2 + \dots . \tag{3}$$

This free energy describes a second-order transition between a hexagonal and a lamellar phase, driven trivially by the parameter a_1, that can depend on various solution conditions, *e.g.* the salt concentration. The same periodic transverse-polarization vector field can induce also more complicated transitions. Assuming now a general form of the order parameter we can start with the following free-energy *ansatz*:

$$\mathcal{F}_1(\eta_i) = a_1 I_1 + a_2 I_1^2 + b_1 I_2 + \dots . \tag{4}$$

The phase diagram in this case is presented in fig. 3. One can observe that in addition to the lamellar phase, stable for $b_1 > 0$, the diagram shows the $(\eta, \eta, 0)$ phase, to be discussed below, stable for $b_1 < 0$. The growth of the complexity of the phase diagram as more non-linear terms are included into the Landau free energy is clearly apparent. *E.g.*, if one starts from the free energy

$$\mathcal{F}_2(\eta_i) = \mathcal{F}_1(\eta_i) + a_3 I_1^3 + c_1 I_3 + d_{12} I_1 I_2 + \dots , \tag{5}$$

this leads to the form of the phase diagram presented in fig. 3, where an additional phase, this time $(\eta_1, \eta_2, 0)$, intercalates between the $(\eta, \eta, 0)$ and the $(\eta, 0, 0)$ phases.

Table I – *Different solutions of the equation of state.*

η_i	$V_{\rm ord}/V_{\rm hex}$	Space group
η, 0, 0	2	C_{2v}^1
η, η, η	4	D_{3h}^1
η, η, 0	4	D_{2h}^1
η_1, η_2, 0	4	C_{2h}^1
η_1, η_1, η_2	4	C_{2v}^1
η_1, η_2, η_3	4	C_S^1

The structure of other phases listed in table I, induced by the periodic ordering of the transverse-polarization vector field obtained in highly non-linear free-energy models can be quite complicated. We have already analysed the lamellar phase, beside this we also have:
1) (η,η,η). The corresponding vector field is given in this phase by the vectors of the different columns in the form

$$\begin{aligned} \boldsymbol{P}_1 &= (P_x, 2P_y), \qquad \boldsymbol{P}_2 = (0,0), \\ \boldsymbol{P}_3 &= (P_x, -P_y), \qquad \boldsymbol{P}_4 = (-2P_x, -P_y). \end{aligned} \tag{6}$$

The vectors of orientation of the columns are expressed in terms of one polar vector (P_x, P_y). Consequently, the displacements of the columns $\boldsymbol{u}_i$ are expressed in the same form but in terms of (u_x, u_y). Note that, because of the specific interaction between the columns in this phase, column 2 is frustrated: its average polar vector is zero. Consequently, its displacement with respect to the position in the 2D hexagonal phase is zero and the dyadic axes of individual NCPs are disordered along this column. All three components of the order parameter are equal to the following combination of $\boldsymbol{P}_i$ vectors: $\eta = P_{x1} + 2P_{y1} + P_{x3} - P_{y3} - 2P_{x4} - P_{y4}$. Analyzing the displacement field in this phase, see fig. 3, it is obvious that the columns of a hexagonal phase are now displaced to make an inverted hexagonal phase, observed also in experiments [6], whose vertices are given by the columns $3'$, 2, $1'$, 2, $4'$ and 2.
2) $(\eta,\eta,0)$. The corresponding dyadic axes orientation vector field in this phase is given by

$$\begin{aligned} \boldsymbol{P}_1 &= \frac{3}{2}(P_x, P_y), \qquad \boldsymbol{P}_2 = \frac{1}{2}(-P_x, P_y), \\ \boldsymbol{P}_3 &= \frac{1}{2}(P_x, -P_y), \qquad \boldsymbol{P}_4 = -\frac{3}{2}(P_x, P_y). \end{aligned} \tag{7}$$

The displacement field in this phase has the same form in terms of (u_x, u_y) and the final structure is easily obtained using these two results. Both non-zero components of the order parameter in this phase are equal to $\eta = \frac{3}{2}(P_{x1}+P_{y1})+\frac{1}{2}(-P_{x2}+P_{y2})+\frac{1}{2}(P_{x3}-P_{y3})-\frac{3}{2}(P_{x4}+P_{y4})$. The columns of a hexagonal phase undergoing the orientational and displacement fields, see fig. 3, now form an orthorhombic phase with a large unit cell. It can be understood as a succession of two series of columns undergoing 1D displacement waves: in the first series the wave is transverse and in the second one it is longitudinal. Note that the point symmetry of this structure shows only slight deviation from the hexagonal symmetry.
3) Along the same line the vector fields and the structures of the low-symmetry phases $(\eta_1,\eta_2,0)$, (η_1,η_1,η_2) and (η_1,η_2,η_3) can be obtained.

Let us now briefly discuss also the fine structure of the lamellar phase. We have shown above how a 2D ordering of a transverse vector polarization field can induce a transition into a lamellar phase $\eta_1 \neq 0$, $\eta_{2,3} = 0$. Its structure can be conventionally called crystal lamellar because it shows correlations between column positions both inside a bilayer and from one bilayer to another. The experimentally observed lamellar phase is however not exactly of this type. The positional order in it is partially destroyed making it basically equivalent to a smectic phase. The difference with respect to a crystal lamellar phase is subtle and refers to the fact that in SAXS in addition to the smectic peaks only reflections with two non-zero indices from the NCPs correlated along the column and within a single lamella are present. No reflection with three non-zero indices which can evidence the correlation between different lamellae is observed. This means that the *macroscopic* symmetry of this phase is higher than that of the crystal lamellar phase with $\eta_i = (\eta, 0, 0)$ and equivalent to the symmetry of a usual lamellar smectic phase ($G = D_{\infty h} \otimes T_d^z$). But within a lamella, both positional and orientational order are equivalent to those in the crystal lamellar phase. One can speculate that this decorrelation is related to the difference in correlation lengths of microscopic interactions which insure the stability of NCP mesophases on the one hand and their colloidal character on the other hand. Indeed, the distances between centers of columns in a 2D hexagonal colloidal crystal are greater than the column diameter (fig. 2a). The displacements of columns during hexagonal-to-lamellar transition in condensed NCP solutions are much greater than typical values of atomic shifts in a solid close-packed hexagonal crystal. Resulting distance d_L between the lamellae in our case can become greater than the correlation length of the interaction between the NCPs but still smaller than the correlation length of the smectic density wave $\xi_{\rho(z)}$. Since the symmetry of the crystal lamellar structure is lower than that of the actual lamellar smectic phase, the decorrelation must be accompanied by vanishing of a physical quantity responsible for this structural change. In other words, the order parameter of the transformation between smectic lamellar and crystal lamellar phases can be defined and easily expressed in terms of the periodic transverse-polarization field [10], along similar lines as first proposed by Bak [17] in a different context.

Let us finally discuss the influence of chirality on the state of NCP columns. Each NCP particle being chiral, their aggregation results in a chiral column with the rotational symmetry of a cylinder without neither mirror planes nor inversion, and the periodicity approximately equal to the individual NCP height. According to the Curie principle, ordering of the polar vector $\boldsymbol{P}_i$ along a chiral column leads to a simultaneous ordering of the axial vector $\boldsymbol{t}_i$, since both $\boldsymbol{P}_i$ and $\boldsymbol{t}_i$ span the same representation of the column symmetry group. In a chiral periodic aggregate the axial vector has an evident physical interpretation. Non-zero average value of $\boldsymbol{t}_i$ corresponds to the correlated tilt of individual NCPs with respect to the column axis (fig. 1d). In the 2D hexagonal phase the NCP columns carry zero average polar vector, then in a column the tilt of the particles is also zero (fig. 1c): the axes of individual particles (usually called S-axes) are parallel to the column axis. On the contrary, in the lamellar phase the ordering of the polar dyadic axes induces a tilt of the NCPs S-axes. This simple consequence of the NCP chirality can explain peculiarities of the SAXS spectrum observed in [8].

REFERENCES

[1] SCHIESSEL H., *J. Phys. Condens. Matter*, **15** (2003) R699.

[2] LUGER K. *et al.*, *Nature*, **251** (1997) 251.

[3] MANGENOT S., Thesis, Université Paris XI, UFR Scientifique d'Orsay (2001).

[4] MANGENOT S. *et al.*, *Eur. Phys. J. E*, **7** (2002) 221.

[5] PODGORNIK R., *J. Chem. Phys.*, **118** (2003) 11286.

[6] MANGENOT S. *et al.*, *J. Mol. Biol.*, **333** (2003) 907.

[7] LEFORESTIER A., DUBOCHET J. and LIVOLANT F., *Biophys. J.*, **81** (2001) 2414.

[8] MANGENOT S. *et al.*, *Biophys. J.*, **84** (2003) 2570.

[9] LORMAN V., PODGORNIK R. and ŽEKŠ B., *Phys. Rev. Lett.*, **87** (2001) 218101.

[10] LORMAN V., PODGORNIK R. and ŽEKŠ B., in preparation.

[11] SALDITT T. *et al.*, *Phys. Rev. Lett.*, **79** (1997) 2582.

[12] ARTZNER F. *et al.*, *Phys. Rev. Lett.*, **81** (1998) 5015.

[13] O'HERN C. S. and LUBENSKY T. C., *Phys. Rev. Lett.*, **80** (1998) 4345; O'HERN C. S., LUBENSKY T. C. and TONER J., *Phys. Rev. Lett.*, **83** (1999) 2745.

[14] LANDAU L. D. and LIFSHITZ E. M., *Statistical Physics* (Pergamon, London) 1958.

[15] IZYUMOV YU. A. and SYROMYATNIKOV V. N., *Phase Transitions and Crystal Symmetry* (Kluwer, Amsterdam) 1990.

[16] KOVALEV O. V., *Representations of the Crystallographic Space Groups: Irreducibles Representations, Induced Representations and Corepresentations* (Gordon and Breach, Amsterdam) 1993.

[17] BAK P., *Phys. Rev. B*, **20** (1979) 2209.