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P. Ziherl[a]; S. Žumer[a]
[a] Department of Physics, University of Ljubljana, Ljubljana, Slovenia

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Director fluctuations in inhomogeneously ordered nematic liquid crystals: An extension of elastic molecular field

by P. ZIHERL* and S. ŽUMER
Department of Physics, University of Ljubljana, Jadranska 19, 1000 Ljubljana, Slovenia

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In order to describe the order director fluctuations in nematic liquid crystalline systems with inhomogeneous order parameter, the approach based on Frank elastic theory is extended by introducing spatially dependent rotational viscosity and elastic constants. Using the proposed model, eigenmodes of director fluctuations in the vicinity of a disclination line of strength 1 are examined. Particular attention is paid to the behaviour of fluctuations in the vicinity of the structural transition in a cylindrical cavity.

1. Introduction

During the past few years, the theoretical understanding of orientational fluctuations in nematic liquid crystals has been broadened from bulk and planar systems to some simple curved geometries [1–4]. Usually, the director dynamics is analysed in terms of Frank theory of liquid crystalline elasticity and the concept of the so-called molecular field [5]. In the case of confined systems, the elementary framework, first introduced by the Orsay group [6], must be extended to allow for the interaction between the liquid crystal and the surrounding material [7].

However, this is usually not enough for the description of the director dynamics in restricted geometries. Due to purely topological reasons, equilibrium director configurations in confined systems are frequently characterized by the presence of disclination lines and point defects. The cores of these objects are often described as regions of isotropic liquid, which are energetically more favourable than the highly distorted nematic liquid crystal even well below the clearing temperature [8, 9]. From this simple point of view, the nematic liquid crystal is not bound only by the surrounding material, but also by the cores of the disclination lines or point defects, whichever occur in a given system. This is actually the basic assumption of the most frequently used model of the interaction between nematic director and disclination line. In this model, the disclination line is represented by a homogeneous cylindrical isotropic core [4, 10, 11]. The static and dynamic properties of the nematic–disclination line interface are characterized by core radius, anchoring at the surface of the core, and the corresponding interfacial rotational viscosity (which is throughout the paper referred to as *surface viscosity*).

The main advantages of this model are simplicity and tractability. On the other hand, it suffers from two severe disadvantages. One of these is the assumption that the core is homogeneous. This implies a somewhat unphysical discontinuity of the nematic order parameter $S = \frac{1}{2}\langle 3\cos^2\theta - 1\rangle$ (where θ is the angle between the long axis of the molecule and the director) at the nematic–disclination line interface. The second questionable point about this model is the determination of physically relevant values of the parameters that describe the surface of the core. While the radius of the core of the disclination line can be determined by Landau–de Gennes theory [9] and the anchoring strength at the surface of the core has been roughly estimated by an analysis of stability of the planar radial structure [4], it is rather difficult to say anything decisive about the surface viscosity at a nematic–disclination line interface. (The first measurement of surface viscosity, for MBBA on treated glass, has been reported only recently [12].) On the contrary, the structure of the core of the disclination lines and point defects is consistently described within Landau–de Gennes theory, which predicts a continuous liquid crystalline ordering [13–17].

Obviously, a description of the director fluctuations in the vicinity of disclination lines and point defects, based on the continuous nematic order parameter profile in these regions, should work better than the hypothetical interface approach. In this paper, such a model—free of nematic–core interface and the corresponding elusive parameters (anchoring strength and surface viscosity)—is introduced. The theoretical framework of the proposed approach is described in § 2, and in § 3 its use

*Author for correspondence.

is illustrated by an analysis of the eigenmodes order director fluctuations in the vicinity of a disclination line of strength 1 and the stability of the planar radial structure in a cylindrical cavity is discussed. § 4 summarizes the advantages of the model and concludes the paper.

2. The model

The complete treatment of the fluctuating orientational order in nematic liquid crystals should start with a tensor order parameter and velocity fields [18]. However, such an analysis would be far too involved. If only fluctuations of the average orientation of the molecules (order director fluctuations) are to be studied, the problem is usually simplified by adopting the director description of the liquid crystal [6], which is assumed to be uniaxial, and by adiabatic elimination of the velocity field. The assumption that the nematic ordering is uniaxial is, of course, an approximation, particularly questionable close to surfaces and defects. For example, the structure of the disclination line of strength 1 consists of a uniaxial core with negative nematic order parameter, surrounded by a radially symmetric region where the ordering is biaxial; at large distances from the core the alignment is, of course, uniaxial with positive order parameter [15, 19]. (These details could be taken into account by introducing the elastic free energy of biaxial nematic ordering [20, 21], but such an approach would be quite involved.) In addition, the role of the hydrodynamic degree of freedom in the dynamics of the director fluctuations in confined systems is generally rather complicated (e.g. they can give rise to surface modes) and, in many cases, cannot be taken into account just by rescaling the rotational viscosity as in bulk samples [22, 23]. Nevertheless, the following analysis is based on the above simplifications, because it is concentrated on the director dynamics in the vicinity of the disclination line where the fluctuations are most likely to be predominantly characterized by the decreased degree of order, whereas the effects of the biaxiality of the equilibrium structure and the coupling to the velocity field are expected to be less important.

Within these approximations, the rate of change of the director is proportional to the molecular field

$$\gamma \frac{\partial \mathbf{n}}{\partial t} = \tilde{\mathbf{h}}. \tag{1}$$

where γ is the effective bulk rotational viscosity (or shortly *bulk viscosity*), $\mathbf{n} \equiv \mathbf{n}(\mathbf{r}, t)$ is the director field and $\tilde{\mathbf{h}} \equiv \tilde{\mathbf{h}}(\mathbf{r}, t) = \mathbf{h}(\mathbf{r}, t) - [\mathbf{h}(\mathbf{r}, t) \cdot \mathbf{n}(\mathbf{r}, t)]\mathbf{n}(\mathbf{r}, t)$ with

$$h_\beta = -\frac{\partial f}{\partial n_\beta} + \partial_\alpha \left(\frac{\partial f}{\partial g_{\alpha\beta}} \right), \quad g_{\alpha\beta} \equiv \frac{\partial n_\beta}{\partial r_\alpha} \tag{2}$$

is the molecular field [5]; the subtracted part ensures the orthogonality of $\tilde{\mathbf{h}}$ and $\mathbf{n}$ [1]. If the two divergence elastic terms $K_{13}\nabla \cdot [\mathbf{n}(\nabla \cdot \mathbf{n})]$ and $K_{24}\nabla \cdot [\mathbf{n} \times \nabla \times \mathbf{n} + \mathbf{n}(\nabla \cdot \mathbf{n})]$ are neglected, the free energy density f consists of splay, twist, and bend terms

$$f = \frac{1}{2} [K_{11}(\nabla \cdot \mathbf{n})^2 + K_{22}(\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + K_{33}(\mathbf{n} \times \nabla \times \mathbf{n})^2] \tag{3}$$

In the case of a spatially uniform nematic order parameter, the elastic constants K_{11}, K_{22}, and K_{33} are also homogeneous and $\mathbf{h}$ is given by

$$\begin{aligned} \mathbf{h} = K_{11}\nabla(\nabla \cdot \mathbf{n}) - K_{22}[A\nabla \times \mathbf{n} + \nabla \times (A\mathbf{n})] \\ + K_{33}[\mathbf{B} \times \nabla \times \mathbf{n} + \nabla \times (\mathbf{n} \times \mathbf{B})], \end{aligned} \tag{4}$$

where $A = \mathbf{n} \cdot \nabla \times \mathbf{n}$ and $\mathbf{B} = \mathbf{n} \times \nabla \times \mathbf{n}$ [5].

The proposed model is concerned with order director fluctuations—dynamical distortions of the director field, which are small in amplitude. Its key assumption is that the equilibrium nematic order parameter profile of a nematic structure does not change significantly upon a fluctuation of the director and is thus a well-defined quantity. This premise is based on the facts that (i) the nematic order parameter is defined on a scale which is small compared to most of the wavelengths of the director fluctuations and (ii) the relaxation times of director fluctuations are much longer than the relaxation times of nematic order parameter fluctuations, the difference being related to the Goldstone dispersion of the former [6].

The nematic order parameter, S, enters the dynamical equation (1) through both bulk viscosity and elastic constants: $\gamma \propto S^2$ and $K_{ii} \propto S^2$ to lowest order [24–27]. (However, the expansions of the elastic constants K_{13} and K_{24}, which are not taken into account at present, also contain terms linear in S [28].) As far as γ is concerned, it is only necessary to insert its spatial dependence in equation (1). On the other hand, non-uniform elastic constants give rise to three more terms that, in addition to those defined by equation (4), contribute to the molecular field. The molecular field, induced by an inhomogeneous nematic order parameter, turns out to be given by

$$\mathbf{h}^* = (\nabla K_{11})(\nabla \cdot \mathbf{n}) - (\nabla K_{22}) \times (A\mathbf{n}) + (\nabla K_{33}) \times (\mathbf{n} \times \mathbf{B}), \tag{5}$$

with A and $\mathbf{B}$ as defined above.

In order to analyse the director fluctuations in inhomogeneously ordered nematic liquid crystals within the model presented, the equilibrium order parameter field must be known. It can be determined by using a Landau–de Gennes free energy density expansion, which is in its simplest uniaxial form usually expressed as

$$f = \frac{1}{2} a(T - T^*)S^2 - \frac{1}{3} BS^3 + \frac{1}{4} CS^4 + \frac{1}{2} L(\nabla S)^2, \quad (6)$$

where a, T^*, B, C, and L are temperature-independent material constants [5]. In most cases, the minimization of the free energy requires numerical treatment.

3. Director modes in vicinity of a disclination line

To demonstrate the use of the proposed approach, the behaviour of order director fluctuations in the vicinity of a disclination line of strength 1 is studied. A disclination line of this type occurs, for example, in the centre of the planar radial director field in cylindrical capillaries with homeotropic anchoring at the wall [9]. In cylindrical coordinates, this structure is described by $\mathbf{n}_0 = \mathbf{e}_r$, where $\mathbf{e}_r$ is the radial unit vector. Its equilibrium nematic order parameter profile has been studied within the uniaxial approximation [16]. Close to the centre of the disclination, the nematic order parameter turns out to be proportional to the distance from the centre (r), while at larger distances it saturates at a temperature-dependent bulk value. This behaviour can be approximated rather well by the model profile

$$S(r) = S_0[1 - \exp(-r/b)], \quad (7)$$

where S_0 is the bulk nematic order parameter and b is a parameter, related to the size of the defect. The temperature dependence of b can be estimated by minimizing the free energy of the model planar radial structure (whose nematic order parameter profile is described by equation (7)) with respect to b. For large capillaries, the temperature dependence of b is approximately given by $0{\cdot}83\ [L/a(T_c - T^*)]^{1/2} \cdot |t - 9/8|^{-0{\cdot}44}$, where $T_c = T^* + 2B^2/9aC$ is the critical temperature and $t = (T - T^*)/(T_c - T^*)$ is the reduced temperature; in this scale, $t = 9/8$ corresponds to the superheating temperature. Within the isotropic core model, the core radius (r_0) is given by the nematic correlation length, which is inversely proportional to $(T_c - T)^{1/2}$ [9]; obviously, r_0 is quite close to b.

The eigenmode analysis is carried out within the one elastic constant approximation, i.e. $K_{11} = K_{22} = K_{33} = K$. Up to linear terms, the fluctuating director field is given by $\mathbf{n}(\mathbf{r}, t) = \mathbf{n}_0 + \mathscr{P}(\mathbf{r}, t)\mathbf{e}_\varphi + \mathscr{A}(\mathbf{r}, t)\mathbf{e}_z$ (the cylindrical coordinate system being spanned by $\mathbf{e}_r$, $\mathbf{e}_\varphi$, and $\mathbf{e}_z$), where both planar and axial components are assumed to be small ($|\mathscr{P}(\mathbf{r}, t)|, |\mathscr{A}(\mathbf{r}, t)| \ll 1$). According to the introductory paragraphs, the dynamics of the fluctuating components of the director field are given by

$$\gamma \frac{\partial \mathscr{P}}{\partial t} = K\nabla^2 \mathscr{P} + \frac{\mathrm{d}K}{\mathrm{d}r} \frac{\partial \mathscr{P}}{\partial r}, \quad (8)$$

$$\gamma \frac{\partial \mathscr{A}}{\partial t} = K\left(\nabla^2 \mathscr{A} + \frac{\mathscr{A}}{r^2}\right) + \frac{\mathrm{d}K}{\mathrm{d}r}\left(\frac{\partial \mathscr{A}}{\partial r} - \frac{\mathscr{A}}{r}\right), \quad (9)$$

where $\gamma \equiv \gamma_0 q(r)$, $K \equiv K_0 q(r)$, and $q(r) = [1 - \exp(-r/b)]^2$. γ_0 and K_0 are the values of bulk viscosity and elastic constant for $S = S_0$, i.e. far from the disclination line.

The planar eigenmodes are of the form

$$P(\mathbf{r}, t) = R_P(r) \begin{Bmatrix} \cos m\varphi \\ \sin m\varphi \end{Bmatrix} \begin{Bmatrix} \cos k_3 z \\ \sin k_3 z \end{Bmatrix} \exp(-t/\tau_P), \quad (10)$$

where m is an integer and k_3 is an integer multiple of $2\pi/d$: to ensure the completeness of the eigenmodes, periodic boundary conditions along the capillary of length d are assumed. Both the radial part of the planar eigenmode, $R_P(r)$, and its relaxation time, τ_P, depend on m and k_3. The axial eigenmodes are decomposed in precisely the same manner. The radial parts of the planar and axial eigenmodes, respectively, are determined by

$$qR_P'' + \left(\frac{q}{x} + q'\right) R_P' - \left(q \frac{m^2}{x^2} - q\lambda_P\right) R_P = 0, \quad (11)$$

$$qR_A'' + \left(\frac{q}{x} + q'\right) R_A' - \left(q \frac{m^2 - 1}{x^2} + \frac{q'}{x} - q\lambda_A\right) R_A = 0, \quad (12)$$

where prime denotes $\mathrm{d}/\mathrm{d}x$ with $x \equiv r/R$ (R is the radius of the capillary),

$$\lambda_{P,A} \equiv \left(\frac{\gamma_0}{K_0 \tau_{P,A}} - k_3^2\right) R^2, \quad (13)$$

and $q = [1 - \exp(-x/\beta)]^2$ with $\beta \equiv b/R$. For simplicity, strong anchoring at the wall of the capillary ($x = 1$) is assumed ($R_P(1) = R_A(1) = 0$). The boundary condition at $x = 0$ (the axis of the cylinder) requires that the eigenmodes be finite, i.e. $R_P(0)$, $R_A(0) < \infty$. To examine the behaviour of planar and axial eigenmodes in the core of the disclination line, approximate forms of equations (11) and (12) valid for $x \ll 1$ must be found. Near $x = 0$, which is a regular singular point for both equations,

$$R_P'' + \frac{3}{x} R_P' - \frac{m^2}{x^2} R_P = 0, \quad (14)$$

$$R_A'' + \frac{3}{x} R_A' - \frac{m^2 + 1}{x^2} R_A = 0 \quad (15)$$

with $R_P(x) = x^{-1 \pm (1 + m^2)^{1/2}}$ and $R_A(x) = x^{-1 \pm (2 + m^2)^{1/2}}$. Physically relevant solutions must be finite, therefore

$$R_P(x \to 0) \approx x^{-1 + (1 + m^2)^{1/2}}, \quad (16)$$

$$R_A(x \to 0) \approx x^{-1+(2+m^2)^{1/2}}. \quad (17)$$

With the aid of these results, equations (11) and (12) can be integrated numerically. Some of the radial parts of planar and axial eigenmodes, corresponding to $\beta = b/R = 0{\cdot}05$, are shown in figures 1 and 2, respectively.

Within the above description of the disclination line, its thickness is determined by the temperature-dependent parameter b (which, as noted above, is nearly equal to the nematic correlation length). The question is whether the effective radius of the line (the quantity that corresponds to the radius of the line in the isotropic core model) is exactly equal to b or is it perhaps two or three times larger? In order to establish the relation between the two quantities, the stability of the planar radial structure against the escape along the capillary axis is analysed [4, 11]. The former is unstable if the relaxation rate of the slowest axial eigenmode is negative.

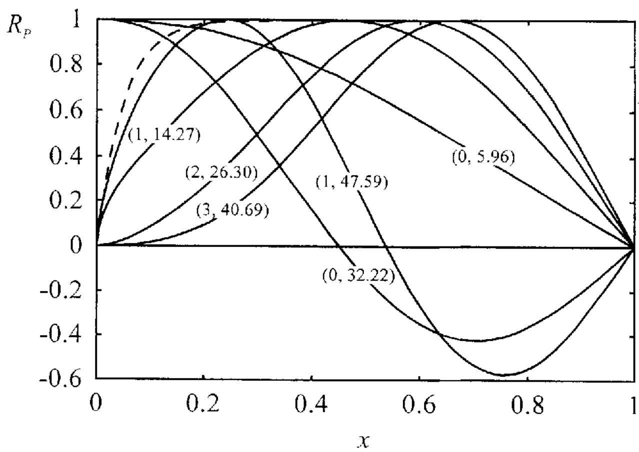

Figure 1. Some of the radial parts of planar eigenmodes in a nematic capillary with $\beta = 0{\cdot}05$ and $K_{11} = K_{22} = K_{33}$. For $m = 0$, the planar modes remain finite right down to the centre of the disclination line; these modes correspond to a spiral distortion of the planar radial structure. For $m > 0$, the modes must vanish at $x = 0$. Each radial part is labelled by (m, λ_P); the nematic order parameter profile $(q = S/S_0)$ is also plotted (dashed line).

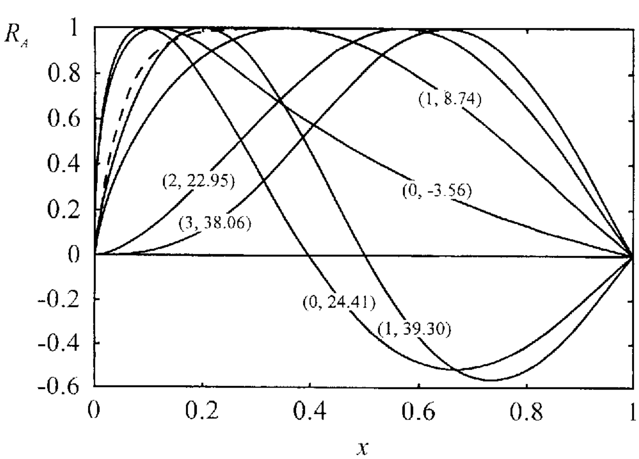

Figure 2. Radial parts of the lowest axial eigenmodes in the capillary, tagged by m and λ_A; as in figure 1, $\beta = 0{\cdot}05$ and $K_{11} = K_{22} = K_{33}$. All axial eigenmodes vanish in the centre of the disclination line. Note that the relaxation rate of the lowest axial eigenmode is negative, which indicates that the planar radial structure is unstable at $\beta = 0{\cdot}05$ and $K_{11} = K_{33}$. Again, the nematic order parameter profile is plotted with the dashed line.

The slowest axial eigenmode is the one with $m = k_3 = 0$ and no radial nodes. In a more elaborate model with anisotropic liquid crystalline elasticity ($K_{11} \neq K_{22} \neq K_{33}$), its radial part, $R_{A,0}$, is determined by

$$a_3 q R''_{A,0} + a_3 \left(\frac{q}{x} + q'\right) R'_{A,0} + \left(\frac{q}{x^2} - \frac{q'}{x} + q\lambda_A\right) R_{A,0} = 0 \quad (18)$$

(where $a_3 \equiv K_{33}/K_{11}$) and the boundary conditions $R_{A,0}(0) < \infty$ and $R_{A,0}(1) = 0$. Close to the centre of the disclination line,

$$R_{A,0}(x \to 0) \approx x^{-1+(1+a_3^{-1})^{1/2}}. \quad (19)$$

The phase diagram is therefore defined in space spanned by β, which depends on the temperature, and a_3, the ratio of bend and splay elastic constant.

In figure 3, the phase diagram resulting from the above stability analysis is compared with predictions of previous calculations, based on the isotropic core model with no anchoring of the nematic director at the disclination line (i.e. the corresponding anchoring strength is set to 0) [4]†, and with the phase diagram obtained by comparison of the free energies of the planar radial structure with a homogeneous isotropic core of the disclination line and the escaped configuration. Qualitatively, all three phase diagrams are the same: the escaped radial structure is stable at small values of the reduced bend elastic constant, a_3, and at large a_3 the planar configuration should be observed. The critical value of a_3 decreases with increasing β and $\rho = r_0/R$, where r_0 is the radius of the disclination line in the isotropic core model. Quantitative discrepancies of the phase diagrams are related to the differences among the models of the nematic order parameter profiles of the planar radial structure on the one hand and the order parameter profile of the escaped radial configuration on the other [4].

According to figure 3, the sole parameter of the described model of the nematic order parameter profile, b, can be regarded as the effective radius of the disclination line. Once this correspondence is established, one

†The phase diagram, of course, does not depend on the value of the surface viscosity [11].

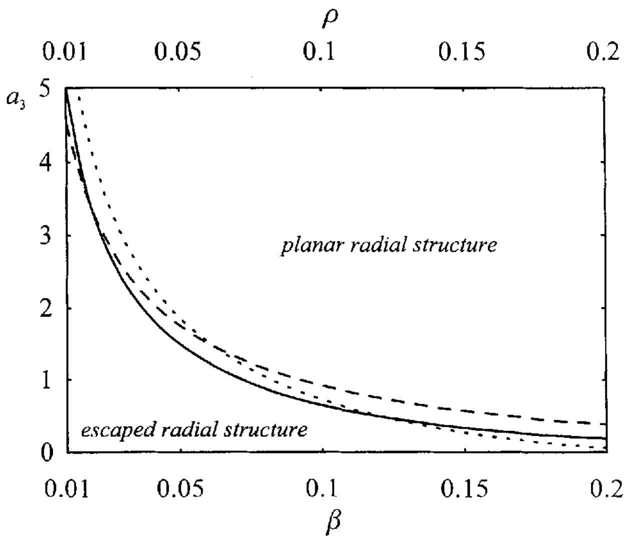

Figure 3. The phase diagram, calculated by eigenmode analyses, based on the proposed description of the disclination line (solid line) and the isotropic core model with core radius r_0 and no anchoring at the surface of the line (dashed line) [11], respectively, and by comparison of the free energies of an escaped and a planar radial structure with a homogeneous disclination line with radius r_0 (dotted line). (The first one is defined in (β, a_3)-space and the other two in a plane spanned by $\rho \equiv r_0/R$ and a_3.) The approaches give quite similar results: for small values of a_3, the escaped configuration is expected, while the planar structure should be stable at large a_3. The only parameter of the proposed model, b, obviously coincides with the common notion of the radius of the disclination line, r_0.

can estimate the surface viscosity needed for the description of the nematic–disclination line interaction within the isotropic core model. This can be accomplished by comparing the shape of the eigenmodes, predicted by the two models. It turns out that the modes, calculated by the isotropic core model [4], are closest to their counterparts from figures 1 and 2 if the surface viscosity is set to 0. This is illustrated by figure 4, where some of the lowest axial modes, determined by the isotropic core model with zero anchoring strength and zero surface viscosity, are compared with the modes from figure 2. The difference between the corresponding modes becomes negligible as m increases and is really significant only for $m = 0$ and 1. For planar modes, the discrepancy between the predictions of the two models is more pronounced than for axial ones, but, again, it is important only for $m < 2$.

The above discussion complements the estimate of the value of the anchoring strength, based on a stability analysis [4]. The results of both analyses suggest that if the disclination line is described by a homogeneous cylinder of isotropic phase, anchoring strength and

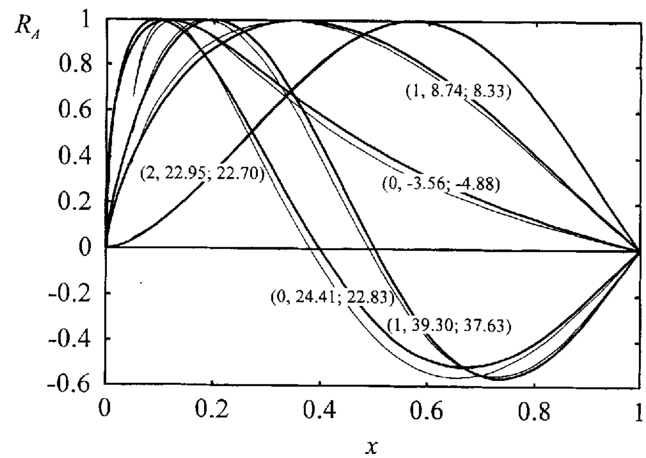

Figure 4. Some of the lowest axial eigenmodes, calculated within the isotropic core model with zero anchoring strength and zero surface viscosity at the nematic–disclination interface (thin line), and their counterparts from figure 2 (thick line). Each pair of eigenmodes is labelled by $(m, \lambda_A; \lambda_{Ai})$, where λ_A and λ_{Ai} are the relaxation rates of the eigenmodes corresponding to the proposed description of the disclination line and the isotropic core model, respectively. The difference between the predictions of both models is largest for $m = 0$; and $m > 2$, they are practically indistinguishable.

surface viscosity at the nematic–disclination line interface should be set to 0.

4. Conclusions

Within the proposed approach, spatially dependent bulk viscosity and elastic constants are introduced in Frank elastic theory of order director fluctuations in nematic liquid crystals. The model is tested by analysing the eigenmodes of fluctuations in a planar radial structure, characterized by a disclination line of strength 1. It seems to offer a sounder and more transparent description of the director dynamics in the vicinity of a disclination line than the isotropic core approach. Its important advantage over the models used so far is the reduced number of parameters needed for the description of the coupling of the nematic director and the disclination line. Within the presented model, this interaction is described only by the coefficients of the Landau–de Gennes expansion (which determine the radius of the core), whereas in the isotropic core model, the anchoring strength and the surface viscosity at the nematic–core interface are needed on top of these material constants.

In micron- and supramicron-size confined systems, the disclination lines and point defects constitute only a minute part of the volume. In such cases, the main role of the concept of inhomogeneous bulk viscosity and elastic constants is to provide boundary conditions for the fluctuating director field at the singularities. But the underlying theory should apply to all nematic systems with inhomogeneous equilibrium order parameter profile, provided the temperature is far enough from critical.

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