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Orientational energy of anisometric particles in liquid-crystalline suspensions

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We obtain a general expression for the orientational energy of an individual anisometric particle suspended in uniform nematic liquid crystals when the main axis of the particle rotates with respect to the nematic director. We show that there is a qualitative and quantitative analogy between the internal and external problems for cylindrical volumes of nematic liquid crystals, and on this basis we obtain an estimate of the orientational energy of a particle of cylindrical (rodlike, needlelike, or ellipsoidal) shape. For an ensemble of such particles we propose a modified form of their orientational energy in the nematic matrix. This orientational energy has the usual second-order term, and additional fourth-order term in the scalar product of the nematic director and the vector which characterizes an average direction of the main axes of the particles. As an example we obtain the expression for the free energy density of ferronematics, i.e., colloidal suspensions of needlelike magnetic particles in nematic liquid crystals. Unlike previous models, the free energy density includes the proposed modified form of the particle orientational energy, and also a contribution describing the surface saddle-splay deformations of the liquid crystal matrix.

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I. INTRODUCTION

Soft condensed matter, which includes such materials as liquid crystals, colloids, foams, gels, polymer melts, and solutions is one of the important areas of modern physics. The presence of internal degrees of freedom and high sensitivity of these materials to external influences leads to a wide variety of observable physical effects in them. Important examples of soft condensed matter are colloidal suspensions of anisometric particles in liquid crystals. The unique physics of these systems is due to the mutual influence of the anisotropic properties of spontaneously ordered liquid-crystalline environment and prolate or oblate particles of the solid phase embedded in it. Therefore, the physical properties of these composite materials are significantly richer than the properties of the constituent components. The collective response of liquid-crystalline suspensions to external fields leads to the existence of many new physical phenomena that are interesting from both fundamental point of view and applied perspectives. Apparently, the first historical examples of liquid-crystalline suspensions are ferronematics [1,2].

Ferronematics (FNs) are magnetic suspensions on the basis of nematic liquid crystals (NLCs). The solid phase of FNs consists of single-domain needlelike ferri- or ferromagnetic particles with length L and diameter $d \sim (L/10)$. Due to the shape anisotropy the particles have a magnetic rigidity at which the magnetic moments of needlelike ferroparticles are always directed along their main axes [3,4]. The length L of the particles and the diameter d are large in comparison with the nematic molecule size a, i.e., the particles represent the mesoscopic objects suspended in a liquid crystal. The volume fraction f of a solid phase is rather small (10^{-7}–10^{-2}), therefore the magnetic impurity in FNs can be considered as an ideal gas of noninteracting magnetic grains. The existence of a small ferroparticle additive does not change the nature of orientational order of a nematic matrix, therefore, FNs having high magnetic susceptibility otherwise behave like usual NLCs.

More than a hundred papers are devoted to theoretical and experimental studies of FNs; their review is given in [5]. Below we discuss the main theoretical investigations devoted to the construction of FN continuum theory, as well as the results of the experiments which influenced the development of theoretical views about the internal structure and behavior of such liquid crystal materials.

The idea of creation of magnetic suspensions was proposed by Brochard and de Gennes in their classical work [1] which initiated the beginning of development of the FN continuum theory. Later FNs were synthesized on a basis of both thermotropic [6–8], and lyotropic [9,10] nematic liquid crystals. The performed experiments [6–10] showed that values of the magnetic fields necessary for reorientation of a liquid crystal matrix of real magnetic suspensions, at least, are two orders of magnitude lower than fields of reorientation of usual nematics. This circumstance expands the use of magnetic liquid crystal materials in applications.

The high magnetic susceptibility of FNs theoretically predicted by Brochard and de Gennes and confirmed experimentally, is caused by an orientational interaction between ferroparticles and a nematic matrix. For magnetized FN in which the magnetic moments of ferroparticles coincide in the direction at each local point of the sample, the natural variables responsible for the orientational order of nematic and magnetic subsystems are the director $\boldsymbol{n}(\boldsymbol{r})$ and magnetization vector $\boldsymbol{M}(\boldsymbol{r})$ averaged at scales much greater than L. Therefore the orientational interaction has to describe the local correlation of spatial distributions of $\boldsymbol{n}$ and $\boldsymbol{M}$. This interaction makes the corresponding contribution to the total free energy of FN which we call "*orientational energy of ferroparticles*" or simply "*orientational energy*."

The detailed theoretical description of orientational interaction of anisometric particles with a nematic matrix is given by Brochard and de Gennes [1]. The analysis of their theory allows us to conclude that the general expression for the

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volume density of orientational energy of ferroparticles can be written as

$$F_{\mathrm{OR}}(\theta) = f\sum_{k=1}^{\infty} A_{2k}\cos^{2k}(\theta) = f\sum_{k=1}^{\infty} A_{2k}(\boldsymbol{m}\cdot\boldsymbol{n})^{2k}, \tag{1}$$

where θ is the angle between the unit vector $\boldsymbol{m} = (\boldsymbol{M}/M)$ of a FN magnetization and the liquid crystal director $\boldsymbol{n}$. Expansion coefficients A_{2k} have the dimensionality of energy volume density ($\mathrm{erg/cm^3}$) and depend on FN material parameters and type of anchoring of nematic molecules at a ferroparticle surface. Let us note that Ref. [1] does not contain expression (1) explicitly; however, it directly follows from the theoretical investigation presented in [1], and this is shown in the main part (Sec. II) of the present paper.

Generally, rigorous calculation of coefficients A_{2k} is not possible since it is connected with the necessity of the detailed theoretical description of a director field in the vicinity of the needlelike particles. Similar problems in colloidal liquid crystal systems arise, for example, at the description of distortions near particles of a spherical shape suspended in a homogeneous nematic matrix. It is well known [11] that those problems lead to a complex nonlinear system of differential equations for the director orientation angles and yet have no exact analytical solutions. At the same time, the use of approximate analytical methods and *Ansätze* based on the topological analysis of orientational distortions and asymptotic behavior of solutions far from spherical particles allows researchers to give quite complete description of possible distributions of a director field [11–13] which are in good agreement with the experiment.

It is also possible to apply the methods of approximate description of orientational distortions in the case of needle-shaped particles. They allow us to estimate the values of coefficients A_{2k}. In particular, Brochard and de Gennes used the approximation of infinitely strong (rigid) anchoring of nematic molecules at particle surfaces which at the time of writing of Ref. [1] was considered as the most probable. Three types of boundary conditions were considered: (I) *longitudinal*, when the easy orientation direction of nematic molecules on the surface of an individual particle is parallel to its main axis; (II) *homeotropic*, when this direction is normal to the particle surface; and (III) *circular*, when the direction of easy orientation is tangential to the particle surface and is normal to the main axis of the particle. Describing orientational distortions for boundary conditions of type (I) Brochard and de Gennes used the electrostatic analogy, and in the case of boundary conditions (II) and (III) they considered needlelike particles as long cylinders and neglected end effects. It was shown that at any of the given types of anchoring the density of orientational energy $F_{\mathrm{OR}}(\theta)$ has a deep minimum when $\theta = 0$. It means that at rigid anchoring in Eq. (1) it is possible to assume $F_{\mathrm{OR}}(\theta = 0) = f\sum_{k=1}^{\infty} A_{2k} \to -\infty$ and consider that $\boldsymbol{m} \parallel \boldsymbol{n}$. Therefore Brochard and de Gennes proposed the continuum model of rigid FNs [1] which implied strict parallelness of the unit magnetization vector $\boldsymbol{m}$ to the liquid crystal director $\boldsymbol{n}$ (so-called Brochard–de Gennes theory). This theory, however, was not valid for real thermotropic suspensions with soft nematic anchoring at surfaces of ferroparticles. In particular, the experiments [6–8] showed convincingly that in the uniform sample of FN the needlelike ferroparticles with soft homeotropic (II) anchoring (and, consequently, the magnetic moments of these ferroparticles, as well) are oriented perpendicular to the unperturbed nematic director. This result was in contradiction with the main postulate of the Brochard and de Gennes theory about the parallelness of $\boldsymbol{m}$ and $\boldsymbol{n}$.

The arisen contradiction between the theory and experiment induced the authors of Refs. [14–16] to study the behavior of magnetic suspensions with soft nematic anchoring at the ferroparticle surfaces. In Refs. [14,15] the influence of soft anchoring on the equilibrium orientation of the individual needlelike particle suspended in uniform nematic sample was considered for the three anchoring types mentioned above [(I), (II), and (III)]. In Ref. [16] these results were generalized for the case of an ensemble of ferroparticles with arbitrary type of anchoring at surfaces, and the continuum model of soft FNs (so-called Burylov-Raikher theory) was proposed. The authors of this theory offered to limit the expansion of orientational energy density (1) only by the quadratic contribution

$$F_{\mathrm{OR},2}(\theta) = A_2 f\cos^2(\theta) = A_2 f(\boldsymbol{m}\cdot\boldsymbol{n})^2. \tag{2}$$

Considering needlelike ferroparticles as cylinders and neglecting end effects by analogy with Ref. [1], the authors of Refs. [14–16] calculated the corresponding coefficient A_2. It appeared that depending on the type of anchoring and material parameters this coefficient can be both less and more than zero. Therefore in the absence of an external magnetic field the particles can be oriented either along or perpendicular to the director. For example, for anchoring (I) the coefficient A_2 is always less than zero, and the particles are oriented along the director. As to anchorings (II) and (III), the sign of this coefficient depends on the value of the dimensionless parameter

$$w = (WR/K), \tag{3}$$

where W is the energy density of nematic anchoring at a surface of a particle, R is the particle radius, and K is the average value of the NLC elastic constants. If w exceeds the certain critical value $w_* \sim 1$, the coefficient $A_2 < 0$, and particles must lie along the director, but if $w < w_*$ then $A_2 > 0$, i.e., the particles must be oriented perpendicular to the director. The estimates given in Refs. [14–16] demonstrated that for real thermotropic FNs [6–8] the parameter $w \ll 1$, therefore the conclusion of the theory about perpendicular orientation of long axes of particles with respect to the director of a nematic matrix makes it possible to explain the results of experiments. As to the theory itself, currently it is widely used [17–31] for the description of orientational effects in FNs induced by magnetic and electric fields.

It should be noted that the authors of the model [16] of soft FNs, limiting the expansion (1) only by quadratic contribution, were guided by the following reasons. First, they assumed that in the absence of a magnetic field there is only one equilibrium position of ferroparticles in magnetized FNs when $\boldsymbol{m} \parallel \boldsymbol{n}$ or $\boldsymbol{m} \perp \boldsymbol{n}$. Secondly, they took into consideration that the quadratic form (2) of the orientational energy of ferroparticles is in a direct analogy with Rapini's heuristic potential [32]. The latter corresponds to the anchoring energy and is widely used for the description of orientational interaction between a nematic and a solid substrate on which the direction of easy orientation of the director is given. In the model [16] of soft FNs a favorable (or unfavorable) axis of the director orientation is represented by a unit magnetization vector along which long axes of ferroparticles are directed at each point of a sample. The rotation of ferroparticles (or the vector $\boldsymbol{m}$), initiated, for example, by an external magnetic field, also involves the rotation of the director. The director $\boldsymbol{n}$ does not strictly follow the direction of $\boldsymbol{m}$ (as in the theory of rigid FNs proposed by Brochard and de Gennes), and is oriented at a certain angle θ which can be found by minimization of total FN free energy. Thus, for the case of soft anchoring, using only the quadratic approximation, the authors of Ref. [16] uncoupled the orientational variables (vector $\boldsymbol{m}$ and director $\boldsymbol{n}$) in the continuum model of FNs.

The theory [16] gives a satisfactory description of available experimental data (see, for example, Refs. [27–29,33,34]), but this theory is not complete because Eq. (2) does not include the next orders of orientational energy expansion in Eq. (1). In the present paper we expand the model [16] and consider the additional fourth-order contribution in Eq. (2):

$$\begin{aligned} F_{\mathrm{OR},4}(\theta) &= f[A_2\cos^2(\theta) + A_4\cos^4(\theta)] \\ &= A_2 f(\boldsymbol{m}\cdot\boldsymbol{n})^2[1-\zeta(\boldsymbol{m}\cdot\boldsymbol{n})^2], \end{aligned} \tag{4}$$

where $\zeta = -(A_4/A_2)$ is the dimensionless phenomenological parameter characterizing the ratio of the fourth- and second-order terms in $(\boldsymbol{m}\cdot\boldsymbol{n})$. The minus sign in Eq. (4) before ζ is chosen by analogy to the corresponding contribution to the surface anchoring energy which corresponds to the interaction of usual NLCs with a solid substrate [35–40]. It is known [39–42] that in general case the anchoring energy density can be presented in the form of infinite series in even degrees of a sine or a cosine of $\tilde{\theta}$, where $\tilde{\theta}$ is the angle of LC director deviation from the easy orientation direction of the director on a substrate. So, this expression formally coincides with Eq. (1). When $\tilde{\theta}$ is small the density of anchoring energy is well described by Rapini form which corresponds only to the quadratic contribution in $\cos\tilde{\theta}$ (or $\sin\tilde{\theta}$). As $\tilde{\theta}$ grows for the correct description of surface effects on the NLC boundary in the anchoring energy density it is also necessary to consider, at least, the fourth order of expansion (see Refs. [35–40]). In the latter case the so-called bistable anchoring characterized not by one, but by two positions of director equilibrium on a cell boundary can take place. It is seen from Fig. 1, where we show the behavior of the function $F_{S,4} = \tilde{A}_2\cos^2\tilde{\theta}(1-\tilde{\zeta}\cos^2\tilde{\theta})$, which corresponds to the anchoring energy of this type. The parameter $\tilde{\zeta}$ can vary from -1 to $+1$, therefore, when $\tilde{A}_2 > 0$ and $0.5 < \tilde{\zeta} \leqslant 1$ the angular function has two local minima $\tilde{\theta} = 0$ and $\pi/2$ [see Fig. 1(a)]. In the absence of some additional orientational distortions initiated, for example, by external fields or the specific geometry of a sample, those minima correspond to two local positions of director equilibrium at the cell boundaries. Therefore the NLC surface energy $F_{S,4}$, unlike Rapini's potential ($\tilde{\zeta} = 0$), is usually known as the modified or bistable anchoring energy [35–37,43–45]. It should be noted that if $\tilde{A}_2 < 0$ and $0.5 < \tilde{\zeta} \leqslant 1$, the energy $F_{S,4}$ allows the tilted orientation of the director at a cell boundary which is observed in the experiments (if a normal or a tangent to this surface is chosen as easy orientation axis) [see Fig. 1(b)].

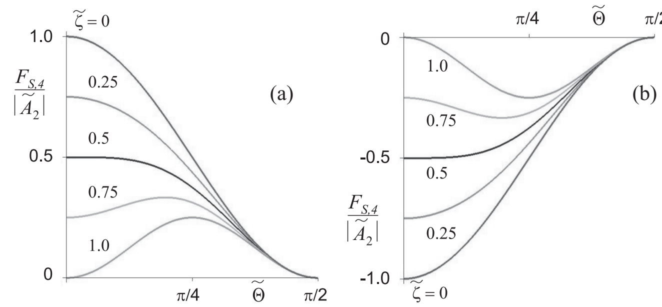

FIG. 1. Profiles of the function $(F_{S,4}/|\tilde{A}_2|)$ which corresponds to the angular part of bistable anchoring energy and angular dependence of orientational energy of ferroparticles (4), with $\tilde{A}_2 > 0$ (a) and $\tilde{A}_2 < 0$ (b) for different values of $\tilde{\zeta}$.

For pure nematic liquid crystals, the use of bistable anchoring energy, of which surface density coincides with Eq. (4) in its angular dependence, is well known [35–40]. Choosing the value of $\tilde{\zeta}$ from theoretical estimates or the analysis of experimental data, many authors gave a correct explanation of different orientational transitions in pure NLCs that would be impossible taking into account only Rapini's contribution in the anchoring energy.

In the physics of ferronematics, the orientational effects with the bistable anchoring at cell walls are discussed in Refs. [43–45], and the fourth-order contribution in $(\boldsymbol{m}\cdot\boldsymbol{n})$ in the orientational energy of ferroparticles with the homeotropic (II) type of anchoring was already studied by Baldin and Zakhlevnykh in Ref. [46]. However, in Ref. [46] the interpretation of this contribution is based only on the analogy presented above between the ferroparticle orientational energy and bistable surface energy of NLCs. In the present paper we show a rigorous derivation of Eq. (1) for the orientational energy from which the necessity of fourth-order contribution (and in the general case of higher orders) in $(\boldsymbol{m}\cdot\boldsymbol{n})$ follows. In addition, we generalize expression (4) for the case of arbitrary anchoring of the director at the surfaces of particles and take into consideration the saddle-splay surface elastic modulus K_{24} of the NLC, which has not yet been taken into account for describing ferronematics behavior.

Analyzing preliminarily the modified potential (4) of ferroparticles orientational energy, we emphasize its fundamental difference from the form of Ref. [16] which is quadratic in $(\boldsymbol{m}\cdot\boldsymbol{n})$. The essence of this difference is that in the absence of a magnetic field the modified potential (4) allows the tilted orientation of particles with respect to the NLC director or the existence of two equilibrium positions of ferroparticles: parallel and perpendicularly to the director. Such tilted or bistable orientation can take place if ζ exceeds the value $\zeta_{\mathrm{BS}} = 0.5$, which is the lower bound of the appearance of these orientational states. From the qualitative point of view, the possibility of the existence of two equilibrium positions is confirmed, for example, by the results of experiments [47,48] on observation of the orientational behavior of nonmagnetic rodlike particles in the homogeneous nematic sample. Let us consider briefly the results of these experiments.

In Ref. [47] the behavior of SiC particles in nematic solvents of various types [MBBA (N-(4-methoxybenzylidene)-4-butylaniline), 8CB (4-octyl-4′-cyanobiphenyl), and NLC mixture E7] was investigated. The particles were rodlike, their average length L was $19 \pm 10\ \mu$m, and their aspect ratio $L : d$ was constant 200:1. Optical experiments [47] showed that depending on a choice of LC solvent and on the existence (or absence) of surfactant on the particles, the latter were oriented either parallel or perpendicular to the nematic director. However, in two cases [when liquid crystals E7 and MBBA were used as solvents and the surfactant prescribed the homeotropic (II) type of anchoring] the bimodal distribution of orientations was observed: A majority of particles was perpendicular to the director and a smaller, but significant, minority of them were along the director. Additionally, all the experiments [47] showed that local orientational distortions of the director field near particles did not contain point and line defects.

In experiments [48] the nematic liquid crystal 5CB (4-n-pentyl-4-cyanobiphenyl) was used. The average length of glass microrods suspended in a nematic matrix was $L \sim 8\mu$m, the diameter was $d = (1.49 \pm 0.07)\mu$m, and the homeotropic type of anchoring was prescribed at particles surface. Optical observations [48], as well as [47], show the bimodal distribution of orientations: more than half of the particles were near $\theta = 0$, i.e., along the director, and the rest of the particles were oriented at various angles from the interval $\pi/3 \leqslant \theta \leqslant \pi/2$. However, unlike [47] in those experiments the particles were surrounded by systems of point and line defects, and in order to make the particles change their state from $\pi/3 \leqslant \theta \leqslant \pi/2$ to $\theta = 0$ it was necessary to overcome the certain energy barrier connected with reorganization of the systems of orientational defects. Let us also note that in some cases the particles, and couples of particles which stuck together by side surfaces, showed in experiments [48] their tilted orientation with respect to the NLC director. It is connected with asymmetrical arrangement of the defects observed at the ends of the studied objects.

For a theoretical interpretation of the obtained experimental results [47,48] and drawing a qualitative analogy with ferronematics we assume that magnetic moments of particles are directed along their main axes. The intrinsic magnetic field of the particles is sufficiently small so it is not able to affect the distribution of the director even in the immediate vicinity of the particles [1]. Therefore, the existence of magnetic moments of particles does not change the structure of the observed local deformations of the director field [47] near their surfaces as well as the configuration [48] of orientational defects. The next step of the theoretical approach is the continuum description of a suspension of similar particles in a liquid crystal. It means that the deformed state of the director field which was observed in experiments, must be averaged at distances much greater than L. For averaging it is possible to use the so-called method of deformation coats [49,50], limiting local orientational distortions near particles. The director field distribution obtained after the averaging will be homogeneous, without any local deformations or point and line defects. Then, for describing the bimodal [47,48] orientational behavior of particles in the suspension we can use the modified potential (4) with $\zeta > \zeta_{\mathrm{BS}}$.

Assuming that in the absence of an external magnetic field the ferroparticles can have two orientational minima or tilted orientation, we, however, do not set a goal of developing the theory of FNs of precisely such magnetic suspensions. Similar systems require extra measures for maintaining locally homogeneous magnetization of a sample typical for FNs. Otherwise the applied magnetic field will inevitably initiate complex spatially nonuniform structures of the director field that can become a subject of independent theoretical research.

In the present paper we consider ferronematics for which in the modified orientational energy (4) the parameter $-1 \leqslant \zeta < \zeta_{\mathrm{BS}}$ and ferroparticles in the absence of an external magnetic field have one equilibrium position. At that we take into account that in real FNs the transverse dimension of particles is significantly less than in the experiments [48] (see, for example, Refs. [6–8]). Therefore we expect (see Sec. III) that systems of point and line defects which surrounded massive particles in Ref. [48], will be purely fictitious in real FNs, i.e. they will be inside ferroparticles. This localization of defects will facilitate their internal reorganization when particles rotate in a nematic matrix that will cause ζ to decrease. This partially confirms comparison of experimental results [47] and [48] given above. In Ref. [47] the diameter of particles was an order of magnitude less than in Ref. [48], the defects near particles were not observed, and bimodal distribution of orientations was much less expressed (in any case it was not observed in all the nematic solvents and could be seen only after special treatment of particles by surfactant).

The structure of the paper is as follows. In Sec. II we derive Eq. (1) for the general form of orientational energy. In Sec. III we discuss the estimations of coefficients A_{2k} in models of rigid [1] and soft [16] FNs, show the limits of validity of these theories, and represent the final form of the modified orientational energy of particles. To clarify the theory as completely as possible we used here one more analogy. This is the analogy between external problems [1,14–16] for the needlelike particles suspended in a nematic matrix, and internal problems [51–54] on determination of equilibrium configurations of the director in cylindrical capillaries. In Sec. IV we present the modified free energy of FNs. The conclusions are given in Sec. V.

II. GENERAL FORM OF PARTICLES ORIENTATIONAL ENERGY

Let us consider in more detail the problem of orientational interaction of magnetic and liquid crystal subsystems of FNs. As the concentration of particles in a suspension is rather small, ferroparticles make additive contributions to this interaction in the density of FN free energy. Therefore the problem stated above is reduced to a description of the orientational behavior of an individual needlelike particle in the nematic matrix with the director $\boldsymbol{n}_0$ set on the infinity (when $r \gg L$).

Brochard and de Gennes in Ref. [1] studied the described problem in a more general case—for a particle of arbitrary shape. They used one-constant approximation, assuming that all elastic volume constants of nematic (i.e., splay K_{11}, twist K_{22}, and bend K_{33}) are equal: $K_{11} = K_{22} = K_{33} = K$. In addition, the authors of Ref. [1] consider that there are rigid boundary conditions for the NLC director on a particle surface. It was shown that at long distances from a particle the director field can be presented as $\boldsymbol{n} = \boldsymbol{n}_0 + \delta\boldsymbol{n}_\perp$, where the small perturbation $\delta\boldsymbol{n}_\perp$ is oriented in the plane that is normal to $\boldsymbol{n}_0$, i.e., $\delta\boldsymbol{n}_\perp \perp \boldsymbol{n}_0$. The equilibrium equation for transverse disturbance $\delta\boldsymbol{n}_\perp$ is Laplace's equation for which the solution can be presented in the form of multipole expansion. This solution is well known and is used by many authors in descriptions of orientational nematic distortions induced by particles of various shapes [11,50,55,56]. It is also known [1,16,50,57,58] that the first—Coulomb ($\sim 1/r$)—contribution to the expansion $\delta\boldsymbol{n}_\perp$ arises only when there is torque of external forces acting on the particle. We should emphasize that this contribution plays the main role in the orientational behavior of magnetic particles in FNs because the rotation of ferroparticles relative to the NLC director can be initiated by an external magnetic field.

Now let us return to the results of Brochard and de Gennes. Further consideration in Ref. [1] was restricted by particles having an axis of cylindrical symmetry (labeled by a unit vector $\boldsymbol{u}$ which in the case of the needlelike particle coincides with the direction of its long axis). Integrating the Coulomb contribution in $\delta\boldsymbol{n}_\perp$ over the NLC volume, Brochard and de Gennes showed that changes of orientational energy of an individual particle at rotation of its axis $\boldsymbol{u}$ by a small angle $\delta\theta$ relative to the director $\boldsymbol{n}_0$ is expressed as follows [1]:

$$\delta\mathcal{F}'_{\mathrm{OR}} = 4\pi K l(\cos\theta)\sin\theta\,\delta\theta, \tag{5}$$

where the prime in this expression means that this is the energy of an individual particle; the distortion amplitude $l(\cos\theta) = \sum_{k=1}^{\infty} l_{2k-1}\cos^{2k-1}\theta$ has the dimension of length, and is an odd function of a cosine of the angle θ between unit vectors $\boldsymbol{u}$ and $\boldsymbol{n}_0$. Taking into account Eq. (5), the torque acting on a nematic from an individual particle is defined as

$$\boldsymbol{\Gamma}' = 4\pi K l(\cos\theta)\boldsymbol{n}_0 \times \boldsymbol{u}. \tag{6}$$

One can see that the torque Γ' is equal to zero, at least in two cases when the particle axis $\boldsymbol{u}$ is parallel or perpendicular to the director $\boldsymbol{n}_0$. Therefore the specified positions of a particle correspond to the equilibrium conditions of the latter in the homogeneous nematic sample. The question of stability of each of those states depends on the function $l(\cos\theta)$.

Equations (5) and (6) are one of the main results used by Brochard and de Gennes in the construction of a continuum model of FNs with rigid anchoring of the director at particles. Similar results were obtained in Ref. [16], where the theory was developed for soft ferronematics. In particular, the authors of Ref. [16] have shown that the general form of Eqs. (5) and (6) does not change under the soft boundary conditions at a particle surface, but the values of an expansion coefficient of $l(\cos\theta)$ do change. Thus, Eqs. (5) and (6) are common for particles with rigid and soft anchoring.

Now let us show how Eq. (1) for the ferroparticles orientational energy in FNs naturally follows from Eq. (5). For an individual particle the general form of the orientational energy $\mathcal{F}'_{\mathrm{OR}}$ can be obtained by integrating Eq. (5) with respect to the angle θ; the result has the following form:

$$\mathcal{F}'_{\mathrm{OR}} = \mathcal{F}'_\perp - 2\pi K\sum_{k=1}^{\infty}\frac{l_{2k-1}}{k}\cos^{2k}\theta, \tag{7}$$

where the integration constant $\mathcal{F}'_\perp$ corresponds to the distortion energy when the axis $\boldsymbol{u}$ of the particle is oriented perpendicular to the unperturbed director, i.e., when $\boldsymbol{u}\perp\boldsymbol{n}_0$ and $\theta = \pi/2$. If $\theta = 0$ then Eq. (7) is equal to $\mathcal{F}'_\parallel$ with $\boldsymbol{u}$ oriented parallel to $\boldsymbol{n}_0$. This allows us to express the sum of expansion coefficients in Eq. (7) through $\mathcal{F}'_\perp$ and $\mathcal{F}'_\parallel$:

$$2\pi K\sum_{k=1}^{\infty}\frac{l_{2k-1}}{k} = \mathcal{F}'_\perp - \mathcal{F}'_\parallel. \tag{8}$$

We will use this relation in further consideration.

Going to the ensemble of needlelike ferroparticles, we multiply Eq. (7) by the local concentration c of a solid phase, here $c = (f/v)$, where v is the volume of an individual particle. Let us relate long axes $\boldsymbol{u}_i$ of ferroparticles to unit vectors $\boldsymbol{\mu}_i$ of their magnetic moments which in magnetized FNs are oriented in one direction in each small volume of a sample. Thereafter we average the obtained relation at distances much greater than the length of particles L. Then θ corresponds to the angle between the local directions of $\boldsymbol{n}$ and the unit magnetization vector $\boldsymbol{m}$. In fact, after averaging at scales $r \gg L$, the fixed value $\boldsymbol{n}_0$ far from each individual particle can be replaced with a local variable $\boldsymbol{n}$, and the average value $\langle\boldsymbol{\mu}_i\rangle$ can be substituted by the variable $\boldsymbol{m}$. As a result, for the ferroparticle orientational energy $F_{\mathrm{OR}} = \langle f\mathcal{F}'_{\mathrm{OR}}/v\rangle$, which is the function of θ, we obtain Eq. (1) with the coefficients

$$A_{2k} = -\frac{2\pi K}{v}\frac{l_{2k-1}}{k} \tag{9}$$

and the sum [see Eqs. (8) and (9)]

$$\sum_{k=1}^{\infty} A_{2k} = \frac{\mathcal{F}'_\parallel - \mathcal{F}'_\perp}{v}. \tag{10}$$

It should be noted that Eqs. (9) and (10) show the dimension of the coefficients A_{2k} which we introduced above in the Introduction. This is the dimension of volume energy density.

III. ESTIMATION OF EXPANSION COEFFICIENTS OF ORIENTATIONAL ENERGY

Now let us consider methods of determining the coefficients A_{2k} in the models [1] and [16] for rigid and soft FNs, and also discuss the applicability limits of the theories. As can be seen from Eq. (10), the key moment in estimation of these coefficients is the calculation of free energy of distortions induced by an individual particle in the uniform nematic sample at the parallel and perpendicular orientations of its long axis $\boldsymbol{u}$ with respect to the unperturbed director $\boldsymbol{n}_0$. Unlike the case of arbitrary orientation of $\boldsymbol{u}$ and $\boldsymbol{n}_0$, such problems can have an exact solution [1,14,15] if we consider needlelike particles as long cylinders and neglect end effects.

A. Analogy between internal and external problems for cylindrical volumes

For definiteness, we assume that the homeotropic (II) type of nematic anchoring at a side surface of a particle is given. Let us consider the results of Refs. [1,14,15] on calculation of the energies $\mathcal{F}'_\parallel$ and $\mathcal{F}'_\perp$ for this type of anchoring. We will not follow the strict calculations shown in the mentioned papers, but use another approach. It consists in carrying out a qualitative and quantitative analogy between internal and external problems for cylindrical volumes. This allows us, first, to present the director field distortions appearing near a

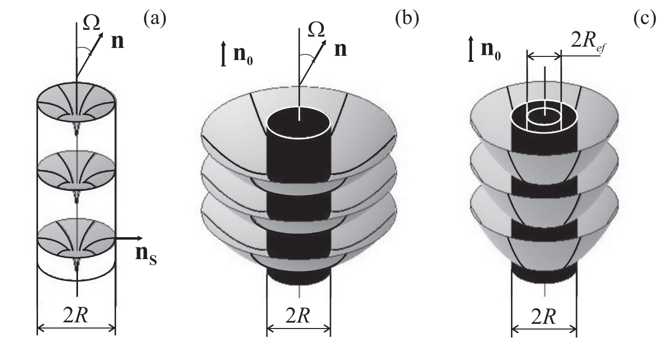

FIG. 2. Qualitative analogy between internal and external problems for cylindrical volumes of NLC: (a) ER structure in cylindrical capillary; (b) director field near a particle with rigid homeotropic anchoring for $\boldsymbol{u} \parallel \boldsymbol{n}_0$, obtained from the ER structure with the use of transformation (11); (c) distribution of the director on going from rigid to soft anchoring at a particle surface, obtained with use of transformation $R \to R_{\text{ef}} = p_{\parallel} R$ [see Eq. (14)].

particle more clearly. Second, this also allows us to show the localization of defects which correspond to the orientational distortions, and the transformation of the defect system upon transition from rigid to soft anchoring. Third, this allows us to specify some of the results of Refs. [1,14,15], in particular, concerning calculations of the energy $\mathcal{F}'_{\parallel}$ at parallel orientation of $\boldsymbol{u}$ and $\boldsymbol{n}_0$.

The qualitative analogy between distributions of a director field for internal and external cylindrical volumes of NLC is shown in Figs. 2 and 3. The well-known escaped radial (ER) structure [51–54] in a cylindrical capillary, presented in Fig. 2(a), upon transformation of inverse-radius vectors

$$r = \frac{R^2}{r'} \tag{11}$$

of the cylindrical coordinate system passes to a configuration which for the variable r' corresponds to a particle suspended in a nematic with $\boldsymbol{u} \parallel \boldsymbol{n}_0$ [see Fig. 2(b)]. In fact, when transforming with Eq. (11) the capillary wall with the coordinate $r = R$ corresponds to the particle surface $r' = R$, and the capillary axis $r = 0$ corresponds to the infinitely remote point $r' = \infty$. Therefore the capillary surface goes into particle surface, the type of anchoring does not change and remains homeotropic, uniform orientation of the director on the capillary axis characterizing the ER structure turns into the uniform distribution of the director field far from a particle, and orientational deformations of the director from the wall to the axis of a capillary are smoothly distributed all over the NLC volume from the particle surface to an infinitely remote point.

The transformation (11) can be used also for the description of orientational distortions near a particle with $\boldsymbol{u} \perp \boldsymbol{n}_0$ [59,60]. In this case, an initial nematic configuration is the so-called planar-polar (PP) structure [52–54] in a cylindrical capillary [see Fig. 3(a)]. As shown in Ref. [54], the orientational distortion corresponding to this structure, in the plane (r,φ) is described by the system of two disclination lines. For rigid anchoring the disclination rest on the capillary surface and have the identical topological charges $q_1 = q_2 = 1$. Let us trace how the charges of these linear defects can change after the transformation (11). Let us surround the points of the plane (r,φ) through which the disclination lines pass, by small contours and set the direction of a circuit of the contours. It is obvious that after transformation (11) the directions of the circuit of disclinations change to the opposite. Therefore, for a particle [Fig. 3(b)], the images of line defects of the PP configuration are the disclination lines with charges $q'_1 = q'_2 = -1$. In addition, one more disclination with the charge $q'_3 = 2$ appears on the particle axis. This disclination is the image of the line defect $q_3 = -2$ spread in the case of the PP structure at the infinity of the plane (r,φ). For better understanding of this question, it is possible to consider the description of disclination lines by means of the functions of complex variables as in Refs. [54,59–62]; then the position of the defect $q_3 = -2$ corresponds to the infinitely remote point of the Riemann sphere. Thus, the distortion induced by the cylindrical particle suspended in the uniform nematic bulk at the perpendicular orientation of its long axis with respect to the unperturbed director, can be described by the system of disclination lines with charges $\{-1,2,-1\}$[see Fig. 3(b)]. The total charge of the defect system is equal to zero. From the topological point of view it means that the system of defects does not induce orientational distortions away from the area of its localization. This circumstance once again confirms the fact that the particle fits into the uniform distribution of the director field.

Considering a quantitative analogy between internal and external problems it is necessary to discuss how transformation (11) affects a type of angular dependence which describes the director field, and also how it affects the changes of the free energy of a system. For two analogies shown in Figs. 2 and 3, this study presents two independent problems, therefore their solution is shown in Appendices A and B, respectively. Below we give the general conclusions according to the results of the performed research.

(1) *For the orientational configurations presented in* Fig. 2: Distortion induced by the particle with $\boldsymbol{u} \parallel \boldsymbol{n}_0$, and the ER structure in a cylindrical capillary are energetically equivalent when the surface elastic modulus K_{24} is formally replaced by the combination $(2K_{11} - K_{24})$ in the relations for the energies of these configurations. The dependence of the angle Ω on the radial coordinate for the external problem can be obtained from

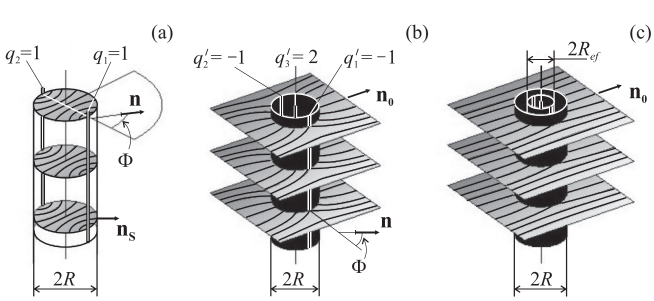

FIG. 3. Qualitative analogy between internal and external problems for cylindrical volumes of NLC: (a) PP configuration in cylindrical capillary; (b) director field near a particle with rigid homeotropic anchoring for $\boldsymbol{u} \perp \boldsymbol{n}_0$, obtained from the PP structure with use the of transformation (11); (c) distribution of the director on going from rigid to soft anchoring at a particle surface, obtained with the use of transformation $R \to R_{\text{ef}} = p_{\perp} R$ [see Eq. (14)].

the known solution for the internal problem (see Refs. [52–54]) with the use of two transformations $r \to (R^2/r')$ and $K_{24} \to (2K_{11} - K_{24})$.

(2) *For the orientational configurations presented in* Fig. 3: The distortion induced by the particle at $\boldsymbol{u}\perp\boldsymbol{n}_0$ and the PP structure in a cylindrical capillary are energetically completely equivalent in the so-called two-constant approximation $K_{11} = K_{33} = \bar{K}$. In the same approximation the dependencies of the angle Φ of the director orientation for the internal and external problems are in a one-to-one correspondence when transformation $r \to (R^2/r')$ is used.

The value $\bar{K}$ in the two-constant approximation is the average value of elastic constants of splay K_{11} and bend K_{33}, i.e., $\bar{K} = (K_{11} + K_{33})/2$, where an overbar is introduced in order to distinguish the two-constant approximation from the one-constant one. The latter is connected with the fact that in real NLC the twist modulus K_{22} is as a rule, always less than elastic constants K_{11} and K_{33}, which are comparable in magnitude. Therefore the two-constant approximation much more corresponds to a real situation than the one-constant one. Additionally, for the homeotropic anchoring, all orientational configurations presented in Figs. 2 and 3 do not depend on the twist modulus K_{22}, therefore the one-constant approximation used in solutions of these problems (as, for example, in Ref. [1]), actually coincides with the two-constant one, but the latter is more accurate.

Further analysis of the results obtained in Appendices A and B for external problems can be split into three stages. At the first stage, we consider these results in approximations which correspond to the model [1] of rigid FNs, and we also give estimates of the applicability limits for this theory. The second stage includes the same consideration for the model [16] of soft FNs. At the third stage we discuss the influence of the saddle-splay surface elastic modulus K_{24} (which was not taken into account in Refs. [1,16]) on the estimates of expansion coefficients A_{2k} from Eq. (1) in existing models of rigid and soft FNs, and we also present a final form of Eq. (4) for the modified orientational energy of ferroparticles.

B. Rigid anchoring

The results of Brochard and de Gennes [1] on calculation of energies $\mathcal{F}'_\parallel$ and $\mathcal{F}'_\perp$ at rigid homeotropic anchoring of the director at a particle surface can be obtained from Eqs. (A15) and (B4) using the approximations $\{W = \infty, K_{11} = K_{33} = \bar{K}, K_{24} = 0\}$; hence,

$$\mathcal{F}'_\parallel = \pi \bar{K} L, \tag{12}$$

$$\mathcal{F}'_\perp = \pi \bar{K} L \ \ln \frac{R}{2r_d}, \tag{13}$$

where r_d is the radius of the disclination core with the order of size a of NLC molecules. Let us remind one that at rigid anchoring and $\boldsymbol{u}\perp\boldsymbol{n}_0$ the disclination lines with the charges $q'_1 = q'_2 = -1$ are on the particle surface [see Fig. 3(b)]. Due to the fact that the radius of the disclination core is $r_d \sim a \ll R$, from Eqs. (12) and (13) we have $\mathcal{F}'_\parallel < \mathcal{F}'_\perp$, therefore the particle has to be oriented parallel to the director.

Let us estimate the value of $\Delta\mathcal{F}' = \mathcal{F}'_\parallel - \mathcal{F}'_\perp \sim -\pi \bar{K} L$ and the sum of the expansion coefficients of orientational energy [see Eq. (10)]. Accepting for these estimates $\bar{K} \sim 5 \times 10^{-7}$ dyne and $L \sim 0.5\mu$m [6–8], we obtain $|\Delta\mathcal{F}'| \sim 2 \times 10^3 k_B T$, i.e., the equilibrium state $\boldsymbol{u} \parallel \boldsymbol{n}_0$ is characterized by a deep minimum of the orientational energy, and the value $|\Delta\mathcal{F}'|$ considerably exceeds the energy of thermal fluctuations. Therefore, the thermal motion cannot disturb the equilibrium state $\boldsymbol{u} \parallel \boldsymbol{n}_0$ of a particle. Supposing a particle as a cylinder, we obtain a sufficiently large negative value $\sum_{k=1}^{\infty} A_{2k} \sim -(\pi \bar{K} L/v) \sim -10^5$ erg/cm^3 for the sum of the expansion coefficients of orientational energy, that allows us to assume $\sum_{k=1}^{\infty} A_{2k} \approx -\infty$. This means that in the case of the ensemble of needlelike ferroparticles in each small volume ($\Delta V \gg L^3$) of magnetic suspension the direction of a unit vector $\boldsymbol{m}$ of FN magnetization is always parallel to the liquid crystal director $\boldsymbol{n}$.

However, this conclusion of the theory [1] may not be true for any values of an external magnetic field $\boldsymbol{H}$. Indeed, the external field initiates the rotation of ferroparticles which for each individual particle is characterized by Zeeman's energy $\mathcal{F}'_Z = -M_S v(\boldsymbol{\mu}\boldsymbol{H}) \sim -M_S v H$, where M_S is the saturation magnetization of the particle material. In order for the local directions $\boldsymbol{m}$ and $\boldsymbol{n}$ remain to parallel, the condition $|\Delta\mathcal{F}'| \gg |\mathcal{F}'_Z|$ needs to hold true. This imposes constraints on the value of the external magnetic field $H \ll H_* = (\pi \bar{K} L/M_S v)$ that with $M_S \approx 5 \times 10^2$G gives $H \ll H_* \approx 160$ Oe.

The estimations given above for the homeotropic (II) type of nematic anchoring at ferroparticles also remain correct for other types of anchoring discussed in Ref. [1] which correspond to the rigid longitudinal (I) and circular (III) boundary conditions. Thus, the theory [1] is true for FNs with rigid anchoring of the director at particles in magnetic fields of about several tens oersted.

C. Soft anchoring

Now let us turn to the results of the model [16] corresponding to soft FNs with finite values of energy W of nematic anchoring at the ferroparticle surfaces [see Eq. (A3)]. As shown in Appendices A and B, a change of anchoring character from rigid to soft from the physical point of view means a reduction of the effective particle radius $R_{\mathrm{ef}} = (pR)$, where the compression coefficient p depends on the orientation of the latter ($\boldsymbol{u} \parallel \boldsymbol{n}_0$ or $\boldsymbol{u}\perp\boldsymbol{n}_0$) and takes values from the interval $0 \leqslant p \leqslant 1$ [see Figs. 2(c) and 3(c)]. Under transformation $R \to R_{\mathrm{ef}}$, the anchoring is rigid at the fictitious particle surface $r' = R_{\mathrm{ef}}$, while on the true surface $r' = R$ the boundary conditions corresponding to the soft anchoring are satisfied. In Refs. [14,15] (which were written before [16] and devoted to solution of these problems), for defining the particle equilibrium position the authors used the approximations $\{K_{11} = K_{33} = \bar{K}, K_{24} = 0\}$. In this case from Eqs. (A21) and (B5) it follows that the compression coefficient p is the function of the dimensionless parameter $\bar{w} = (WR/\bar{K})$ which for the homeotropic anchoring (two-constant approximation) is analogous to the parameter w from Eq. (3); the result takes the following form:

$$p = \begin{cases} p_\parallel = \sqrt{\frac{\bar{w}}{2+\bar{w}}} & \text{for} \quad \boldsymbol{u} \parallel \boldsymbol{n}_0 \\ p_\perp = \sqrt{\frac{\sqrt{4+\bar{w}^2}-2}{\bar{w}}} & \text{for} \quad \boldsymbol{u}\perp\boldsymbol{n}_0. \end{cases} \tag{14}$$

The energies $\mathcal{F}'_\parallel$ and $\mathcal{F}'_\perp$ with an accuracy up to the multiplier $(\pi \bar{K} L)$ are also functions of the parameter $\bar{w}$:

$$\mathcal{F}'_\parallel = \pi \bar{K} L \frac{\bar{w}}{1+\bar{w}}, \tag{15}$$

$$\mathcal{F}'_\perp = \pi \bar{K} L \left[1 + \frac{\bar{w}}{2} - \frac{\sqrt{4+\bar{w}^2}}{2} - \ln \frac{4(\sqrt{4+\bar{w}^2}-2)}{\bar{w}} \right]. \tag{16}$$

These expressions are obtained from Eqs. (A15) and (B4) at $K_{11} = K_{33} = \bar{K}$ and $K_{24} = 0$. They correspond to the results of Ref. [15] where it is shown that the equality $\mathcal{F}'_\parallel = \mathcal{F}'_\perp$ takes place at $\bar{w} = \bar{w}_* = 1.396$. If $\bar{w} < \bar{w}_*$, then $\mathcal{F}'_\parallel > \mathcal{F}'_\perp$, and the particle has to be oriented perpendicular to the director. For $\bar{w} > \bar{w}_*$ we have $\mathcal{F}'_\parallel < \mathcal{F}'_\perp$, so the particle has to be along the director.

In the model [16] it is suggested to limit the general expansion of orientational energy from Eq. (1) only by the quadratic term, then from Eq. (10) it follows that the coefficient A_2 is determined by the expression

$$A_2 = \frac{\mathcal{F}'_\parallel - \mathcal{F}'_\perp}{v}, \tag{17}$$

where Eqs. (15) and (16) correspond to the energies $\mathcal{F}'_\parallel$ and $\mathcal{F}'_\perp$. However, such form of the coefficient A_2 is quite cumbersome and inconvenient for practical use. In addition, in the other types of anchoring, in particular, longitudinal (I) and circular (III), the expressions for $\mathcal{F}'_\parallel$ and $\mathcal{F}'_\perp$ differ from Eqs. (15) and (16). It does not allow us to write down the rigorous expression for A_2 generalized for the anchorings (I)–(III).

The authors of Ref. [16] offered the following way out from this predicament. It was shown that for real FNs the parameter $\bar{w}$ in the case of homeotropic anchoring and the generalized parameter w in the case of anchorings (I)–(III) take values much less than unity. Therefore the authors of [16] actually used the series expansions of $\mathcal{F}'_\parallel$ and $\mathcal{F}'_\perp$ in those small parameters. For Eqs. (15) and (16) such expansions take the form

$$\begin{aligned} \mathcal{F}'_\parallel &\approx \pi \bar{K} L(\bar{w} - \bar{w}^2 + \bar{w}^3 + \cdots) \\ &= \pi R W L(1 - \bar{w} + \bar{w}^2 + \cdots), \\ \mathcal{F}'_\perp &\approx \pi \bar{K} L\left(\frac{\bar{w}}{2} - \frac{\bar{w}^2}{16} + \frac{\bar{w}^4}{512} + \cdots\right) \\ &= \frac{\pi R W L}{2}\left(1 - \frac{\bar{w}}{8} + \frac{\bar{w}^3}{256} + \cdots\right). \end{aligned} \tag{18}$$

When calculating A_2 in Ref. [16] they took into account only the first orders of expansion; then for cylindrical particles with the homeotropic anchoring from Eqs. (17) and (18) we have

$$A_2 \cong \frac{\pi \bar{K} L \bar{w}}{2v} = \frac{W}{d}. \tag{19}$$

As can be seen from Eqs. (2), (18), and (19), in the first order in $\bar{w}$, the particle orientational energy represents only a surface energy. And moreover, the latter one corresponds to the situation when the director near a particle is not distorted and has the direction $\boldsymbol{n}_0$. This conclusion allows one to generalize Eq. (19) for the case of an arbitrary type of anchoring which is defined by the fixed direction $\boldsymbol{n}_S$ of the director easy orientation at a particle surface. After this generalization for A_2 the following relation was obtained [16]:

$$A_2 \cong -2\frac{W}{d} P_2(\cos\alpha), \tag{20}$$

which in the case of an arbitrary type of anchoring is valid at $w \ll 1$; here $P_2(\cos\alpha) = (3\cos^2\alpha - 1)/2$ is the second-order Legendre polynomial and α is the angle between the easy orientation direction $\boldsymbol{n}_S$ of the director and the long axis $\boldsymbol{u}$ of a particle. From Eq. (20) it is possible to determine the equilibrium state of a particle in the uniform nematic bulk at the given type of anchoring at its surface (i.e., at the given angle α of orientation $\boldsymbol{n}_S$). If $\alpha < \alpha_* = \arccos(1/\sqrt{3})$, then $A_2 < 0$ and the particle long axis is directed along the director. If $\alpha > \alpha_*$, then $A_2 > 0$, i.e., the particle is oriented perpendicular to the director. It should be noted that this result along with Ref. [16] was obtained independently in Ref. [63]. Let us notice also that A_2 from Eq. (20) is the function of only the azimuth angle α and does not depend on the polar angle which sets up the orientation $\boldsymbol{n}_S$ in the plane, perpendicular to the direction of the particle long axis $\boldsymbol{u}$. In this sense homeotropic (II) and circular (III) types of anchoring are equivalent in the approximation $w \ll 1$.

Use of the quadratic type of orientational energy (2) with the coefficient A_2 in the form of Eq. (20) in the continuum model of soft FNs gives a chance to unify the theory for various types of anchoring at the ferroparticles surface and to remove the restriction on the maximum value of the external magnetic field which can be applied to FN. Thus, the theory [16] of soft FNs is valid at any value of an external magnetic field for magnetic suspensions with $w \ll 1$. As the coefficient A_2 from Eq. (20) is determined only by the surface energy, the term "ferroparticles surface energy" is used in Ref. [16] instead of the term "orientational energy of ferroparticles."

D. Effect of saddle-splay deformations on particle orientation

Let us estimate the models of rigid and soft FNs presented above from the standpoint of the new results received in Appendix A on energy $\mathcal{F}'_\parallel$ calculation; in particular, we will consider the influence of the saddle-splay nematic elastic modulus K_{24} on the particle orientation and coefficients A_{2k}. The above analysis we will carry out for the homeotropic type of anchoring.

As shown in Appendices A and B, the surface elastic modulus K_{24} contributes only to the energy $\mathcal{F}'_\parallel$ which can be calculated for any arbitrary values of FN material parameters [see Eq. (A15)]. In order to compare this result with $\mathcal{F}'_\perp$ more accurately for the cases of rigid (13) and soft (16) anchorings, let us write Eq. (A15) in two-constant approximation:

$$\mathcal{F}'_\parallel = \begin{cases} \pi L \bar{K}\left[\bar{k}_{24} + \frac{\bar{w} - \bar{k}_{24}}{1 + \bar{w} - \bar{k}_{24}}\right] & \text{for } \bar{w} > \bar{k}_{24} \quad \text{or} \quad W > (K_{24}/R) \\ \pi R L W & \text{for } \bar{w} \leqslant \bar{k}_{24} \quad \text{or} \quad W \leqslant (K_{24}/R), \end{cases} \tag{21}$$

where $\bar{k}_{24} = (K_{24}/\bar{K})$. For rigid anchoring with $\bar{w} \to \infty$, from the first line of Eq. (21) it follows that $\mathcal{F}'_{\parallel} = \pi L\bar{K}[1 + (K_{24}/\bar{K})]$. The value of K_{24} can be in the interval (0–2)$\bar{K}$ [64], and the expected radius of particles in FN is such that $\ln(R/2r_d) \approx \ln(R/2a) \approx 2$–3. Therefore when the values of K_{24} are close to $2\bar{K}$, the obtained estimation $\mathcal{F}'_{\parallel} \approx 3\pi L\bar{K}$ exceeds $\mathcal{F}'_{\perp}$ from Eq. (13), i.e., the particles in the absence of a field are oriented perpendicular to the director. This once again emphasizes that the theory [1] based on the approximation $\sum_{k=1}^{\infty} A_{2k} \approx -\infty$ and the postulate of parallelness of $\boldsymbol{m}$ and $\boldsymbol{n}$, cannot be used for the description of magnetic suspensions, at least those with homeotropic anchoring of the director at particles surfaces and with large values of the nematic modulus K_{24}.

On the contrary, for the model [16] of soft FNs the increase of K_{24} leads to the conclusion that the result of calculation of A_2 from Eq. (19) becomes more accurate. Indeed, for $\boldsymbol{u} \parallel \boldsymbol{n}_0$ and $\bar{w} \leqslant \bar{k}_{24} \neq 0$, i.e., $W \leqslant (K_{24}/R)$, the director near a particle is not distorted and the value of $\mathcal{F}'_{\parallel}$ is determined only by the surface anchoring energy [see Appendix A and Eq. (21)]. Therefore at $\bar{w} \leqslant \bar{k}_{24} \neq 0$ the first order of smallness with respect to $\bar{w}$ is the exact result for $\mathcal{F}'_{\parallel}$. Thus, the value (19) of A_2 calculated in this approximation, becomes more accurate. With an increase of $\bar{k}_{24}$ the applicability limit of this approximation gets extended towards larger values of $\bar{w}$. It confirms the comparative analysis of the normalized dependencies $\bar{A}_2 = (A_2 v/\pi\bar{K}L)$ presented in Fig. 4 and calculated according to the exact and approximate results [see Eqs. (17) and (19), respectively]. If we estimate an admissible calculation error of A_2 within 10%, then for $\bar{k}_{24} = 1$–2 the approximate result of (19) is possible to use up to values $\bar{w} = 1$. From Fig. 4 it can also be seen that with an increase of $\bar{k}_{24}$ the limits of stability of a particle state with $\boldsymbol{u} \perp \boldsymbol{n}_0$ expand. For example, if $\bar{k}_{24} = 1$ and 2 then the transition to the orientation $\boldsymbol{u} \parallel \boldsymbol{n}_0$ takes place at values $\bar{w}_* = 8.627$ and 27.441, respectively.

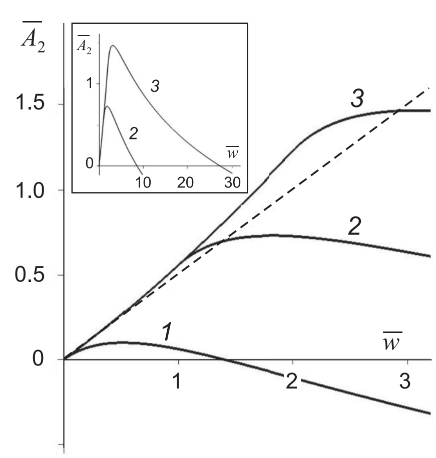

FIG. 4. Normalized coefficient $\bar{A}_2$ as a function of the dimensionless parameter $\bar{w}$. Solid curves are the results of exact calculation of $\bar{A}_2$ with the use of Eqs. (16), (17), and (21) for $\bar{k}_{24} = 0$ (curve 1), 1 (curve 2), and 2 (curve 3); the dashed line is the approximate value of $\bar{A}_2$ from Eq. (19). The plot in the inset shows the behavior of curves 2 and 3 at large values of $\bar{w}$.

One can show that for soft FNs this type of influence of the elastic modulus K_{24} on the value of A_2 is typical not only for homeotropic, but also for other types of anchoring of the nematic director at particles surface. Therefore the generalized form (20) of this coefficient can be used in a final form of Eq. (4) for the modified orientational energy of ferroparticles which have cylindrical (rodlike, needlelike, or ellipsoidal) shape with $(L/d) \geqslant 10$; it gives

$$F_{\mathrm{OR},4}(\theta) = -2\frac{W}{d}P_2 f(\boldsymbol{m}\cdot\boldsymbol{n})^2[1 - \zeta(\boldsymbol{m}\cdot\boldsymbol{n})^2],$$
$$P_2 = P_2(\cos\alpha), \quad \zeta = \zeta(\cos^2\alpha). \tag{22}$$

Here we take into account that in the general case the phenomenological parameter $\zeta = -(A_4/A_2)$, as well as expansion coefficients of orientational energy, has to depend on the type of nematic anchoring at particles surface and to be the even function of $\cos\alpha$. The latter follows from identity of the director easy orientations corresponding to the angles α and $(\pi - \alpha)$.

The need to incorporate the fourth-order term in $(\boldsymbol{m}\cdot\boldsymbol{n})$ in orientational energy of ferroparticles can be caused by end effects, local deformations of the director field near individual particles, the deviation of the particles shape from cylindrical (for example, ellipsoidal particles) that in the model [16] was not taken into account. To account for these effects we use the phenomenological parameter ζ and make a correction of the energy $(\mathcal{F}'_{\parallel} - \mathcal{F}'_{\perp})$ of an individual particle at the parallel and perpendicular orientations of its long axis with respect to the director of NLC [see Eq. (10)]. In addition, a more complete examination of the surface interactions based on bistable type of anchoring energy of the director at ferroparticles surfaces, the term of fourth order in $(\boldsymbol{m}\cdot\boldsymbol{n})$ appears in the orientational energy from Eq. (22) automatically.

Finishing the discussion of the modified type of ferroparticles orientational energy, one should be reminded once again of the applicability limits of Eq. (22) for describing soft FNs. They depend on the value of the saddle-splay nematic elastic modulus K_{24}. When $0 \leqslant K_{24}/K \leqslant 0.1$, then this expression is true in $w \ll 1$ approximation, and at $0.1 \leqslant (K_{24}/K) \leqslant 1$ it gives a correct result for $w \leqslant (K_{24}/K)$; in the case of $1 \leqslant (K_{24}/K) \leqslant 2$ it can be used at $w \leqslant 1$.

IV. FREE ENERGY OF A FERRONEMATIC

Before passing directly to the description of free energy of ferronematics, we briefly discuss their magnetic properties dependent on an orientation of needlelike particles in a nematic matrix. As shown above, in the FN homogeneous bulk with the director $\boldsymbol{n}_0 =$ const. the long axes of ferroparticles are directed either along or perpendicular to the director. Therefore in the first case for the directions $\boldsymbol{\mu}_i$ of orientation of their magnetic moments there is an easy-axis anisotropy $\boldsymbol{\mu}_i \parallel \boldsymbol{n}_0$, and in the second case there is an easy-plane anisotropy $\boldsymbol{\mu}_i \perp \boldsymbol{n}_0$. If no extra measures for orienting the FN magnetic particles in one direction are taken in preparation of a suspension, then the considered system will be in the so-called compensated state when the macroscopic density of the FN magnetic moment $\boldsymbol{M} = M_S f \sum_i \boldsymbol{\mu}_i$ is equal to zero. However, to control the mesophase orientational order, ferronematics with nonzero (spontaneous) magnetization are required. How to obtain magnetized ferronematics at $\boldsymbol{\mu}_i \parallel \boldsymbol{n}_0$ and $\boldsymbol{\mu}_i \perp \boldsymbol{n}_0$ is described in Ref. [16] in detail. In particular, it is shown, that at $\boldsymbol{\mu}_i \perp \boldsymbol{n}_0$ (i.e., when, for example, the homeotropic type of anchoring at particle surfaces is given) the bias field $\boldsymbol{H}_b \perp \boldsymbol{n}_0$ is to be applied to the ferronematic. This field magnetizes the FN along the direction $\boldsymbol{H}_b$ according to the relation

$$M = M_S f \frac{I_1(\rho)}{I_0(\rho)}, \tag{23}$$

where I_0 and I_1 are the modified Bessel functions, and $\rho = (M_S v H_b / k_B T)$ is the Langevin parameter characterizing the ratio of the magnetic energy of an individual particle and the energy of its thermal motion. As shown in Ref. [16], the magnetization of Eq. (23) reaches a saturation state $M \to M_S f$ at $\rho \geqslant 10$. For real FN, $\rho = 10$ corresponds to rather weak magnetic fields $H_b \leqslant 1$ Oe. In this case the bias field sets the initial magnetization of the system, without breaking a uniform state of the nematic matrix. For example, in the experiments [6–8] to create a uniform magnetized sample with the magnetic moment density $\boldsymbol{M} = M_S f \boldsymbol{m}$, the terrestrial magnetic field was used as the bias field.

For the magnetized FN in the external uniform magnetic field $\boldsymbol{H}$, the modified form of the volume density of free energy can be written as follows:

$$\begin{aligned} F = {} & \frac{1}{2}\{K_{11}(\nabla \cdot \boldsymbol{n})^2 + K_{22}[\boldsymbol{n} \cdot (\nabla \times \boldsymbol{n})]^2 + K_{33}[\boldsymbol{n} \times (\nabla \times \boldsymbol{n})]^2 \\ & - K_{24}\nabla \cdot [\boldsymbol{n} \times (\nabla \times \boldsymbol{n}) + \boldsymbol{n} \cdot (\nabla \cdot \boldsymbol{n})]\} \\ & + \frac{f k_B T}{v} \ln f - 2\frac{W}{d} P_2 f (\boldsymbol{m} \cdot \boldsymbol{n})^2 [1 - \zeta (\boldsymbol{m} \cdot \boldsymbol{n})^2] \\ & - M_S f (\boldsymbol{m} \cdot \boldsymbol{H}) - \frac{\chi_a}{2} (\boldsymbol{n} \cdot \boldsymbol{H})^2, \end{aligned} \tag{24}$$

where χ_a is the NLC diamagnetic anisotropy. The averaging in Eq. (24) is performed at scales much greater than the particle length L. It is implied also that at $\boldsymbol{\mu}_i \perp \boldsymbol{n}_0$ the external field $\boldsymbol{H}$ has the extra component $\boldsymbol{H}_b$.

Let us describe briefly each contribution in Eq. (24). The first term in curly brackets represents the Frank elastic energy of the nematic matrix. In comparison with the models [1,16] the saddle-splay surface contribution K_{24} is supplemented in the elastic energy density because by analogy with pure NLC this contribution must be considered at a description of ferronematics as well. The second term in Eq. (24) is the contribution of mixing entropy. The third one represents the orientational energy (22) which depends on the type of nematic anchoring at the surfaces of ferroparticles. The fourth term is the Zeeman energy describing the dipole mechanism of interaction of ferroparticles with an external magnetic field. The last one represents the diamagnetic energy which corresponds to the quadrupole interaction of nematic matrix with an external field.

For all the presented FN theories, Eq. (24) is the most general one. At $K_{24} = 0$ and $\zeta = 0$ it results in the volume density of free energy for the model [16] of soft FNs. The corresponding expression for the continuum theory [1] describing rigid FNs can also be obtained from (24) with $K_{24} = 0$, $\zeta = 0$, and formal replacement of the orientational energy expansion coefficient $A_2 = -(2WP_2/d)$ by $(-\infty)$. Then, the condition of parallelness of $\boldsymbol{m}$ and $\boldsymbol{n}$ immediately follows from Eq. (24). It is appropriate to mention here that the theory [1] is true only in magnetic fields of about several tens oersted. The diamagnetic matrix of a nematic is practically unreceptive to such values of a field, therefore, in the model [1] the diamagnetic interaction of NLC with an external magnetic field was not taken into account, i.e., the last term in Eq. (24) was not considered.

In the modified theory of soft FNs presented above, as well as in the model [16], the equilibrium state of magnetic suspension is characterized by three spatial distributions: volume fraction $f(\boldsymbol{r})$ of ferroparticles, director $\boldsymbol{n}(\boldsymbol{r})$, and unit magnetization vector $\boldsymbol{m}(\boldsymbol{r})$. Therefore, the equilibrium state of FNs corresponds to the set of equations which can be obtained by minimization of the free energy functional $\mathcal{F} = \int F\, dV$ with respect to the independent variables f, $\boldsymbol{n}$, and $\boldsymbol{m}$.

To summarize this section, we briefly discuss one more question. It consists in determining the limits of applicability of the above presented continuum approach to the description of FNs and concerns limitations on the concentration f of magnetic particles in the suspension. As shown in Refs. [1,16], under the action of an external magnetic field there are two types of behavior of a ferroparticle ensemble and a nematic matrix: individual and collective. Individual behavior means that each individual particle rotates independently of the rest, without disturbing the total orientational order of the nematic matrix. In this case the rotation of ferroparticles in the FN bulk with the characteristic scale D requires costs of orientational energy $\mathcal{F}_{\mathrm{OR}} \sim (fW/d)D^3$. In the collective behavior the rotation of ferroparticles induces the NLC director rotation, i.e., reorientation of the nematic matrix as a whole. This behavior is characterized by the Frank energy $\mathcal{F}_{\mathrm{FR}} \sim (K/D^2)D^3 = KD$. The continuum approach to the description of FNs discussed above is valid for suspensions, in which the collective response of the particles and nematic matrix to the applied magnetic field is realized. It means that the condition $\mathcal{F}_{\mathrm{FR}} < \mathcal{F}_{\mathrm{OR}}$ must be fulfilled. Due to this the concentration of particles in the suspension is bounded from below: The volume fraction f of solid phase has to be more than the so-called critical concentration $f_* \sim (Kd/WD^2)$ of collective behavior. Assuming that $K \approx 5 \times 10^{-7}$ dyn, $d \approx 7 \times 10^{-6}$ cm, $W \sim 5 \times 10^{-2}$ erg/cm^2, and $D \sim 100$–500 μm [6–8,16,34], the estimation for the critical concentration becomes $f_* \sim 10^{-8}$–10^{-6}.

On the other hand, we consider dilute suspensions and by analogy with Refs. [1,16], we neglect the effect of magnetic dipole-dipole interaction between ferroparticles in Eq. (24). As shown in Ref. [1], this approach is allowable for submicronic particles at $f \leqslant f_{**} \sim 10^{-2}$. So, the suggested modified continuum model of soft FNs is valid for suspensions with ferroparticles volume fraction $f_* < f \leqslant f_{**}$.

V. CONCLUSIONS

In this paper we have carried out the theoretical analysis of existing models of rigid [1] and soft [16] FNs. We have shown the restriction of these approaches for the description of the orientational interaction between the ensemble of ferroparticles and nematic matrix, and proposed a modified version of the continuum theory of FNs. Compared to Refs. [1] and [16] we have studied in more detail the questions of a mesoscopic description of the magnetic suspensions (at the level of an individual particle) and have received a number of important results.

It is shown, in particular, that the orientational energy of an individual particle can be represented as an expansion in even powers of the cosine of the angle of deviation of the main particle axis $\boldsymbol{u}$ from the director $\boldsymbol{n}_0$ of the uniform nematic matrix [see Eq. (7)]. It is found that the sum (8) of the coefficients of this expansion is equal to the difference of the free energies for the states with perpendicular ($\boldsymbol{u}\perp\boldsymbol{n}_0$) and parallel ($\boldsymbol{u} \parallel \boldsymbol{n}_0$) particle orientations with respect to the director $\boldsymbol{n}_0$. These results are valid both for rigid and soft character of anchoring with arbitrary direction of easy orientation of the director at the particle surface.

Developing the theoretical description of FNs at the mesoscopic level, and using as the example the homeotropic type of anchoring, we have also shown qualitative (Figs. 2 and 3) and quantitative (Appendices A and B) analogy in the description of the orientational distortions between the internal and external problems for cylindrical volumes of NLCs. We have determined the conditions under which the exact solutions for internal problems transform into exact solutions for external problems (and vice versa). We have established that the transition from the rigid to the soft character of anchoring for internal problems is equivalent to the effective increase in the radius of the cylinder, and for the external problems—to the effective decrease of the particle radius. Using these results, we have investigated the influence of surface saddle-splay elastic modulus K_{24} of the NLC, which was not considered in Refs. [1,14–16], on the equilibrium orientation of the individual needlelike (cylindrical) particle in a uniform nematic matrix. It is shown that the increase in K_{24} leads to the expansion of the limits of stability of orientational state with $\boldsymbol{u}\perp\boldsymbol{n}_0$ (the particle lies across the director), compared with the state $\boldsymbol{u} \parallel \boldsymbol{n}_0$ (the particle lies along the director) (see Fig. 4).

These results are quite general. In particular, the expansion (7) for the orientational energy and the sum (8) of the coefficients of this expansion are applicable not only to the needlelike particles in ferronematics, but also to any anisometric objects (particles or their conglomerates with an axis of cylindrical symmetry) embedded in liquid crystals: ferroelectric, dielectric, and semiconductor nanoparticles, gold nanorods, and carbon nanotubes, etc. (see Refs. [5,65–68]). In addition, if these objects have a rodlike (needlelike or cylindrical) shape, then for them the results of the qualitative and quantitative analogy between the internal and external problems for the cylindrical volumes of the NLC and the conclusions presented in Sec. III and Appendices A and B, also remain valid.

It is shown that when passing to the continuum (macroscopic) description of magnetic suspensions the orientational energy of the ferroparticles ensemble is an infinite series in even powers of the scalar product ($\boldsymbol{m}\cdot\boldsymbol{n}$) [see Eq. (1)]. In fact, this expansion establishes the succession of theoretical approaches to the description of magnetic suspensions from model [1] of rigid FNs to the model [16] of soft FNs, and then to the proposed modified model. Indeed, in the Brochard–de Gennes theory [1] it is supposed that $\boldsymbol{m} = \boldsymbol{n}$ and the orientational energy of ferroparticles is a constant that can be excluded from the expression for the free energy density of FN. In the Burylov-Raikher model [16] the expansion (1) is limited only by a quadratic term as in Eq. (2), and in the proposed model the fourth order in ($\boldsymbol{m}\cdot\boldsymbol{n}$) is additionally considered, which is the next step in the development of a continuum theory of FNs [see Eqs. (4) and (22)].

Mainly due to the change of orientational energy of ferroparticles in this paper we have proposed the modified expression (24) for the free energy density of FNs. Note that unlike Ref. [46], which considered the FNs with homeotropic anchoring on the surface of magnetic particles, expression (24) is generalized to the case of an arbitrary type of anchoring. In addition, by analogy with the pure NLC, in modified FN free energy density the term describing the surface (saddle-splay) nematic matrix deformations was introduced that is associated with the elastic modulus K_{24}. Accounting for K_{24} contribution to describe the FNs is extremely important, since it allows one to expand the limits of applicability of the proposed continuum approach. Recall that the model [16] of soft FNs is valid for magnetic suspensions, in which the anchoring energy density W of a nematic at the surface of an individual particle, the radius R of the latter, and the average value K of the NLC elastic constants satisfy the condition $w = (WR/K) \ll 1$. As shown above, for $K_{24} \sim K$ (see, for example, Refs. [52,53,69]) our modified theory corresponds to a wider class of FNs with $w \leqslant 1$.

Application of this theory to the description of the orientational behavior of ferronematics in an external magnetic field and theoretical consideration of magneto-optical effects, which can be observed in experiments on the birefringence of suspensions, will be presented in a future paper.

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APPENDIX A

Let us consider a quantitative analogy between equilibrium equations and boundary conditions corresponding to the ER structure in a cylindrical capillary on the one hand, and to the particle with $\boldsymbol{u} \parallel \boldsymbol{n}_0$ suspended in a nematic on the other hand (see Fig. 2).

Let us start from the general expression for the total free energy,

$$\mathcal{F}_{\mathrm{NLC}} = \int_V F_{\mathrm{FR}} dV + \int_S F_S \, dS, \qquad \text{(A1)}$$

where F_{FR} is the Frank potential for NLC elastic deformations; it has the form

$$\begin{aligned} F_{\mathrm{FR}} = \tfrac{1}{2}\{ & K_{11}(\nabla\cdot\boldsymbol{n})^2 + K_{22}[\boldsymbol{n}\cdot(\nabla\times\boldsymbol{n})]^2 \\ & + K_{33}[\boldsymbol{n}\times(\nabla\times\boldsymbol{n})]^2 \\ & - K_{24}\nabla\cdot[\boldsymbol{n}\times(\nabla\times\boldsymbol{n}) + \boldsymbol{n}\cdot(\nabla\cdot\boldsymbol{n})]\}. \end{aligned} \qquad \text{(A2)}$$

As mentioned above, the moduli K_{11}, K_{22}, and K_{33} are the elastic volume constants corresponding to the splay, twist, and bend deformations, respectively. The saddle-splay constant K_{24} is called the surface elastic modulus, because the divergence K_{24} contribution can be written as a surface integral (see, for instance, [52–54]).

A surface density of nematic anchoring energy F_S in Eq. (A1) we take in the Rapini form (see Refs. [14–16,32,52–54])

$$F_{S,2} = \frac{W}{2}(\boldsymbol{n} \times \boldsymbol{n}_S)^2, \tag{A3}$$

where the density W of anchoring energy corresponds to the quadratic in $(\boldsymbol{n} \times \boldsymbol{n}_S)$ term in the expansion of F_S, and $\boldsymbol{n}_S$ is the direction of easy orientation of the director at the sample surface S. In the considered problems corresponding to the homeotropic type of anchoring with $W > 0$, the easy orientation direction $\boldsymbol{n}_S$ may be chosen along the direction of an outward normal to the cylindrical surface.

Let us note that in Refs. [14,15] in the calculation of $\mathcal{F}'_\parallel$ two-constant approximation $K_{11} = K_{33}$ was used, while in Ref. [1] the general approach to the problem of behavior of an individual particle in NLC was based on the one-constant approximation $K_{11} = K_{22} = K_{33}$. In addition, in Refs. [1,14,15] the contribution with the surface elastic modulus K_{24} was not taken into account for calculations of $\mathcal{F}'_\parallel$. Here, by using the analogy between the internal and external problems for cylindrical capillaries, it is possible to obtain a general form of the solution for arbitrary values of material parameters entered into Eqs. (A1)–(A3).

The director distribution for the internal and external problems in the cylindrical coordinate system can be found as follows:

$$\boldsymbol{n} = \sin\,\Omega(\beta)\cdot \boldsymbol{e}_\beta + \cos\,\Omega(\beta)\cdot \boldsymbol{e}_z, \quad \beta = r,r'. \tag{A4}$$

Here $\beta = r$ and $\beta = r'$ correspond to the internal and external problems, respectively; at the initial stage of carrying out the analogy, these values are assumed as independent variables.

Substituting Eq. (A4) into Eqs. (A1)–(A3) we obtain the following expressions for the energies of ER structure:

$$\begin{aligned}\mathcal{F}_{\rm ER} = \pi L\Bigg[\int_0^R \Bigg\{&[K_{11}\cos^2\Omega(r) + K_{33}\sin^2\Omega(r)]\\ &\times\left(\frac{d\Omega(r)}{dr}\right)^2 + \frac{K_{11}\sin^2\Omega(r)}{r^2}\Bigg\} r\,dr\\ &+ (WR - K_{11} + K_{24})\cos^2\Omega(R) + K_{11} - K_{24}\Bigg]\end{aligned} \tag{A5}$$

and for the configuration corresponding to orientational distortion induced by the particle at $\boldsymbol{u} \parallel \boldsymbol{n}_0$,

$$\begin{aligned}\mathcal{F}'_\parallel = \pi L\Bigg[\int_R^\infty \Bigg\{&[K_{11}\cos^2\Omega(r') + K_{33}\sin^2\Omega(r')]\\ &\times\left(\frac{d\Omega(r')}{dr'}\right)^2 + \frac{K_{11}\sin^2\Omega(r')}{r'^2}\Bigg\} r'\,dr'\\ &+ (WR + K_{11} - K_{24})\cos^2\Omega(R) - K_{11} + K_{24}\Bigg],\end{aligned} \tag{A6}$$

where L is the capillary and the particle length.

The integrands in Eqs. (A5) and (A6) formally coincide, therefore the equilibrium equations for the internal and external problems, obtained by minimization of functionals Eqs. (A5) and (A6), have identical form

$$\begin{aligned}&[\cos^2\Omega(\beta) + \eta\sin^2\Omega(\beta)]\left[\frac{d^2\Omega(\beta)}{d\beta^2} + \frac{1}{\beta}\frac{d\Omega(\beta)}{d\beta}\right]\\ &\quad + \sin\,\Omega(\beta)\cos\,\Omega(\beta)\left[(\eta - 1)\left(\frac{d\Omega(\beta)}{d\beta}\right)^2 - \frac{1}{\beta^2}\right] = 0,\end{aligned} \tag{A7}$$

where $\eta = (K_{33}/K_{11})$. The boundary conditions for the internal problem have the form

$$\begin{aligned}&\Omega(r=0) = 0,\\ &\left.\frac{d\Omega(r)}{dr}\right|_{r=R} = \frac{\sigma\,\sin\,\Omega(R)\cos\,\Omega(R)}{R[\cos^2\Omega(R) + \eta\,\sin^2\Omega(R)]},\\ &\sigma = \frac{WR - K_{11} + K_{24}}{K_{11}},\end{aligned} \tag{A8}$$

and for the external problem as follows:

$$\begin{aligned}&\Omega(r'=\infty) = 0,\\ &\left.\frac{d\Omega(r')}{dr'}\right|_{r'=R} = -\frac{\sigma'\sin\,\Omega(R)\cos\,\Omega(R)}{R[\cos^2\Omega(R) + \eta\,\sin^2\Omega(R)]},\\ &\sigma' = \frac{WR + K_{11} - K_{24}}{K_{11}}.\end{aligned} \tag{A9}$$

Now let us consider how the formulation of the problem for the ER structure can change if transformation (11), i.e., $r = (R^2/r')$, is used, and compare it with the problem formulation for the particle with $\boldsymbol{u} \parallel \boldsymbol{n}_0$. This results in the following. The equilibrium equation (A7) is invariant with respect to that transformation, the first boundary condition (A8) turns into the first boundary condition (A9), and the second boundary condition (A8) can be written as

$$\left.\frac{d\Omega(r')}{dr'}\right|_{r'=R} = -\frac{\sigma\,\sin\,\Omega(R)\cos\,\Omega(R)}{R[\cos^2\Omega(R) + \eta\,\sin^2\Omega(R)]}. \tag{A10}$$

To make Eq. (A10) coincide with the boundary condition (A9) at the particle surface, it is necessary to use the substitution $\sigma \to \sigma'$. Then the solution for the external problem can be obtained from the well-known internal problem solution from Refs. [52–54], using two simple substitutions $r \to (R^2/r')$ and $\sigma \to \sigma'$. The latter substitution is equivalent to $K_{24} \to (2K_{11} - K_{24})$. Let us note that the conditions $0 \leqslant (2K_{11} - K_{24}) \leqslant 2K_{11}$ fulfill for the value $(2K_{11} - K_{24})$ because the possible values of K_{24} belong to the interval $0 \leqslant K_{24} \leqslant \min\{2K_{11},2K_{33}\}$ (see Ref. [64]).

To write the final result, let us recall the well-known solution [52–54], corresponding to the ER structure. It depends on the value of σ. For $\sigma > 1$, the equilibrium distribution $\Omega(r)$ is given by the implicit function

$$\begin{aligned}&r = R\sqrt{\frac{\sigma+1}{\sigma-1}\frac{\Lambda-1}{\Lambda+1}}\exp\left(\sqrt{\eta-1}\arctan\frac{\sqrt{\eta-1}(\Lambda-\sigma)}{\Lambda\sigma+\eta+1}\right),\\ &\Lambda = \sqrt{1+\eta\,\tan^2\Omega}, \quad 0 \leqslant \Omega \leqslant \pi/2.\end{aligned} \tag{A11}$$

The result is written in terms of Ref. [54]. It is valid both for $\eta \geqslant 1$ and for $0 < \eta < 1$; in the latter case one has to use the identity replacement in the exponent:

$$\sqrt{\eta-1}\,\arctan[\sqrt{\eta-1}\lambda(\eta,\ldots)] \equiv \sqrt{1-\eta}\,\mathrm{arctanh}[\sqrt{1-\eta}\lambda(\eta,\ldots)], \tag{A12}$$

where $\lambda(\eta,\ldots)$ is the sign-definite function for any value of $\eta > 0$. For $\sigma > 1$, the boundary value of Ω at the capillary surface is determined by the equation $\Omega(r = R) = \arctan(\sqrt{(\sigma^2-1)/\eta})$. For $\sigma \leqslant 1$ the boundary value of Ω is equal to zero, and the state of complete escaping takes place in cylindrical capillary, when $\Omega(r) = 0$ all over the bulk of the sample, and the director is parallel to the capillary axis. The uniform state, which corresponds to the complete escaping of ER configuration, is called the "axial (AX) structure" [54]. Total free energies of these configurations are determined by the relations

$$\mathcal{F}_{\mathrm{ER}} = \begin{cases} \pi L K_{11}\left[2 - \frac{K_{24}}{K_{11}} + \frac{\eta}{\sqrt{\eta-1}}\arctan\frac{\sqrt{\eta-1}(\sigma-1)}{\sigma+\eta-1}\right] & \text{for} \quad \sigma > 1\\ \pi R L W & \text{for} \quad \sigma \leqslant 1. \end{cases} \tag{A13}$$

Using the substitutions $\sigma \to \sigma'$ [i.e.. $K_{24} \to (2K_{11} - K_{24})$] and $r \to (R^2/r')$ in Eqs. (A11) and (A13), we obtain the final solution for the external problem, which corresponds to the particle suspended in a nematic with the parallel orientation of its axis with respect to the unperturbed director. In the case $\sigma' > 1$ [or $W > (K_{24}/R)$], the implicit function $\Omega(r')$ takes the form

$$r' = R\sqrt{\frac{\sigma'-1}{\sigma'+1}\frac{\Lambda+1}{\Lambda-1}}\exp\left(\sqrt{\eta-1}\,\arctan\frac{\sqrt{\eta-1}(\sigma'-\Lambda)}{\Lambda\sigma'+\eta+1}\right), \tag{A14}$$

and the boundary angle Ω at the particle surface is determined from the expression $\Omega_R = \Omega(r' = R) = \arctan(\sqrt{(\sigma'^2-1)/\eta})$. If $\sigma' \leqslant 1$ [or $W \leqslant (K_{24}/R)$] the director distribution becomes uniform [$\Omega(r') = 0$] all over the bulk, i.e., the particle does not induce orientational distortions in the nematic matrix. The total free energy of the system obtained from Eq. (A13) by the substitutions presented above can be written as

$$\mathcal{F}'_{\parallel} = \begin{cases} \pi L K_{11}\left[\frac{K_{24}}{K_{11}} + \frac{\eta}{\sqrt{\eta-1}}\arctan\frac{\sqrt{\eta-1}(\sigma'-1)}{\sigma'+\eta-1}\right] & \text{for} \quad \sigma' > 1 \quad \text{or} \quad W > (K_{24}/R)\\ \pi R L W & \text{for} \quad \sigma' \leqslant 1 \quad \text{or} \quad W \leqslant (K_{24}/R). \end{cases} \tag{A15}$$

Note that for $0 < \eta < 1$ in Eqs. (A14) and (A15) the identity replacement similar to Eq. (A12) is to be used.

To verify the presented analogy between the internal and external problems we substitute the solution of Eq. (A14) into the equilibrium equation (A7) and boundary conditions (A9). In this case it is expedient to perform the following change of variables: We consider Ω as the independent variable and the coordinate r' as the following function of this angle: $r' = R\Theta(\Omega)$. The latter expression, differentiated twice with respect to r' as an implicit function, gives

$$\frac{d\Omega(r')}{dr'} = \left(R\frac{d\Theta(\Omega)}{d\Omega}\right)^{-1}, \quad \frac{d^2\Omega(r')}{dr'^2} = -R^{-2}\frac{d^2\Theta(\Omega)}{d\Omega^2}\left(\frac{d\Theta(\Omega)}{d\Omega}\right)^{-3}. \tag{A16}$$

The equilibrium equation (A7) and the boundary conditions (A9) in terms of the function $\Theta = \Theta(\Omega)$ and the variable Ω take the form

$$\Theta(\cos^2\Omega + \eta\sin^2\Omega)\left[\Theta\frac{d^2\Theta}{d\Omega^2} - \left(\frac{d\Theta}{d\Omega}\right)^2\right] + \sin\Omega\cos\Omega\left[\left(\frac{d\Theta}{d\Omega}\right)^3 - (\eta-1)\Theta^2\frac{d\Theta}{d\Omega}\right] = 0, \tag{A17}$$

$$\Theta(\Omega = 0) = \infty, \quad \left.\frac{d\Theta(\Omega)}{d\Omega}\right|_{\Omega=\Omega_R} = -\frac{\cos^2\Omega_R + \eta\sin^2\Omega_R}{\sigma'\sin\Omega_R\cos\Omega_R}. \tag{A18}$$

Expressing the function $\Theta(\Omega)$ from Eq. (A14) and substituting it into Eqs. (A17) and (A18) one can show that these relations can be transformed into identities.

The validity of expression (A15) for the energy can be seen if we substitute the function $\Omega(r') = 0$ into Eq. (A6) for $\mathcal{F}'_{\parallel}$ at $W \leqslant (K_{24}/R)$ and Eq. (A14) at $W > (K_{24}/R)$. In the latter case, for calculating the integral in Eq. (A6) we can again use the replacement of variables $r' = R\Theta(\Omega)$ and Eq. (A16). The results of calculation of $\mathcal{F}'_{\parallel}$ completely coincide with Eqs. (A15). Let us note also that in the approximation $\{K_{11} = K_{33},\ K_{24} = 0\}$ the obtained results coincide with the results of Refs. [14,15] [see Eq. (15)], and in the approximation $\{K_{11} = K_{33},\ K_{24} = 0,\ W = \infty\}$, with the result of Ref. [1] [see Eq. (12)]. Thus, the presented quantitative analogy between the internal and external problems is proper.

Let us add that the mentioned quantitative analogy could be obtained in a simpler way, applying the transformation $r = (R^2/r')$ directly to Eq. (A5) for the ER structure energy. In this case the volume integral from Eq. (A5) transforms into the volume integral in Eq. (A6) for the distortion energy induced by the particle at $\boldsymbol{u} \parallel \boldsymbol{n}_0$, and the surface contributions (A5) and (A6) become equal under the replacement $K_{24} \to (2K_{11} - K_{24})$, i.e., $\sigma \to \sigma'$. However, here we preferred to perform a more detailed consideration in order to study the influence of this transformation on the equilibrium equation, boundary conditions, and, finally, distribution of the director near the particle.

In the conclusion of this Appendix we consider the question of physical interpretation of transition from rigid to soft anchoring in the studied problem about the particle suspended in a nematic with $\boldsymbol{u} \parallel \boldsymbol{n}_0$. The obtained solution (A14) for the implicit dependence $\Omega(r')$ can be written as

$$r' = (p_{\parallel} R)\Theta_{W=\infty} = R_{ef,\parallel}\Theta_{W=\infty}, \tag{A19}$$

where $\Theta_{W=\infty}$ is the function of Ω corresponding to the rigid anchoring:

$$\Theta_{W=\infty} = \left(\frac{r'}{R}\right)\Big|_{W=\infty} = \sqrt{\frac{\Lambda+1}{\Lambda-1}}\exp\left(\sqrt{\eta-1}\arctan\frac{\sqrt{\eta-1}}{\Lambda}\right), \tag{A20}$$

the dimensionless coefficient $p_{\parallel}$ is determined by the expression

$$p_{\parallel} = \sqrt{\frac{\sigma'-1}{\sigma'+1}}\exp\left(-\sqrt{\eta-1}\arctan\frac{\sqrt{\eta-1}}{\sigma'}\right), \tag{A21}$$

and the subscript $p_{\parallel}$ means that this coefficient corresponds to the parallel orientation of $\mathbf{u}$ and $\mathbf{n}_0$. As follows from Eq. (A21), at the given value of the particle radius R and material parameters of the liquid crystal, i.e., K_{11}, K_{33}, and K_{24}, the coefficient $p_{\parallel}$ is the function of anchoring energy density W. With the increase of W starting from (K_{24}/R) [which corresponds to $\sigma' = 1$] to infinity [i.e., $\sigma' \to \infty$], the value of $p_{\parallel}$ monotonously increases from zero to one and has the following asymptotics:

$$p_{\parallel} = \begin{cases} \left[\left(\frac{\sigma'-1}{2}\right)^{1/2} + \frac{3\eta-4}{2\eta}\left(\frac{\sigma'-1}{2}\right)^{3/2}\right]\exp(-\sqrt{\eta-1}\arctan\sqrt{\eta-1}) & \text{for} \quad \sigma' \to +1 \\ 1 - \frac{\eta}{\sigma'} + \frac{\eta^2}{2\sigma'^2} & \text{for} \quad \sigma' \to \infty. \end{cases} \tag{A22}$$

Thus, the parameter $0 \leqslant p_{\parallel} \leqslant 1$ plays a role of compression coefficient indicating to what extent the particle effective radius $R_{ef,\parallel} = p_{\parallel} R$ decreases upon the transition from rigid to soft anchoring [see Eq. (A19) and Fig. 2(c)]. Equations (A19)–(A22) allow us to give a physical interpretation of the transition from nonuniform distribution (A14) to a uniform one with $\Omega(r') = 0$, which occurs at $W = (K_{24}/R)$ or $\sigma' = 1$. In this case the coefficient $p_{\parallel}$—and consequently, the particle effective radius $R_{ef,\parallel}$—is reduced to zero, which means the formal absence of the particle in the nematic matrix with uniform distribution of the director field.

APPENDIX B

In this Appendix we consider the quantitative analogy between the PP structure in a cylindrical capillary and the orientational distortion induced by the particle suspended in the nematic at perpendicular orientation of its long axis with respect to the unperturbed director (see Fig. 3).

The director field in these problems is two dimensional, and in the cylindrical coordinate system it is determined by the angle $\Phi = \Phi(\beta,\varphi)$ of director deviation from the direction of polar axis:

$$\boldsymbol{n} = \cos(\Phi - \varphi)\cdot \boldsymbol{e}_{\beta} + \sin(\Phi-\varphi)\cdot \boldsymbol{e}_{\varphi}, \tag{B1}$$

where, as well as in Appendix A, the variables $\beta = r$ and $\beta = r'$ corresponding to the internal and external problems are independent at the initial stage of consideration. The angle $\Phi_S = \Phi_S(\beta = R,\varphi)$ of the easy director orientation $\boldsymbol{n}_S$ on the cylindrical surface can be chosen as

$$\Phi_S = \begin{cases} \varphi, & 0 < \varphi < \pi \\ \varphi - \pi, & \pi < \varphi < 2\pi. \end{cases} \tag{B2}$$

The expressions for total free energy of internal and external problems are found by substituting (B1) and (B2) into (A1)–(A3); it yields

$$\begin{aligned} \mathcal{F}_i = \frac{L\bar{K}}{2}\int_{\beta_{i,1}}^{\beta_{i,2}} \beta\, d\beta \int_0^{2\pi} \Bigg\{ & \left(\frac{\partial\Phi}{\partial\beta}\right)^2 + \frac{1}{\beta^2}\left(\frac{\partial\Phi}{\partial\varphi}\right)^2 \\ & + k\left[\left(\frac{\partial\Phi}{\partial\beta}\right)^2 - \frac{1}{\beta^2}\left(\frac{\partial\Phi}{\partial\varphi}\right)^2\right]\cos(2\Phi - 2\varphi) \\ & + 2\frac{k}{\beta}\frac{\partial\Phi}{\partial\beta}\frac{\partial\Phi}{\partial\varphi}\sin(2\Phi-2\varphi)\Bigg\} d\varphi \\ & + \frac{LWR}{2}\int_0^{2\pi}\sin^2(\Phi - \Phi_S)\, d\varphi, \end{aligned} \tag{B3}$$

$$\bar{K} = \frac{K_{11}+K_{33}}{2}, \quad k = \frac{K_{33}-K_{11}}{2}, \quad i = 1,2.$$

Here the energy $\mathcal{F}_1 \equiv \mathcal{F}_{\mathrm{PP}}$ corresponds to the internal problem for the PP structure, and $\mathcal{F}_2 \equiv \mathcal{F}'_{\perp}$ corresponds to the external problem for the particle with $\boldsymbol{u} \perp \boldsymbol{n}_0$; the intervals of integration over β for internal ($i = 1$, $\beta = r$) and external ($i = 2$, $\beta = r'$) problems are $[\beta_{11},\beta_{12}] = [0,R]$ and $[\beta_{21},\beta_{22}] = [R,\infty]$, respectively.

As seen from Eq. (B3), the total free energy depends only on two NLC elastic constants K_{11} and K_{33}. Unfortunately, in the general case of arbitrary values of K_{11} and K_{33} the energies of internal $\mathcal{F}_{\mathrm{PP}}$ and external $\mathcal{F}'_{\perp}$ problems are not invariant relative to the transformation (11), i.e., $r = (R^2/r')$. At such transformation the sign of the contribution $2k(\partial\Phi/\partial\beta)(\partial\Phi/\beta\partial\varphi)\sin(2\Phi - 2\varphi)$ in Eq. (B3) is reversed, and this change of sign cannot be compensated by substitutions as were made in Appendix A. For example, use of the substitution $k \to -k$ does not lead to the desired result since in Eq. (B3) there are other contributions proportional to the parameter k. Therefore, in the general case when $K_{11} \neq K_{33}$ it is not possible to use the exact solution obtained in Ref. [54] for the PP configuration. At the same time in the two-constant approximation when $K_{11} = K_{33} = \bar{K}$ and $k = 0$, the expressions for the energies $\mathcal{F}_{\mathrm{PP}}$ and $\mathcal{F}'_{\perp}$ are completely invariant with respect to transformation of the inverse-radius vectors, i.e., their values have to coincide. This conclusion is confirmed by the results of Refs. [15,52–54] where the energies $\mathcal{F}_{\mathrm{PP}}$ and $\mathcal{F}'_{\perp}$ were calculated independently:

$$
\begin{aligned}
&[52\text{–}54]\colon \mathcal{F}_{\mathrm{PP}} = \pi \bar{K} L\left[-\ln\left(\frac{c^4-1}{c^4}\right) + \frac{\bar{w}}{2}\frac{c^2-1}{c^2}\right] \\
&\Leftrightarrow [15]\colon \mathcal{F}'_{\perp} = \pi \bar{K} L\left[-\ln(1-p_{\perp}^4) + \frac{\bar{w}}{2}(1-p_{\perp}^2)\right],
\end{aligned}
\tag{B4}
$$

where

$$
\begin{aligned}
c &= \sqrt{\frac{\sqrt{4+\bar{w}^2}+2}{\bar{w}}}, \\
p_{\perp} &= \frac{1}{c} = \sqrt{\frac{\sqrt{4+\bar{w}^2}-2}{\bar{w}}}, \\
\bar{w} &= \frac{WR}{\bar{K}}.
\end{aligned}
\tag{B5}
$$

As shown in Ref. [54], for the PP structure the parameter $1 \leqslant c \leqslant \infty$ acts as an expansion coefficient which defines the effective radius $R_{\mathrm{ef,PP}} = (cR)$ of the cylindrical capillary upon the transition from rigid to soft anchoring. So, for the particle with $\boldsymbol{u}\perp\boldsymbol{n}_0$ the compression coefficient $0 \leqslant p_{\perp} \leqslant 1$ determines the effective radius $R_{\mathrm{ef},\perp} = (p_{\perp}R)$ of this particle upon the same transformation [see Fig. 3(c)]. It is confirmed by expressions for the angles $\Phi_{\mathrm{PP}} = \Phi(r,\varphi)$ and $\Phi_{\perp} = \Phi(r',\varphi)$ of the director orientation in the case of external and internal problems which are connected by transformation (11), i.e.,

$$
\begin{aligned}
&[52\text{–}54]\colon \Phi_{\mathrm{PP}} = \arctan\frac{r^2\sin 2\varphi}{r^2\cos 2\varphi-(cR)^2} + \frac{\pi}{2} \overset{r=R^2/r'}{\longleftrightarrow} \\
&[15]\colon \Phi_{\perp} = \arctan\frac{(p_{\perp}R)^2\sin 2\varphi}{(p_{\perp}R)^2\cos 2\varphi - r'^2} + \frac{\pi}{2},
\end{aligned}
\tag{B6}
$$

and also the asymptotic behavior of compression coefficient $p_{\perp}$:

$$
p_{\perp} = \begin{cases} \frac{\bar{w}^{1/2}}{2} - \frac{\bar{w}^{5/2}}{64} & \text{for} \quad \bar{w}\to 0 \\ 1 - \frac{1}{\bar{w}} + \frac{1}{2\bar{w}^2} & \text{for} \quad \bar{w}\to\infty. \end{cases}
\tag{B7}
$$

From Fig. 3 and Eqs. (B6) and (B7) one can see that for rigid anchoring when $\bar{w}=\infty$ and $p_{\perp}=1$, the disclination lines with the charges $q'_1 = q'_2 = -1$ remain at the particle surface. For finite values of $\bar{w}$ when $0 < p_{\perp} < 1$, they are localized inside the particle and become purely fictitious. When $\bar{w}$ tends to zero, i.e., $p_{\perp}\to 0$, the disclinations $q'_1 = q'_2 = -1$ and the linear defect with the charge $q'_3 = 2$, located at the particle axis, annihilate, so that the director field becomes uniform.

This behavior of disclinations in the case of the external problem is completely analogous to the behavior of linear defects corresponding to the internal problem. In the case of PP configuration in the cylindrical capillary [see Fig. 3(a)] with $\bar{w}$ decreasing from infinity to zero the disclination lines with the charges $q_1 = q_2 = 1$ first disappear from the capillary surface and become fictitious, and then annihilate with the defect $q_3 = -2$ at the infinite point of the Riemann sphere.

So, in the two-constant approximation $K_{11} = K_{33} = \bar{K}$ the director orientation angles for the internal and external problems are uniquely related by the transformation of the inverse-radius vectors $r = (R^2/r')$, and the distortion corresponding to the particle with $\boldsymbol{u}\perp\boldsymbol{n}_0$ is energetically completely equivalent to the PP structure in the cylindrical capillary.

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