Disclination dynamics on the surfaces of nematic liquid crystal spheres

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The possible number of disclinations on the surface of a sphere is determined. It is shown that any configuration with more than two disclinations is unstable. The disclination dynamics was investigated on individual spheres and for two coalescing spheres having at the center a "hedgehog" or two surface disclinations. It was established that two disclinations of negative sign are spontaneously produced at the instant of sphere coalescence. The law governing the variation of the distance r between disclination was found to be of the form $r^2 \propto t$.

Depending on the boundary conditions, spherical drops of nematic liquid crystals (NLC) can contain either hedgehog-type defects in the bulk, or a number of disclination states on the surface. Molecule orientation is maintained homogeneous in the bulk,[1–3] while the topological stability and the number of point defects on the surface are determined by the Euler characteristics of the latter.[4,5] A change in the boundary conditions can cause two surface disclination (boojums) of strength $+1$ and a hedgehog to go over into two surface disclinations. These statements were checked in a number of studies by varying the boundary conditions for the director on the sphere surface; in particular, transitions of a hedgehog into two disclinations on a sphere have been observed.[6]

The purpose of the present paper is to study the dynamics of various disclination configurations produced on NLC spheres as a result of coalescence of two drops with two surface disclinations, two hedgehogs, a hedgehog, and a sphere with two defects on the surface. Another aim was to elucidate the conditions for stabilization of such configurations and investigation of the subsequent dynamics of the disclinations when these conditions are violated. To study these processes of interaction and annihilation of disclinations on spherical NLC drops, we used high-speed motion-picture photography. The temperature was stabilized to within 0.01 °C using heat-stabilized stages with two heaters each, one with slow response time. The optical system was based on the MKU-2 microscopic setup. The signs of the disclinations were determined by a polarization-optics method: the sign is positive if the rotation of the extinction bands near the disclination core coincides with the direction of polarizer rotation. We investigated nematic drops measuring 1–100 μm, produced in the vicinity of the phase transition from an isotropic liquid to an NLC. Equilibrium states with more than two disclinations on the surface were produced by stabilizing the latter by attaching impurity particles to the sphere surface.[7] The particles used in this case where dispersed and readily sublimating I_2 and Br_2 compounds. This made it possible to investigate equilibrium symmetric configurations of the disclinations when their number on the surface was large. To alter the boundary conditions, rigid-chain polymer impurities, such as polydiphenylene, were introduced in the NCL. This has made it possible, at certain impurity densities ($\sim 1\%$), to produce spheres with tangential or homeotropic orientation of the molecules on their surfaces.

POSSIBLE SYMMETRIC DISCLINATION CONFIGURATIONS ON SPHERES

When two spherical NLC surfaces interact, a certain new configuration of disclinations is observed on the resultant surface. The number of disclinations is generally speaking not equal to that on the initial surfaces. It is determined by the condition that the elastic energy be a maximum, and the resultant disclination configuration is determined by the equation of state. Let us consider the possible configuration of defects on the surface of NLC spheres. We denote by $\mathbf{N}$ the director, for which the equilibrium equation on the sphere surface takes in the single-constant approximation the form

$$\Delta \mathbf{N}=0.$$

The boundary conditions require that the director $\mathbf{N}$ be tangent to the sphere surface, i.e., in spherical coordinates

$$\mathbf{N}=\mathbf{e}_\theta N_\theta+\mathbf{e}_\varphi N_\varphi.$$

By virtue of the equivalence of $\mathbf{N}$ and $-\mathbf{N}$, in the NLC upon inversion we have $\mathbf{N}\to\mathbf{N}$, i.e.,

$$\left.\begin{array}{l}\theta\to\pi-\theta\\ \varphi\to\pi+\varphi\end{array}\right\}, \quad \text{with} \quad N_\theta\to N_\theta, \quad N_\varphi\to -N_\varphi.$$

There exists also a symmetry plane and an axis perpendicular to this plane, i.e.,

$$\left.\begin{array}{l}\theta\to\pi-\theta\\ \varphi\to-\varphi\end{array}\right\}, \quad \begin{array}{l}\text{with} \quad N_\theta\to N_\theta, \quad N_\varphi\to N_\varphi,\\ \text{or} \quad N_\theta\to -N_\theta, \quad N_\varphi\to -N_\varphi.\end{array}$$

The solution of the Leplace equation for N on the surface of a sphere, with tangential boundary conditions, is in the general case[8]

$$N=\sum_{n=1}^{\infty}\sum_{m=1}^{n}(b_{mn}\mathbf{B}_{mn}+c_{mn}\mathbf{C}_{mn}),$$

where $\mathbf{B}_{mn}$ and $\mathbf{C}_{mn}$ are spherical vector harmonics of second and third kind, respectively:

$$\mathbf{B}_{mn}=[n(n+1)]^{-1/2}r\,\mathrm{grad}\,[X_n^m(\theta,\varphi)],$$
$$\mathbf{C}_{mn}=[n(n+1)]^{-1/2}\,\mathrm{rot}\,[\mathbf{r}X_n^m(\theta,\varphi)],$$

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TABLE I. Possible sphere disclination configurations that follow from the equations of state of NLC.

Indices of spherical functions	Number of disclination (strength)	N_θ N_φ	Map of disclination locations
$n=1$ $m=1$ $\mathbf{N}=\mathbf{B}_{11}^0$	2(+1) Stable	$\cos\theta\sin\varphi$ $\cos\varphi$	see Fig. 1a
$n=2$ $m=1$ $\mathbf{N}=\mathbf{C}_{12}^0$	4(+1) 2(−1) Unstable	$\cos\theta\cos\varphi$ $(2\cos^2\theta-1)\sin\varphi$	Fig. 1b
$n=2$ $m=2$ $\mathbf{N}=\mathbf{C}_{22}^0$	4(+1) 2(−1) Unstable	$\sin\theta\cos 2\varphi$ $\sin\theta\cos\theta\sin 2\varphi$	Fig. 1c
$n=3$ $m=1$ $\mathbf{N}=\mathbf{B}_{13}^0$	6(+1) 4(−1) Unstable	$(15\cos^2\theta-11)\cos\theta\sin\varphi$ $(5\cos^2\theta-1)\cos\varphi$	Fig. 1d
$n=3$ $m=2$ $\mathbf{N}=\mathbf{B}_{23}^0$	8(+1) 6(−1) Unstable	$(3\cos^2\theta-1)\sin\theta\sin 2\varphi$ $\sin\theta\cos\theta\cos 2\varphi$	Fig. 1e
$n={}^3/_2$ $m={}^3/_2$ $\mathbf{N}=\mathbf{C}^0_{3/2\,3/2}$	3(+1) 2(−¹/₂) Unstable	$\sin\theta\cos{}^3/_2\varphi$ $\sin\theta\cos\theta\sin{}^3/_2\varphi$	Fig. 1f

where

$$X_n^m(\theta,\varphi)=P_n^m(\cos\theta)e^{im\varphi}$$

is a scalar spherical harmonic. The constant coefficients b_{mn} and c_{mn} are chosen to satisfy the normalization condition $\mathbf{N}^2=1$.

Inversion leads to

$$\mathbf{B}_{mn}\to(-1)^{n+1}\mathbf{B}_{mn},\quad \mathbf{C}_{mn}\to(-1)^n\mathbf{C}_{mn};$$

hence

$$\mathbf{N}=\sum_{n=1}^{\infty}\sum_{m=1}^{n}[\alpha_n b_{mn}\mathbf{B}_{mn}+(1-\alpha_n)c_{mn}\mathbf{C}_{mn}],$$

where

$$\alpha_n=\begin{cases}1, & n=2k+1\\ 0, & n=2k+2.\end{cases}$$

Considering the symmetry configuration, we obtain

$$\mathbf{N}=\sum_{n=1}^{\infty}\sum_{m=1}^{n}[\alpha_n b_{mn}\mathbf{B}_{mn}{}^0+(1-\alpha_n)c_{mn}\mathbf{C}_{mn}{}^0],$$

the superscript "0" labels the imaginary part.

The director distribution on the NLC sphere surface is determined therefore in the general case by one of the harmonics

$$\mathbf{N}_{mn}=\alpha_n b_{mn}\mathbf{B}_{mn}{}^0+(1-\alpha_n)c_{mn}\mathbf{C}_{mn}{}^0$$

or by one of their linear superposition. The singular points of the vector field $\mathbf{N}$ are zeros; these points are none other than the disclinations on the surface of the sphere. For each solution $\mathbf{N}_{mn}$ we have a different disclination picture (see the table).

Using the expression for the number of zeros of the associated Legendre polynomials $P_h^m(\cos\theta)$ and analyzing the expressions for $\mathbf{B}_{mn}$ and $\mathbf{C}_{mn}$, we obtain the number $\mathbf{N}_{mn}$ of the zeros of the solution, i.e., the number of disclinations on the surface of the sphere for the given solution:

$$l=4m(n-m)+2m+2(1-\delta_{1m}),$$

n and m are integer positive numbers. If the number of disclinations l satisfies the relation $l=4k-2$, $k=1,2,3,\ldots$, we have, for a given solution $\mathbf{N}_{mn}$, $2k$ disclination of strength $+1$ and $2k-2$ disclinations of strength -1. By considering $\mathbf{N}_{mn}$ for half-integer n and m we can obtain disclination configurations with half-integer strength (see Table I). In general, for an arbitrary solution $\mathbf{N}_{mn}$ the number of the indices of the zeros is equal to two.

Let us examine the stability of the disclination configuration corresponding to the solution $\mathbf{N}_{mn}$. Using the relations

$$\operatorname{div}\mathbf{C}_{mn}=0,\quad \operatorname{rot}\mathbf{B}_{mn}=0,$$
$$\operatorname{rot}\mathbf{C}_{mn}=k_n\mathbf{e}_r X_n{}^m,\quad \operatorname{div}\mathbf{B}_{mn}=-k_n X_n{}^m,$$

where kn is a constant that depends on n, we obtain for the free-energy surface density in the single-constant approximation:

$$F_d={}^1/_2k[(\operatorname{div}\mathbf{N}_{mn})^2+(\operatorname{rot}\mathbf{N}_{mn})^2],$$

with accuracy to within a constant

$$F_d=[X_n{}^{m0}]^2=[P_n{}^m(\cos\theta)\sin m\varphi]^2.$$

Next,

$$\sin m\varphi=\sum_{k=0}^{E[(m-1)/2]}(-1)^k C_m^{2k+1}\times[\cos\varphi]^{m-2k-1}[\sin\varphi]^{2k+1},$$

where E[. . .] stands for the integer part; hence

$$\sin^2 m\varphi=\sum_{k=1}^{m}(-1)^{k+1}A_k^{(m)}\sin^{2k}\varphi.$$

In addition,

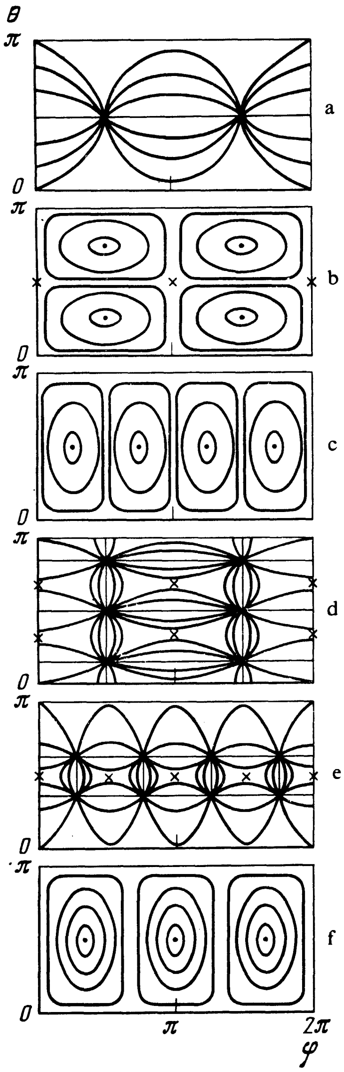

FIG. 1. Map of dislocation arrangement (see the table).

$$[P_n^m(\cos\theta)]^2=\frac{(n+m)!}{(n-m)!}\sum_{k=m}^{n}(-1)^{k+m}\times\frac{(2k)!}{2^{2k}(k-m)!(k+m)!(k!)^2}\cdot\frac{(n+k)!}{(n-k)!}\sin^{2k}\theta.$$

Thus,

$$F_d=\left(\sum_{i=m}^{n}(-1)^{i+m}B_i^{(m)}\sin^{2i}\theta\right)\times\left(\sum_{k=1}^{m}(-1)^{k+1}A_k^{(m)}\sin^{2k}\varphi\right),$$

where $B_i^{(m)}$, $A_k^{(m)}>0$ and are constant.

To assess the stability of this disclination configuration we must investigate the sign of $\partial^2/F_d/\partial\xi^2$, where ξ is a small displacement of the disclination. If $\partial^2F_d/\partial\xi^2>0$ in the vicinity of each disclination, this means that F_d of the given configuration has a minimum and the configuration is stable. At $\partial^2F_d/\partial\xi^2<0$ the configuration is unstable.

It is convenient to consider the displacements in θ and φ, i.e., $\xi_1=\sin\theta$, $\xi_2=\sin\varphi$. For $n\neq1$ we have $\partial^2F_d/\partial\xi_1^2<0$, and $\partial^2F_d/\partial\xi_2^2<0$, in the disclination region by virtue of the alternating signs of the terms in F_d.

All the disclination configurations corresponding to solutions $\mathbf{N}_{mn}$ with $n\neq1$ are thus unstable, as are superpositions of such configuration. The only stable solution is $\mathbf{N}_{mn}$ with $n=m=1$. In this case $F_d=\sin^2\theta\sin^2\varphi$, there are two disclination of strength $+1$ and $\partial^2F_d/\partial\xi_1^2>0$, $\partial^2F_d/\partial\xi_2^2>0$, i.e., two stable disclinations.

RESULTS OF EXPERIMENTAL INVESTIGATIONS OF DISCLINATION DYNAMICS ON SPHERE SURFACES

a) Symmetric configuration of disclinations on the surface of a sphere with an arbitrary number of disclinations

Realization of such a disclination configuration became possible by sublimation of impurity particles trapped by the poles of the spheres. Vanishing of the impurity particles produces on the poles, as a rule, a pair of negative disclinations with indices $-1/2$, -1, or -2; one of the possible configurations listed in the table becomes correspondingly possible. One example of such configurations, $\mathbf{N}=\mathbf{C}_{12}^0$, consisting of four $(+1)$ and two (-1) disclinations, is shown in Fig. 2a. Spontaneous symmetry violation, caused for example by fluctuations of the NLC, leads to instability of the entire disclination system as a whole, and to near-annihilation of two pairs of disclinations with opposite indices. In the upshot only one pair of disclinations of like sign remains on the sphere. This interaction is illustrated in detail in Figs. 2a and 3a.

b) Interaction of spheres with different disclination orientations

To assess the general laws that govern the dynamics of disclination structures on spheres, investigations were made of interactions between two hedgehogs, two spheres with planar boundary conditions, and a hedgehog and a sphere with planar boundary conditions. The sphere were brought into contact either by mechanical collisions or by thermally induced changes of their radii. All the possible configurations of the initial disclinations on the spheres and the results of their interaction are illustrated by motion-picture photographs b–f of Fig. 2 and c and d of Fig. 3. The mechanism of the first stage of sphere coalescence is common to all these configurations: At the instant when the spheres coalesce or are joined by a bridge, two disclinations with negative index (-1) are produced. The subsequent dynamics of the disclinations depends on the disclination configuration produced at the instant of sphere coalescence. For interacting spheres with hedgehogs there can be produced either a sphere with a hedgehog (see Figs. 2b and 3b), or a sphere with two disclinations on the surface. In the the first case one hedgehog emerges to the surface via interaction with one of the surface disclinations. The hedgehog breaks up near the surface into two positive surface disclinations,[6] which are annihilated by the two negative disclinations produced at the instant of sphere coalescence. In the second case (Figs. 2c and 3b), both hedgehogs emerge to the surface, and the end result of the disclination reaction is a sphere with two dis-

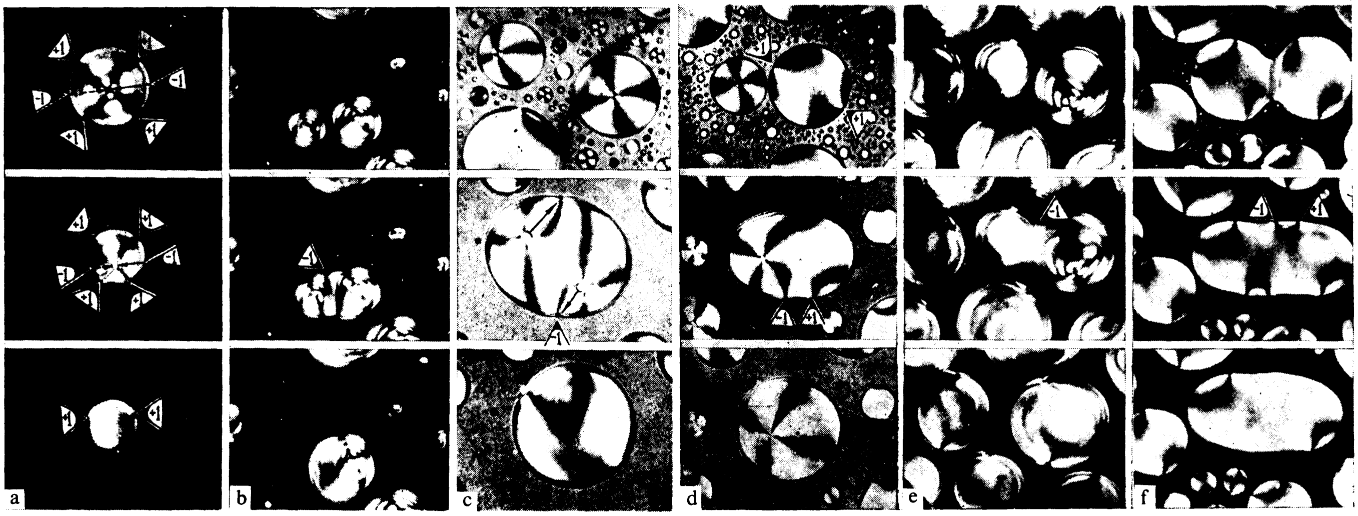

FIG. 2. Dynamics of disclinations in interactions between spheres: a) Temporal instability of sphere with four (+ 1) and two (− 1) disclinations on the surface. The first frame corresponds to the instant of vanishing of the impurity particles in the poles of the sphere and production of two negative disclinations. b, c) Interaction of spheres, with hedgehogs at the center. d, e) Interaction of hedgehog and sphere with two disclinations on the surface. f) Interaction of two spheres with two disclinations on the surface. The disclination indices are marked on the figure by numbers, and the approach directions for annihilation are marked by arrows.

clinations on the surfce.

Let us consider the interaction of a hedgehog with a sphere having two disclinations on the surface. Here, too, two manners of sphere interaction are possible. The net result is either two produced spheres with two diclinations on the surface, or a hedgehog. In this case the processes described for hedgehog interaction (Figs. 2,d–e; 3c): the hedgehog either emerges to the surface or vanishes on annihilation with two disclinations on the surface. The result of these processes is illustrated in Figs. 2d,e.

We consider finally the interaction of spheres with two surface disclinations. This process leads only to formation of spheres with two disclinations on the surface (Figs. 2f and 3d). It is a partial stage of an interaction of the hedgehog-hedghog and hedgehog-sphere type, with two surface disclinations. The processes in the dynamics of the disclinations and their annihilation are illustrated in Fig. 2f. The instantaneous disclination configuration produced at the instant of sphere coalescence depends on the orientation of the symmetry axes of the spheres in space. When the axes of the spheres with two disclinations on each surface are strictly parallel, a symmetric disclination configuration is produced with four disclinations on the equator with Frank index (+ 1) and two disclinations with Frank index (− 1) at the poles. The subsequent dynamics of the disclinations is similar to that shown in Figs. 2a and 3a. All the pictures considered are derived from a disclination configuration of the type $\mathbf{N} = \mathbf{C}_{12}^{0}$ as a result of violation of its symmetry in the fields on change of the boundary conditions. We note that in the absence of external fields the final state of the sphere and the coalescence variants described above (Figs. 2 and 3) depend on the boundary conditions and on the spatial arrangement of the defects at the instant when the spheres come in contact.

c) Investigation of temporal characteristics of disclination dynamics on spheres

It follows thus from the experimental investigations of the disclination dynamics that violation of the symmetry of one of the configurations with more than two disclinations leads to annihilation of a number of disclinations and to conservation, in the upshot, of only two disclinations on the surface or of a hedgehog at the center of the sphere. The speed of the processes is determined by the rate of approach of two disclinations of opposite sign. The law governing the variation of the distance between cores was theoretically estimated by Dreĭzen and Dykhne[9] and was experimentally verified in Ref. 10. It was established that the distance between the approaching disclination cores varies in this like $(t - t_c)^{1/2}$, where tc is the disclination-annihilation time. Reduction of the motion-picture photographs shown in Fig. 2 demonstrated strict agreement with this distance variation in disclination interactions on sphere surfaces (Fig. 2f), and interaction of the hedgehog-surface disclination type. The results of these investigations are illustrated in Fig. 4, together with data on the dynamics of disclinations on a flat surface of a semi-infinite nematic crystal. It is important to note that the constants a in the relation $r^2 = at$ are the same in all the situations considered.

Let us consider the coalescence of two viscous drops without allowance for the disclinations on their surface. A problem of this type for drops of equal radius R was apparently first considered by Frenkel'.[11] The law he obtained for the variation of the contact bridge of the coalescing drops was $x^2 \approx bt$, where $b = \sigma R / \eta$. For drops of unequal radius, $R_1 > R_2$, only the constant b changes and is equal to

$$b = (\sigma/\eta) R Q^{-1}(R_1, R_2),$$
$$Q = 1 + (R_2/R_1)^3 + (R_1/R_2)^3 + (R_2/R_1).$$

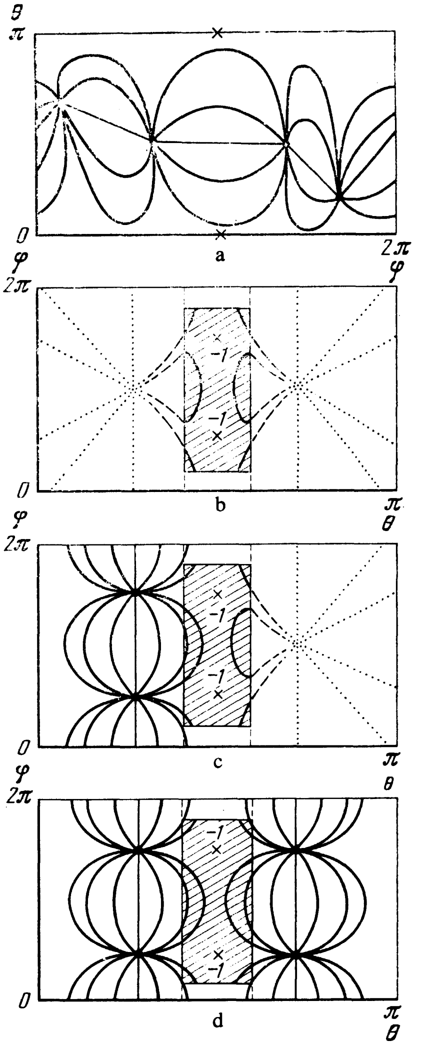

FIG. 3. Director-distribution maps: a) for loss of equilibrium of the disclination configuration corresponding to the solution C^0_{12}; b) for coalescence of two hedgehogs; c) for coalescence of a hedgehog with a sphere with two disclinations on the surface. The shaded regions in Figs. b–d correspond to a contact bridge; the projections of the director on the sphere surface in the case of hedgehogs are arbitrarily marked by dashes of various lengths.

Curve 4 of Fig. 4 corresponds to the variation of the sizes of the contact necks (or to the distance between the drop centers), and leads to two important conclusions. First, this dimension varies like $t^{1/2}$; second, $b \gg a$. This means that the presence of disclinations on the surfaces of the spheres does not influence significantly the character of the coalescence, so that the spheres themselves can be regarded in the calculations as isotropic. In the framework of this approximation, the straight line 3 of Fig. 4 corresponds to calculation in accordance with the Frenkel' theory.[11] This conclusion is corroborated by the agreement, within experimental error, between the calculated and experimental data.

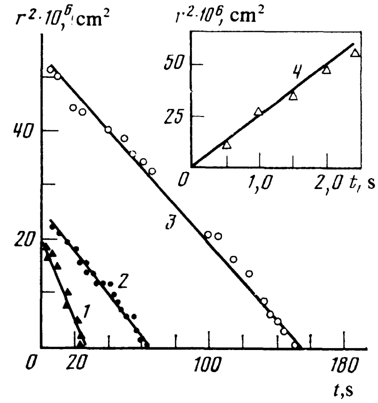

FIG. 4. Dependence of the square of the distances between the disclination cores on the time in accordance with Figs. 2b–2f: 1—in interaction of a hedgehog with a surface disclination; 2—in interaction of two surface disclinations; 3—in annihilation of disclinations on the surface of an "infinite" NLC: 4—change of contact bridge in coalescence of two drops (in the inset).

CONCLUSION

Many various symmetric configurations of disclinations can thus be realized on a spherical surface. The sum of the disclination indices on the spheres is always equal to the Euler characteristic of the surface, i.e., to two. Global stability is possessed by a configuration with two disclinations, so that ultimately coalescence of the spheres leaves only two disclinations. This answers the question why a state with homogeneous director orientation remains after phase transitions in large volumes of nematics. The result is of importance for the understanding of the processes whereby certain materials with specified properties, such as graphite, are obtained from NLC.

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