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# Symmetry-breaking and symmetry-recovering occurring during coalescence of nematic droplets with a bipolar structure

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We have directly observed the coalescence process of elementary nematic droplets having a bipolar structure. The coalescence results in a symmetry-breaking of the elementary droplet and then creation of new defects at the surface of the coalesced droplet. The annihilation of the defects with opposite signs and the shape change result in symmetry-recovering and finally formation of a large droplet that has absolutely the same structure as the former elementary droplets.

For nematic liquid crystals consisting of rod-like molecules, a unit vector **n**, having a characteristic of $\mathbf{n} = -\mathbf{n}$ because the rod-like molecules have no preferred polarity, is introduced to describe the orientational order in the nematic phase. Its mean orientation is defined as an order parameter that is non-zero in the nematic state, but that vanishes in the isotropic phase for symmetry reasons. This implies that the nematic phase has a lower symmetry than the isotropic phase [1]. Therefore, the isotropic–nematic transition is a symmetry-breaking phase transition which can create topological defects in the nematic phase [2]. Experimental results concerning the defect formation or defect annihilation dynamics in liquid crystals can be used to verify some theoretical models relevant to similar issues in cosmology and condensed matter physics [2, 3]. In this work, we report experimental verification concerning the formation of topological defects caused by symmetry-breaking when nematic droplets coalesce, and concerning the annihilation of the defect pairs in the coalesced droplets which results in symmetry-recovering of the droplet.

In the biphasic region of nematic liquid crystals, or when nematic liquid crystals are dispersed in an immiscible isotropic medium, they form droplets having certain special structures. Figure 1 (*a*) shows the configuration of a nematic droplet with a bipolar structure, which is one of the simplest but most important liquid crystal droplets [4]. In the 'one-constant' approximation†, the director components in cylindrical polar coordinates are taken as $n_r = -\sin\theta$, $n_\alpha = 0$ and $n_z = \cos\theta$, thus

$$\theta = \tan^{-1}\left(\frac{rz}{R^2 - z^2}\right) \qquad (1)$$

where $R$ is the radius of the droplet. The energy for this bipolar structure is

$$E \approx 5\pi k R \qquad (2)$$

[5]. This droplet has a cylindrical symmetry in molecular orientation with respect to the axis connecting the two poles ($D_{\infty h}$) and the molecular orientation at the surface of the droplet is parallel to the surface (tangential boundary). Two surface point defects (boojums) with strength $s = +1$ distribute at the surface of the droplet at the bipolar region. The liquid crystal molecules are subjected to splay and bend deformations in this bipolar droplet. When viewed between crossed polarizers, the bipolar droplet shows a certain optical texture, as shown in figure 1 (*b*) showing the optical texture of a bipolar droplet having the same size and orientation as that in figure 1 (*a*). Two dark brushes emanate from the two point defects at the poles and the director of the unit vector in the areas occupied by the dark brushes is parallel to the analyser [4, 6, 7].

Before describing the present work with nematic droplets, we illustrate a mathematical theorem, the *Poincaré* theorem, which will be used for analysing in a precise way the number and the features of defects distributed at the surface of the nematic droplet. The *Poincaré* theorem indicates that the sum of the indices $m_i$ of the vector field fixed on the closed and smooth surface in the tangential boundary condition equals the *Euler* characteristic of this surface, i.e.

$$\sum_i m_i = E \qquad (3)$$

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†In the 'one-constant' approximation, $k_{11} = k_{22} = k_{33} = k$ where $k_{11}$, $k_{22}$ and $k_{33}$ are the Frank splay, twist, and bend constants, respectively.

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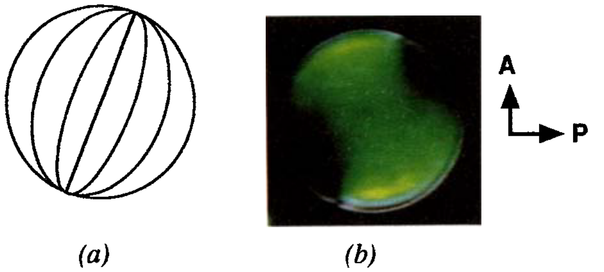

Figure 1. (*a*) Distribution of the director field in a nematic droplet with a bipolar structure; two surface point defects (boojums) localize in the bipolar region; (*b*) PLM micrograph of a bipolar nematic droplet having the same size and orientation as that in (*a*). The orientation of the crossed polarizers is indicated by the letters A (analyser) and P (polarizer).

where $E$ is the *Euler* characteristic of a two-dimensional surface [8–10]. In the case of a sphere-like volume surrounded by a smooth surface, the *Euler* characteristic $E$ is equal to 2 [8]. When the mathematical theorem is used for the nematic droplet, the indices $m_i$ of the vector field are the strengths $s_i$ of defects. For example, the *Poincaré* theorem indicates that only two surface point defects with $s = +1$ can distribute on the surface of the bipolar droplet at equilibrium, see figure 1 (*a*). In addition, in this work we refer to the droplet in figure 1 as the elementary droplet in order to distinguish it from the droplet formed by coalescence. The latter is referred to as the coalesced droplet.

We experimentally tracked the transient structure of the coalesced droplet by recording pictures of the nematic droplets with the bipolar structure before and after coalescence. The material we used was *N*-(4-methoxybenzylidene)-4-butylaniline (MBBA) with a purity of 98% which was obtained from Aldrich and used without further purification. The isotropic to nematic transition for this MBBA was determined to occur at 36°C.

The specimen containing the elementary bipolar droplets was prepared by sealing a very small amount of the material between two glass slides with epoxy resin. The

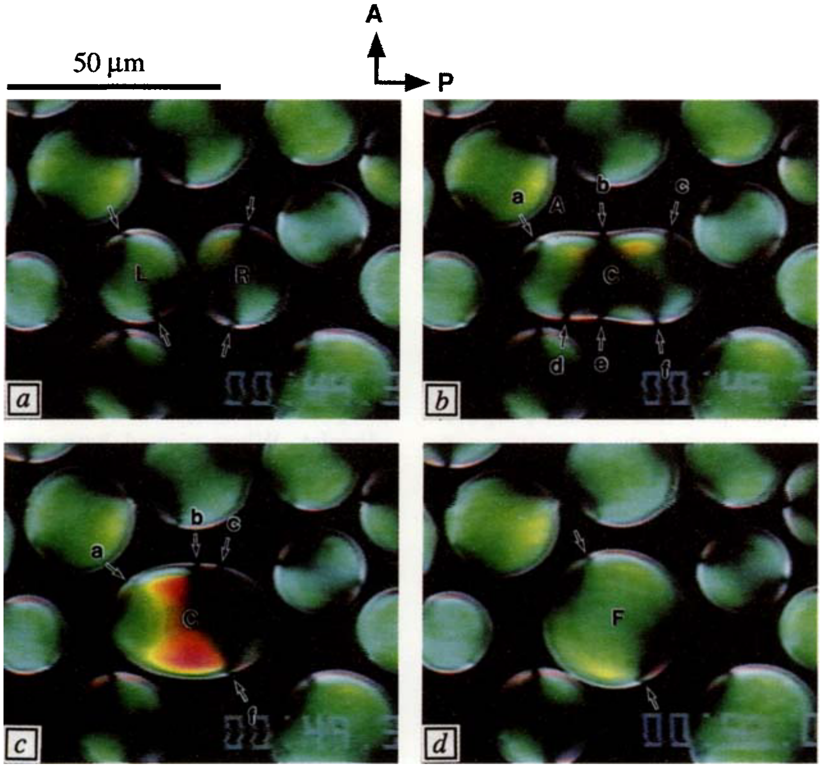

Figure 2. PLM micrographs showing the coalescence of two bipolar droplets and the time-evolution of the ordering process in the coalesced droplet in two-dimensional space. (*a*) Two elementary droplets denoted by the letters L (left) and R (right) before coalescence, the position of four surface point defects shown by arrows; (*b*) coalesced droplet C containing six surface defects, four positive indicated by arrows a c d and f and two negative denoted by arrows b and e; the negative defects were created by coalescence; (*c*) at this stage the defect pair (d–e) has annihilated and the two defects of the pair (b–c) have moved towards each other, meanwhile the coalesced droplet changes its shape to reduce surface tension; (*d*) the large elementary droplet F thus developed by coalescence has absolutely the same structure as the former small droplets. The images (*b*) to (*d*) were taken at 0·7, 3·0 and 25 s after taking the image (*a*). The orientation of the crossed polarizers is indicated by the letters A (analyser) and P (polarizer).

spacing between the two glass slides was 15 µm. We carefully controlled the amount of the material to be less than that needed to fill the sample chamber. In this case the liquid crystal formed some large domains. When the chamber was heated to 40°C, the domains became isotropic. When the test temperature was decreased from 40 to 29°C, many small droplets having the bipolar structure formed in the areas around the domains. In this case the matrix surrounding the bipolar droplets was air. They coalesced and then their size increased. The texture of the elementary and coalesced droplets was observed *in situ* by polarizing light microscopy (PLM) and was recorded by a CCD camera and a video recorder.

The PLM micrographs in figure 2 show the coalescence of two bipolar droplets and the time-evolution of the ordering process in the coalesced droplet in two-dimensional space. The corresponding change in the trajectory of the molecular director in the elementary droplets and the coalesced droplet is approximately shown in figure 3. Figure 2(*a*) shows the birefringence pattern of the elementary droplets with the bipolar structure. The dark area is the isotropic matrix (air). Here we focus on the two elementary droplets, denoted by the letters L (left) and R (right), having a size of *c*. 22 and 26 µm, respectively. The positions of the two point surface defects at the surface are indicated by two arrows in each droplet in figure 2(*a*) and the orientation of the two droplets is indicated in figure 3(*a*).

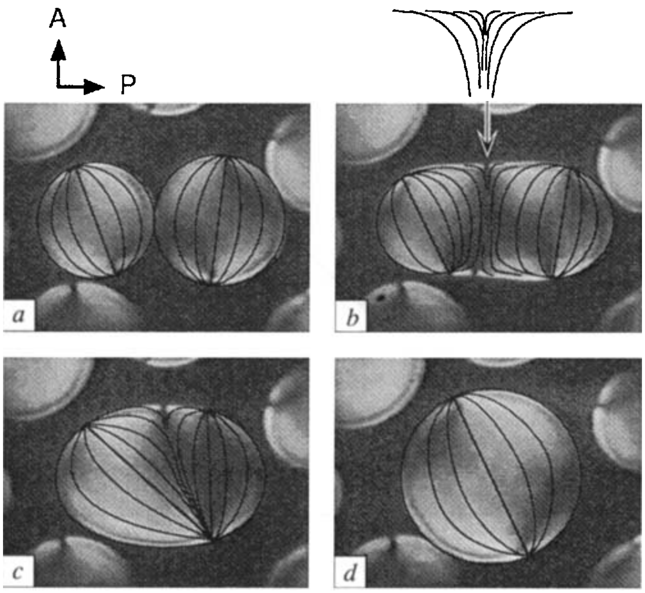

Figure 3. The trajectory of the molecular director superimposed on the elementary or coalesced droplets in figure 2. Thus micrographs (*a*) to (*d*) show the trajectory change caused by coalescence of the elementary droplets and by annihilation of defect pairs with opposite signs in the coalesced droplet. The trajectory of the elementary droplets in (*a*) and (*d*) was approximately determined on the basis of the relation between their optical textures and the distribution of the director field, as shown in figure 1(*a*), and that of the coalesced droplet in (*b*) and (*c*) according to the trajectory of the elementary droplets in (*a*) and (*d*) and the optical texture. For example, the director in the areas showing the dark brushes is approximately parallel to the analyser. The details of the trajectory around the negative defect indicated by the arrow in (*b*) are shown outside (*b*) [11]. The orientation of the crossed polarizers is indicated by the letters A (analyser) and P (polarizer).

Figure 2(*b*) shows the optical texture of the coalesced droplet (denoted by the letter C) formed by the two elementary droplets. The arrows labelled a, c, d and f point out the two pairs of point surface defects originating in figure 2(*a*). This coalesced droplet does not have any symmetry, as the former droplets have†. This is due to the difference in size of the two elementary droplets, their interior structure or the distribution of the molecular director and their relative orientation before coalescence. This symmetry-breaking causes a mis-match of the molecular director at the contact area and hence creates two new defects, as indicated by the arrows labelled b and e in figure 2(*b*). The newly created defects distribute at the surface of the coalesced droplet and the trajectory of the directors around them is shown in figure 3(*b*). According to the *Poincaré* theorem (equation 3), we can reason that the strength of each of the two newly created defects is $-1$, because there are four $s=+1$ defects at the surface of the coalesced droplet and $E$ is 2 for the coalesced droplet [8]. The feature of this negative surface defect is the same as that discussed by Meyer [11]. The defect pairs with opposite signs attract each other, move together on the surface and finally annihilate. From figure 2(*b*) to 2(*d*), we see that two pairs of defects, (b–c) and (d–e), gradually disappear from view, and from figure 3(*b*) to 3(*d*) we see the corresponding change in the trajectory of the directors. Figure 4 shows a double logarithmic plot of the distance $D$ between the defect pair (b–c) (in figure 2) versus $(t_0-t)$, where $t_0$ is the time of annihilation. In figure 4 the slope of the solid line is 0·5. Therefore, figure 4 shows further that the annihilation kinetics of the defect pair obey the scaling relation $D \sim (t_0-t)^{1/2}$ shortly before annihilation [12–14]. The data in the early stage deviate from the scaling relation due to the influence of the defects around the pair, as well as the shape change in the coalesced droplet. The coalesced droplet changes its shape to form a spherical droplet. Finally, a large

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†If the two droplets have the same size beforehand, the coalesced droplet can have a certain symmetry, but it is lower than $D_{\infty h}$, for example $D_{2h}$ in the case that their axes are parallel to each other.

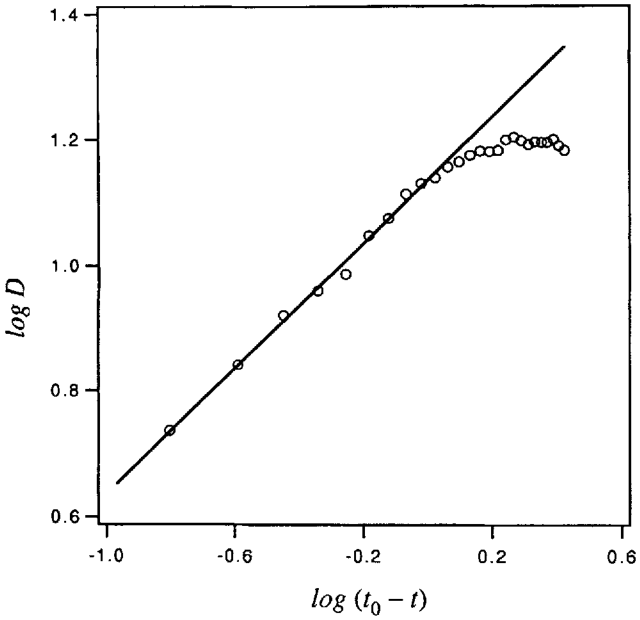

Figure 4. A double logarithmic plot of the distance $D$ between the defect pair (b–c) versus ($t_0 - t$), where $t_0$ is the time of annihilation. The slope of the solid line is 0·5, showing the scaling relation $D \sim (t_0 - t)^{1/2}$ [12–14].

elementary droplet containing only two $s = +1$ surface defects and having absolutely the same structure as the two former droplets is formed, as shown by the droplet F in figure 2 (*d*). This fact indicates that annihilation of the defects and the shape change in the coalesced droplet result in symmetry-recovery.

The experimental observation shown in figure 2 indicates that the following processes occur. In order to lower the interfacial area between the liquid crystal and the isotropic matrix and hence the interfacial tension, the elementary nematic droplets with a bipolar structure coalesce to form a large coalesced droplet. The coalescence causes the symmetry-breaking of the elementary nematic droplet, and then the new defects are created at the surface of the coalesced droplet. The properties (strength and sign) of the newly created defects are determined by the *Poincaré* theorem. Formation of the new defects in the coalesced droplet increases the free energy of the system. The excess of free energy is relaxed by the annihilation of the defect pair with opposite signs and by the shape change in the coalesced droplet. The large droplet finally formed has absolutely the same structure as the original droplets before coalescence, indicating a symmetry-recovering process. The *Poincaré* theorem gives a theoretical basis for the structure invariability of the bipolar nematic droplet before and after coalescence. Therefore, all droplets observed in a specimen should have totally the same structure, independent of their size, except for those undergoing the transient changes.

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