Defect-antidefect correlations in a lyotropic liquid crystal from a cosmological point of view

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In this work, we analyze the defect and antidefect distribution in the nematic calamitic phase of a lyotropic liquid crystal [the ternary mixture formed by potassium laurate (KL), decanol (DeOH), and water]. We obtain defects with wedge disclinations of strength $\pm 1/2$, and the scaling exponent determined by the defect-antidefect correlation was 0.29 ± 0.07. This value is in good agreement with the theoretical value of 1/4 obtained by the Kibble mechanism. The constant of the scaling relation of the defect and antidefect distribution is also discussed. We compare our results with the values obtained by Digal *et al.* [Phys. Rev. Lett. **83**, 5030 (1999)] who used a thermotropic liquid crystal.

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I. INTRODUCTION

Topological defects have appeared from a phase transition with spontaneous breaking of symmetry. These defects are commonly studied in cosmology [1] and also condensed matter systems [2,3]. For instance, in the cosmological case, cosmic strings are one-dimensional objects, which are formed when an axial or cylindrical symmetry is broken; such objects also occur in liquid crystals when a uniaxial symmetry is broken. In models of elementary particle physics (and cosmology), symmetry breaking is most often described in terms of a scalar field, the Higgs field [1,4]. In condensed matter physics, this field is the order parameter. It can also be a vector or tensor field [5].

Kibble, in 1976, was the pioneer in describing the formation of defects in the early Universe [6]. He showed that the description of defects depends only on the symmetry of the order parameter and space dimensions. As a consequence of the adiabatic expansion of the early Universe, the Kibble mechanism predicts that phase transitions can occur with spontaneous breaking of symmetry, producing domains with different configurations of minimum energy. The coalescence of these domains produces topological defects such as wall domains, cosmic strings, monopoles, and textures. Furthermore, many authors [7–11] have shown that it is possible to use condensed matter systems, such as superfluid helium [7], thermotropic liquid crystals [8–10], and, more recently, superconductors [11], as a good cosmologic laboratory. All these systems are analyzed by means of Kibble and Kibble-Zurek mechanisms in accordance with each type of phase transition.

Lyotropic liquid crystals are organized molecular systems formed by amphiphilic molecules in a solvent (usually water). The amphiphilic molecules can self-assemble in superstructures called micelles. The phase transitions occur as a function of temperature and/or pressure as well as of the change in the relative concentration of the compounds [12]. These systems are richer than thermotropic ones due to the freedom of choice of parameters to obtain the defects. We verified that in lyotropic liquid crystals the defects are more stable than those in thermotropic liquid crystals. In the last ones the defects quickly disappear.

In this work, we utilized a nematic calamitic phase of a lyotropic liquid crystal (KL-DeOH-water) as a new branch to analyze topological defects [13]. In comparison with the primordial Universe, an investigation is proposed describing defects and antidefects formed in the lyotropic liquid crystal, and we also compared our results with the values obtained by Digal *et al.* [14] who used a thermotropic liquid crystal. Furthermore, the formation of a wedge disclination of strength $m = \pm 1/2$, experimentally obtained in a lyotropic liquid crystal, is presented. In order to analyze the formation and the observation of these defects, the polarizing optical light microscopy technique was used. Recently, topological defects (cosmic strings and monopoles) have been analyzed by Satiro *et al.* only by numerical simulation [15] for liquid crystals in general.

II. THEORY

In this work, we considered the same theoretical approach proposed in Refs. [10,14,23], but applied to lyotropic liquid crystals. The orientational order of micelles is represented by the director vector $\vec{n}$. For liquid crystals $\vec{n}$ and $-\vec{n}$ are equivalent states.

For a uniaxial nematic, the order parameter space $\mathcal{M}$ (manifold of all possible values of the order parameter which do not alter the thermodynamical potentials of the system) is a sphere of unit radius: any point on the sphere corresponds to a different orientation of the director $\vec{n}$. Since $\vec{n} = -\vec{n}$, any two diametrically opposite points on the sphere describe not just energetically equivalent states but rather indistinguishable states. This unit sphere with identified antipodal points is denoted $\mathcal{M} = \mathcal{S}^2 / \mathcal{Z}_2$ [16]. The space of ground-state nematic configurations $\mathcal{M}$ is given by $\mathrm{SO}(3)/D_\infty$, where SO(3) is the symmetry group of the disordered (isotropic) phase and D_∞ is the unbroken subgroup consisting of rotations about the molecular axis and 180° rotations about axes perpendicular to the molecular axis [2,9]. For $\pi_1(\mathcal{M}) = \mathcal{Z}_2$ we have stringlike defects as those analyzed in this paper. Therefore disclinations are formed by a spontaneous breaking of symmetry in liquid crystals when a transition from an isotropic phase [SO(3)] to a nematic phase [O(2)] occurs. The director orientation changes by an angle of $2\pi m$ (Fig. 1) on going around the line (string), where parameter m labels the wedge disclination (or strength of disclination [16]). The director configuration around disclinations (string defects) can be de-

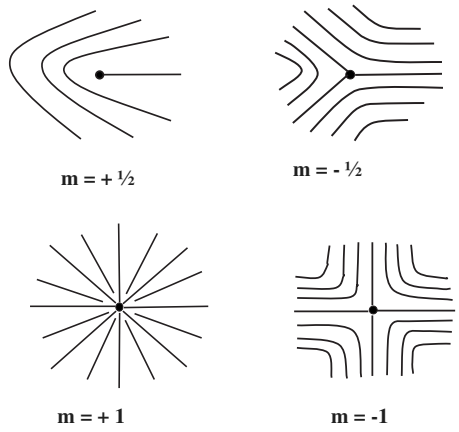


FIG. 1. Sketch of disclinations [14,16,17,20] with $m=\pm 1/2$ and ± 1 for $\theta_0=0$.

scribed by Frank's [17,18] free energy density as follows:

$$\mathcal{F}_V = \frac{1}{2}K_{11}[\nabla \cdot \vec{n}]^2 + \frac{1}{2}K_{22}[\vec{n} \cdot (\nabla \times \vec{n})]^2 + \frac{1}{2}K_{33}[\vec{n} \times (\nabla \times \vec{n})]^2, \quad (1)$$

where K_{11}, K_{22}, and K_{33} are splay, twist, and bend elastic constants (or Frank's constants), respectively. These constants are positive with dimension of energy times length^{-1} and are temperature dependent. Typical values for elastic constants are 10^{-11} N for both thermotropic [16,17] and lyotropic [19] liquid crystals.

The solution of Eq. (1) is

$$\theta = m\phi + \theta_0 \quad (2)$$

when the free energy is minimized by assuming one-constant approximation. This approximate solution, which is correct from the point of view of symmetries, assumes the z-axis perpendicular to the x,y plane. $\theta(x,y)$ is the angle between the director $\vec{n}$ and x, and $\phi(\vec{r})$ is the angle between $\vec{r}$ and $x(\phi=\tan^{-1} y/x)$. θ_0 changes from 0 to π [17,20]. In fact, the solution (2) restricts the nematic director to two dimensions (Fig. 2).

In 1976, Kibble proposed a theory [6] in which the phase transition proceeds with the formation of uncorrelated domains which subsequently coalesce, leaving behind defects. This mechanism hinges upon the fact that, during a cosmological phase transition, any correlation length is always limited by the particle horizon (maximum distance over which a massless particle could have propagated since the time of the Bang) [21]. This defect formation process is known as the Kibble mechanism. This coarsening happens due to the following processes [22]:

(1) string intercommutation: where two strings can exchange partners at a crossing point and

(2) oscillating loops which emit radiation and eventually shrink away.

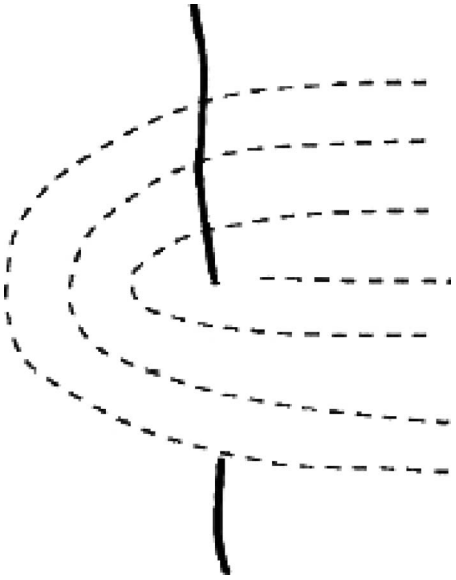


FIG. 2. Sketch of the string formed in the nematic phase.

This mechanism of the formation of the topological defect is based on two postulates: (1) At a phase transition the Universe is broken up into domains of broken symmetry phase, and the order parameter randomly varies from one domain to the other. (2) The order parameter, between two domains, interpolates following the shortest path on the vacuum manifold (order parameter space). This is called the geodesic rule [23].

In general, the Kibble mechanism in cosmology is based on topological ideas and is very difficult to analyze due to experimental restrictions. However, to verify the Kibble Mechanism there are two specific theoretical predictions which can be checked [23]:

(1) density of defects per domain and
(2) Strong correlation in defects and antidefects.

In this work, we followed the second prediction of the Kibble mechanism, which was proposed for the first time by Digal *et al.* [14]. In this way, we employed the following scaling relation for σ:

$$\sigma = CN^{\nu}. \quad (3)$$

Here, N is the number of defects in region Ω of area A, C is the constant of the proportionality (defined by strength of disclinations m), σ represents the width of the distribution which depends on the correlations of the defects and antidefects [24], and ν is the scaling exponent (determined by the defect-antidefect correlation). For instance, the exponent ν is $1/2$ for the uncorrelated case. In contrast, $\nu=1/4$ when the Kibble mechanism is obeyed. This value has universal nature describing other physical systems with the same symmetry of order parameter and space dimensions, as well as some systems for condensed matter. Digal *et al.* [14] obtained $\nu=0.26\pm0.11$ and $C=0.76\pm0.21$ for strength of disclinations $m=\pm 1$ (square lattice) analyzing stringlike defects using the thermotropic liquid crystal $K15$ (nematic phase). The theo-

retical value for C predicted by the Kibble mechanism is 0.71. In this work, we analyzed the formation of stringlike defects for strength of disclinations $m=\pm 1/2$ in a nematic calamitic phase of a ternary lyotropic liquid crystal.

III. EXPERIMENT

A. Sample

The lyotropic liquid crystal (LLC) investigated in this paper is formed by potassium laurate (KL), decanol(DeOH), and water. The composition in weight is 25.30% KL, 6.24% DeOH, and 68.46% water. The sequence of phases for this mixture determined by optical microscopy observations is I 13.5 °C N_D 17.6 °C N_{Bx} 18.7 °C N_C 36.6 °C I, where I is the isotropic phase, and N_D, N_{Bx}, and N_C are the discotic, biaxial, and calamitic nematic phases, respectively. In this work, the I-N_C phase transition was analyzed.

B. Experimental technique

In order to observe defects in the lyotropic liquid crystal, the polarized optical light microscopy technique was utilized. The sample was encapsulated in one flat glass microslide (0.2 mm thick, 2.5 mm wide, and 50 mm long) and was observed between crossed polarizers. The laboratory frame axes are defined as follows: x is the long axis of the microslide and z is the axis normal to the largest surface of the microslide. The optical microscopy observations were performed using a Leica ($5\times$objective) microscope [connected to a charge coupled device (CCD) camera] along the z-axis. The heating of the sample was controlled using a device called *hotstage* (INSTEC-HS1-I, accuracy of 10^{-3} °C) connected to a computer and cooled in a water bath (accuracy of 10^{-3} °C).

C. Production of defects

As mentioned above, the lyotropic mixture (Kl-DeOH-water) was encapsulated in a planar glass cell with a 200 μm light path. In order to avoid changes in the mixture concentration, the borders of the cell were sealed with a paraffin film followed by a nail polish coat. The cell was displayed in the *hotstage* at 50 °C. At this temperature the mixture shows an isotropic texture. After that, the temperature was adjusted to 30 °C, where a nematic calamitic phase (N_C) is expected. After about 20 h maintaining the temperature at 30 °C the desired defects were formed. Figure 3 shows a typical texture of the lyotropic mixture (obtained by the CCD camera) showing strength of disclinations $m=\pm 1/2$ and ± 1. In this picture $m=\pm 1/2$ (circle) is identified as points with two black brushes originating from them, while $m=\pm 1$ (rectangle) is a point with four black brushes. The brushes in Fig. 3 are regions where the mean micellar alignment is parallel to the plane of polarization of the incident light. It is interesting to note that for the lyotropic liquid crystal the defects with strength of disclinations $m=\pm 1/2$ occur most frequently.

In order to identify whether the disclinations are positive (defect, $m=+1/2$) or negative (antidefect, $m=-1/2$), we used the following procedure: we rotated the polarizer clockwise; some brushes rotated in the same direction (defects) and others in the opposite direction (antidefects). In this way, two pictures can be obtained with the sample between crossed polarizers: one without the rotated polarizer [Fig. 4(a)] and another with the rotated polarizer [Fig. 4(b)]. We compared these pictures and identified [Fig. 4(b)] defects with gray bullets (see circle) and antidefects with black bullets (see rectangle).

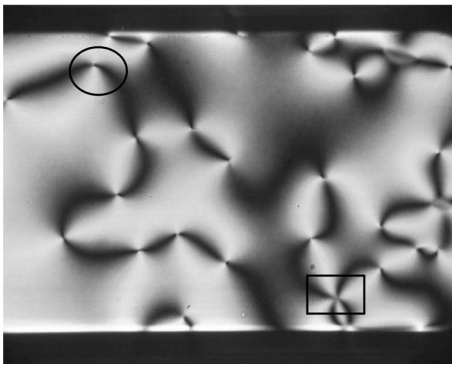


FIG. 3. Schlieren texture in a 200-μm-thick nematic film of a lyotropic mixture formed by KL-DeOH-water at $T=30$ °C. Strengths of disclinations $\pm 1/2$ are indicated by a circle and ± 1 by a rectangle. The points from which only two (or four) brushes emanate is located in a string (perpendicular to the plane).

D. Correlation between defects and antidefects

From the obtained data and by using the method described in the previous section, we employed a statistical method to investigate the correlation between defects and antidefects. The procedure was as follows:

(1) a maximum number of regions in each picture with N stringlike defects was selected and

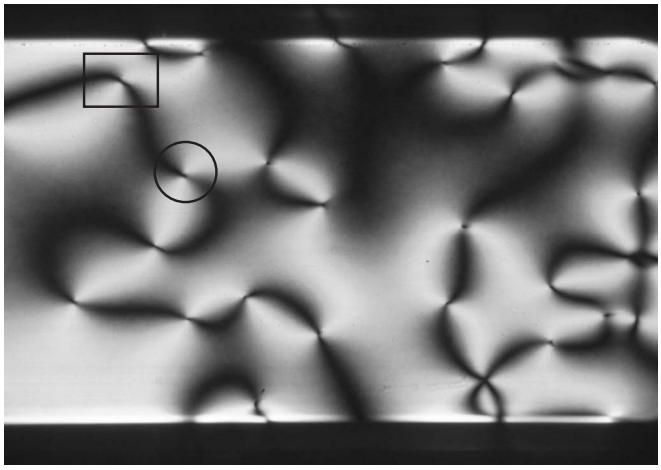
(2) the number of defects (gray bullets) and antidefects (black bullets) in each selected region was computed. The difference between the number of defects and antidefects is called Δn.

In order to correlate defects and antidefects (theoretically predicted by the Kibble mechanism), we have utilized the procedure described before for $N=10$, 14, and 18. In this process, we analyzed 28 pictures such as those shown in Fig. 3. For $N<10$ or $N>18$, it is not possible to obtain enough data to perform a statistical analysis.

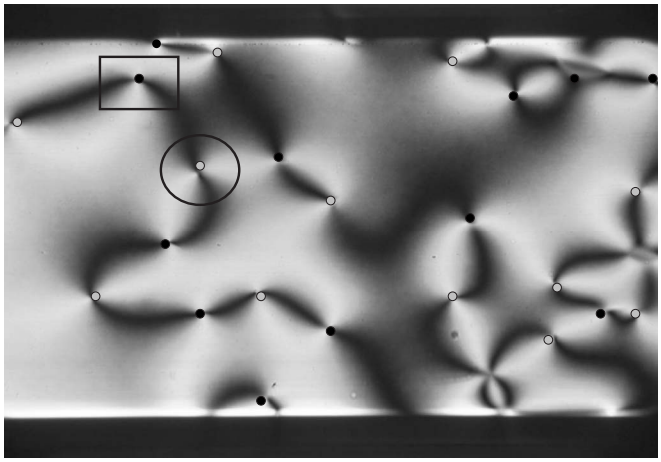
IV. RESULTS AND DISCUSSION

Figure 5 shows $f(\Delta n)$ versus Δn in the regions analyzed from the pictures. The lines are very good Gaussian fittings for $N=10$ (solid line), $N=14$ (dashed line), and $N=18$ (dotted line). Table I shows the values obtained from the Gaussian fitting.

Using the data from Table I, we obtained a good linear fit (Fig. 6). Notice that the maximum of the Gaussians is consistent with $\overline{\Delta n}$. The values for the width of the Gaussian



(a)



(b)

FIG. 4. Sequence of pictures showing the procedure utilized to identify defects (marked with a circle) and antidefects (marked with a rectangle) in textures of LLC. (a) This picture indicates the positions of the points before rotating the polarizer. (b) After rotating the polarizer clockwise, the black brushes rotated in the opposite direction (rectangle) are represented by black bullets—antidefects (−1/2). Those rotated in the same direction (circle) are represented by gray bullets—defects (+1/2).

distributions (σ parameter) are also shown in Table I. Notice that the Gaussian shape depends on the total number (N) of defects and antidefects. These values are strongly conditioned to the area of the sample analyzed and the different amounts of possible Δn in this region. We analyzed other quantities of possible distributions for Δn. For the points (Δn) concentrated around zero, the distribution is similar to a delta function. On the other hand, we have found some distribution with great dispersion due to the low frequency of Δn, for each Δn analyzed. These distribution cases do not contribute to our analysis.

Observing the $N=10$ case, we obtained $\sigma=0.65\pm0.01$. Comparing this value to the ones obtained by Digal *et al.* (1.41 ± 0.07) and the Kibble mechanism (1.26) we verified that these values are approximately twice as large as the value we obtained. The value of σ is correlated to the density of defects and antidefects in a specific region which reflects

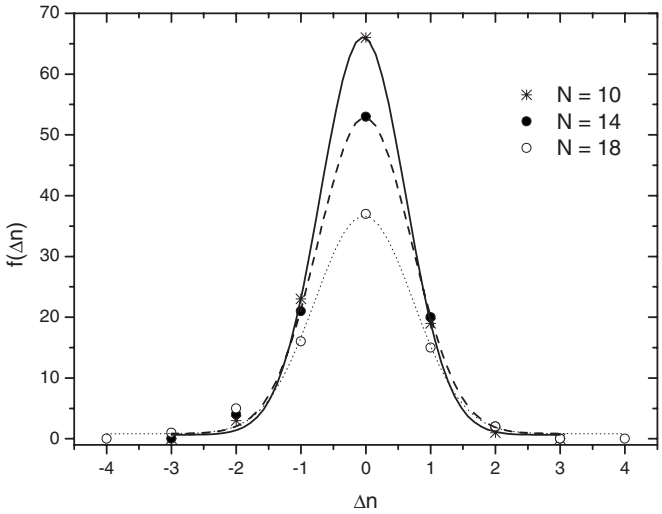


FIG. 5. Gaussian fitting for the frequency of occurrence of the difference between defects and antidefects $f(\Delta n)$ versus Δn. The fitting function is $f(\Delta n)=a\exp[-(\Delta n-\overline{\Delta n})^2/(2\sigma^2)]$, where σ is the variance of distribution and N is the total number of defects in each region.

the correlation in the production of defects and antidefects. The σ value we obtained can be associated to the nature of the sample (a mixture of amphiphilic molecules and water). In this micellar solution, the density of micelles is lower in comparison to the molecular density of the thermotropic liquid crystals.

We found the parameter ν (Fig. 6) to be 0.29 ± 0.07. This value is in good agreement with the theoretical value 1/4 from the Kibble mechanism for this correlation in the early Universe. In addition, our result for ν is also in agreement with that obtained by Digal *et al.* using a thermotropic liquid crystal ($\nu=0.26\pm0.11$).

From the values of σ, N, and ν, we obtained the value of the proportionality constant, C, as 0.34 ± 0.06. By using the thermotropic liquid crystal $K15$, Digal *et al.* [14] obtained $C=0.76\pm0.21$ for stringlike defects with $m=\pm1$ (points from which four brushes emanate). The theoretical value predicted by the Kibble mechanism is 0.71 for square-shaped elementary domains. When comparing our results to those obtained

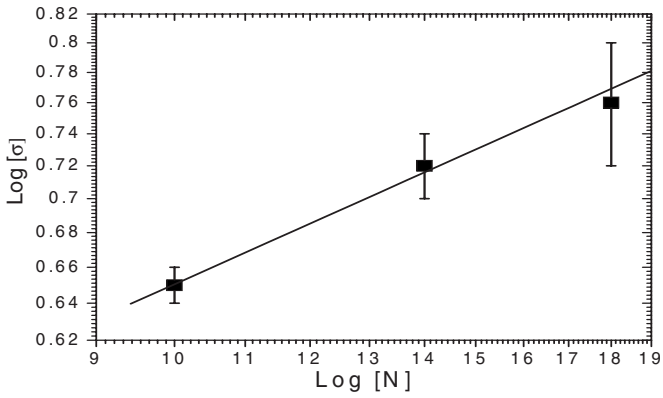


FIG. 6. A log-log graph to determine the exponent $\nu(\nu=0.29\pm0.07)$.

TABLE I. Values obtained from the Gaussian fitting for the results, where N is the total number of defects in each region, a the amplitude, $\overline{\Delta n}$ the mean of the difference between defects and antidefects, and σ the variance.

N	a	$\overline{\Delta n}$	σ
10	65.45±1.20	−0.04±0.02	0.65±0.01
14	52.02±1.51	−0.02±0.02	0.72±0.02
18	35.87±1.60	−0.03±0.04	0.76±0.04

by Digal *et al.* and by the Kibble mechanism, consistent with the σ values mentioned in the previous paragraphs, it is interesting to note that the theoretical value is approximately twice as large as the ones we obtained with $m=\pm 1/2$ (points from which only two brushes emanate) in the same-shaped elementary domains. From this fact we may say that the C value depends on the defect's strength of disclinations.

V. CONCLUSIONS

In this work, we presented some indications that LLC is a good cosmological laboratory. In comparison to thermotropic liquid crystals, the lyotropic mixture KL-DeOH-water has retained the optical textures for a much longer time (~6 h under the same physical conditions). This fact supplies good experimental conditions to analyze the formed disclinations. The mean-square-root equation used in this work reflects the correlation in the production of defects and antidefects. For instance, the exponent ν is $1/2$ for the uncorrelated case. On the other hand, $\nu=1/4$ when the Kibble mechanism is obeyed. We have verified that our experiment indicates the prediction of the Kibble mechanism, since the value of the exponent determined by the defect-antidefect correlations for a lyotropic liquid crystal is $\nu=0.29\pm0.07$. This value is in good agreement (including deviation) with the theoretical value $1/4$ from the Kibble mechanism for this correlation in the early Universe. In addition, our result for ν is also in accordance with that obtained by Digal *et al.* using a thermotropic liquid crystal $\nu=0.26\pm0.11$. As for the constant C, we obtained $C=0.34\pm0.06$ for $m=\pm 1/2$ and Digal *et al.* [14] found $C=0.76\pm0.21$ for $m=\pm 1$. Our value is approximately half the theoretical value $C=0.71$ predicted by the Kibble mechanism when $m=\pm 1$. We attributed this discrepancy to its dependence on the strength of disclinations (in both square-shaped elementary domains). Thus this value is not only dependent on the elementary domains of the defects, but also on the strength of disclinations. As for the value of σ, it is correlated to the density of defects in a specific region which reflects the correlation in the production of defects and antidefects. We observed that the σ value can be associated with the nature of the sample (a mixture of amphiphilic molecules and water). In this micellar solution, the density of micelles is lower compared to the density of molecules (dense network of defects) of the thermotropic liquid crystals. As far as we know, this is the first time that stringlike defects with strength of disclinations $m=\pm 1/2$ have been experimentally obtained on a large scale using a lyotropic liquid crystal.

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[1] A. Vilenkin, Phys. Rev. D **23**, 852 (1981); E. W. Kolb and M. S. Turner, *The Early Universe* (Addison-Wesley, New York, 1993); A. Vilenkin and E. P. S. Shellard, *Cosmic String and Other Topological Defects* (Cambridge University Press, Cambridge, England, 1994).

[2] N. D. Mermin, Rev. Mod. Phys. **51**, 591 (1979).

[3] O. D. Lavrentovich, Liq. Cryst. **24**, 117 (1998); *"Nematic Liquid Crystals: Defects,"* in Encyclopedia of Materials: Science and Technology (Elsevier Science Ltd., 2001) pp. 6071–6076.

[4] C. Quigg, *Gauge Theories of the Strong, Weak, and Electromagnetic Interactions* (Addison-Wesley, New York, 1983); R. Dominguez-Tenreiro and M. Quirós, *An Introduction to Cosmology and Particle Physics* (World Scientific, Singapore, 1988).

[5] R. Durrer, M. Kunz, and A. Melchiorri, Phys. Rep. **364**, 1 (2002).

[6] T. W. B. Kibble, J. Phys. A **9**, 1387 (1976); Phys. Rep. **67**, 183 (1980); Aust. J. Phys. **50**, 697 (1997).

[7] W. H. Zurek, Nature (London) **317**, 505 (1985); P. C. Dendry, N. S. Lawson, R. A. Lee, P. V. E. McClintock, and C. D. H. Williams, *ibid.* **368**, 315 (1994).

[8] I. Chuang, R. Durrer, N. Turok, and B. Yurk, Science **251**, 1336 (1991).

[9] M. J. Bowick, L. Chandar, E. A. Schiff, and A. M. Srivastava, e-print arXiv:hep-ph/9208233; Science **263**, 943 (1994).

[10] R. Durrer, e-print arXiv:astro-ph/0003363; R. Ray and A. M. Srivastava, eprint arXiv:hep-ph/0110165; Phys. Rev. D **69**, 103525 (2004).

[11] R. Monaco, J. Mygind, M. Aaroe, R. J. Rivers, and V. P. Koshelets, Phys. Rev. Lett. **96**, 180604 (2006).

[12] P. Toledano and A. M. Figueiredo Neto, *Phase Transitions in Complex Fluids* (World Scientific, Singapore, 1998).

[13] B. F. de Oliveira, M.Sc. thesis, Departamento de Física, Universidade Estadual de Maringá, Brazil, 2006 (unpublished).

[14] S. Digal, R. Ray, and A. M. Srivastava, Phys. Rev. Lett. **83**, 5030 (1999).

[15] C. Satiro and F. Moraes, Mod. Phys. Lett. A **20**, 2561 (2005); Eur. Phys. J. E **20**, 173 (2006).

[16] O. D. Lavrentovich and M. Kleman, *Chirality in Liquid Crystals*, edited by Heinz-Siegfried Kitserow and Christian Bahr (Springer, New York, 2001), Chap. 5.

[17] P. G. de Gennes, *The Physics of Liquid Crystals*, 2nd ed. (Clarendon, Oxford, 1993).
[18] G. Barbero and L. R. Evangelista, *An Elementary Course on the Continuum Theory for Nematic Liquid Crystals* (World Scientific, Singapore, 2000).
[19] T. Kroin, A. J. Palangana, and A. M. Figueiredo Neto, Phys. Rev. A **39**, 5373 (1989).
[20] S. Chandrasekhar, *Liquid Crystals* (Cambridge University Press, Cambridge, England, 1980); S. Chandrasekhar and G. S. Ranganath, Adv. Phys. **35**, 507 (1986).
[21] E. W. Kolb and M. S. Turner, *The Early Universe* (Ref. [1]).
[22] A. M. Srivastava, Pramana, J. Phys. **53**, 1069 (1999).
[23] R. Ray, Pramana, J. Phys. **53**, 1087 (1999).
[24] A. Rajantie, Int. J. Mod. Phys. A **17**, 1 (2002).