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Light-transmission study of coarsening in a nematic liquid crystal

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We have used light transmission to follow for four decades in time the annealing of defects generated by pressure jumps in a uniaxial liquid crystal. Two distinct scaling regimes for the light intensity as a function of time were observed: an early-time regime in which light propagates diffusively to the detector, and a late-time regime in which unscattered light dominates the signal reaching the detector. The measured values for the scaling exponent ν for the string density $\rho_s \propto t^{-\nu}$ are within 10% of the expected value $\nu = 1$ over the time interval 10 msec $< t <$ 100 sec.

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There is considerable interest in the dynamics of topological defects formed during a symmetry-breaking phase transition in fields as diverse as condensed-matter physics, particle physics, and cosmology [1]. Most of the theoretical and experimental work has been carried out for the case when the topological defects are two-dimensional structures [2–5], domain walls, such as those produced during the spinodal decomposition of a binary fluid. Systems for which the topological defects have a lower dimensionality have received considerably less attention [6–13]. Experimental work has primarily concentrated on studies of coarsening in two-dimensional nematic liquid-crystal films exhibiting stringlike [14,15] or pointlike defects [16–18]. We have studied the coarsening of three-dimensional defect tangles in a uniaxial nematic liquid [1,19] crystal. For uniaxial nematics the transition from the isotropic phase to the nematic phase involves the breaking of global SO(3) symmetry to O(2). The topological defects created by this symmetry breaking [20–22] include type-$\frac{1}{2}$ disclination lines belonging to the π_1 homotopy class, monopoles or hedgehogs belonging to the π_2 homotopy class, and texture defects belonging to the π_3 homotopy class. The defects that appear in greatest abundance after a phase quench are the type-$\frac{1}{2}$ disclination lines. These stringlike defects appear to control the coarsening dynamics of the defect tangle [19]. Arguments have been put forth that the string density ρ_s should scale with time as t^{-1} for three-dimensional defect tangles [8,13]. In previous work we have confirmed this prediction [1,19].

Here we report on light-transmission experiments which have allowed us to extend our coarsening studies by three decades in time. Using this technique we have been able to measure string densities as large as 3×10^4 mm^{-2}. For these experiments, one of the cyclohexylcyclohexanes, a nematic liquid crystal with a small optical anisotropy $\Delta n = 0.027$, was chosen in order to minimize problems associated with light scattering due to director field fluctuations. For the pressure jumps employed in these experiments, the isotropic to nematic transition takes place in less than 8 msec as determined from observing the time it takes for the transmitted light intensity to drop from maximum transmission (isotropic phase) to maximum opacity. The time at which the liquid crystal became maximally opaque was taken to be the starting time for our measurements of t. There are two regions in which the scaling of the transmitted light intensity with time can be modeled simply: an early-time regime, 10^{-2} sec $< t < 1$ sec, in which the light propagates through the cell diffusively [23] by a random-walk process and a late-time regime, $t > 3$ sec, when most of the light is transmitted through the cell without scattering and the Lambert-Beer absorption law can be applied. The scaling exponent ν for the string density as a function of time $\rho_s \propto t^{-\nu}$ is expected to be 1. The scaling exponent inferred from our early-time intensity measurements is $\nu = 1.13$ and for late times it is $\nu = 0.93$. These values are close to the expected scaling exponent of 1. A direct measurement, using optical microscopy, of the late-time disclination line density gave a scaling exponent $\nu = 1.10$.

The liquid crystal used in these experiments, *trans*-(*trans*)-4-methoxy-4′-*n*-pentyl-1,1′-bicyclohexyl (Merck, CCH-501 or ZLI-3005), was chosen for its low optical anisotropy. The difference between the extraordinary index of refraction $n_e = 1.496$ and the ordinary index of refraction $n_o = 1.469$ for this material is $\Delta n = 0.027$ which is a factor of 5 smaller than typical optical anisotropies for nematic liquid crystals. The liquid crystal was housed inside a heated pressure cell with an entrance window and an exit window which allowed one to observe the defect tangle via optical microscopy or to pass laser light through the sample in light-transmission studies. The windows were made of glass coated with homeotropic alignment material and the window apertures were 3 mm in diameter. The cell thickness was 450 ± 50 μm. The temperature of the liquid crystal was measured using a Chromel-Alumel (type K) thermocouple projecting into the cell. At atmospheric pressure the liquid crystal exhibited an isotropic-to-nematic phase transition at 37.3 °C, close to the listed value of 36.8 °C. Our measured pressure-temperature phase diagram of the material is shown in Fig. 1. Over the temperature range 37.3 to 40.9 °C the coexistence curve is linear and has the slope $\Delta P/\Delta T = 2.8$ MPa/K. In a typical operation the cell was heated to 38.8 ± 0.2 °C, putting the liquid crystal into the isotropic phase. The transition to the nematic phase was induced by increasing the cell pressure from 0 to 6.9 MPa (1000 psi).

Video microscopy was used to directly observe the defect tangle. The liquid crystal was viewed in transmission with an objective (Nikon, E Plan, 4×, 0.1 NA) with a deep depth of field so that disclination lines throughout the sample would appear in focus. The coarsening of the defects was continuously recorded with an HSV-400 (NAC, Inc.) video recorder at a rate of 200 frames/sec. For light-transmission studies the cell was removed from the microscope stage and mounted on an optical bench. Linearly polarized light, with 632.8-nm wavelength, from a 0.5-mW polarization stabilized He-Ne laser (Edmund Scientific, Uniphase, model no. 1508P-0) was passed through the cell. The laser light intensity was monitored by extracting some of the light via a 50-50 beam splitter placed between the laser and the sample cell. The intensity of the laser output and the light transmitted through the sample cell were both monitored with photodiodes (United Detector Tech., PIN-5DP). The photodiode outputs were amplified and recorded using a multichannel analog-to-digital converter with 12-bit resolution (Labmaster from Scientific Solutions).

The coarsening of the defect tangle as it appears via transmission microscopy is shown in the sequence of photographs in Fig. 2. The defect tangle consists primarily of line disclinations. Each picture shows a region about 500 μm across and is labeled with the time since the isotropic-to-nematic phase transition. Incident light does not propagate through the defect tangle, without scattering, until after 1 sec. Before this time, only diffuse light that has undergone multiple-scattering events passes through the sample cell. The frame at 3 sec shows numerous regions where the incident light penetrates the tangle without scattering, indicating the average mean free path is on the order of the cell thickness. In the frame at 10 sec the defect tangle has coarsened sufficiently that individual defect lines can be identified.

The line density as a function of time was measured by recording the optical microscope images with the HSV-400 video system. Figure 3 is a log-log plot of the defect density versus time obtained by averaging the values ob-

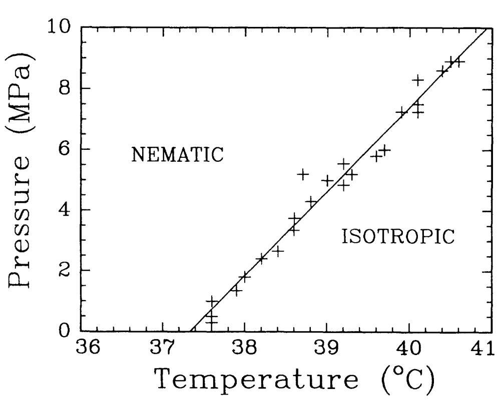

FIG. 1. The pressure-temperature phase diagram for the CCH-501 liquid crystal.

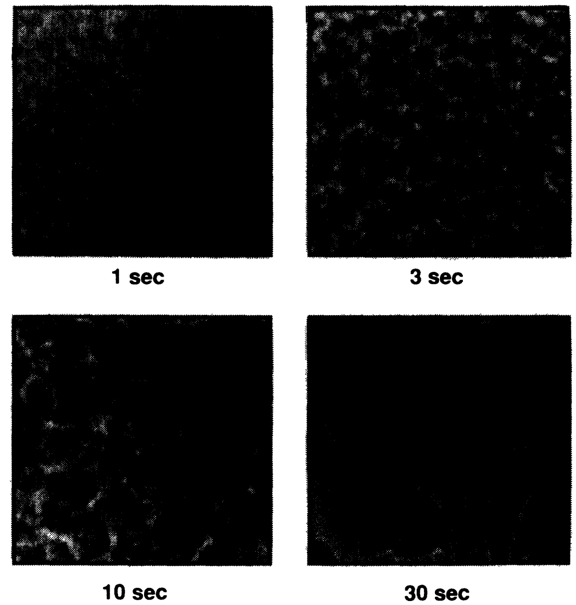

FIG. 2. A sequence of pictures of the defect tangle in the nematic phase. Each frame is labeled with the time after the phase transition occurred.

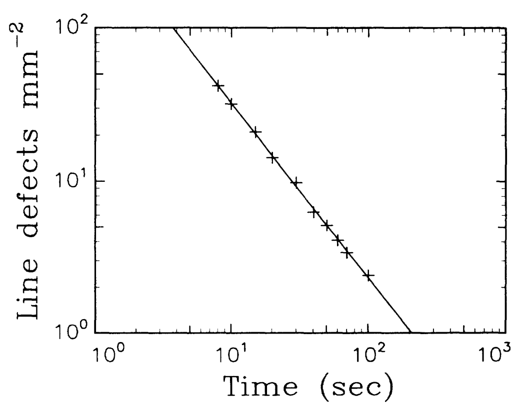

FIG. 3. String density (line defects per mm^2), as measured by optical microscopy, vs time on a log-log scale. Data for a pressure jump from 0 to 6.9 MPa and a temperature of 38.9 °C are shown. The solid line is a least-squares fit with a slope of −1.10.

tained from nine separate runs. The defect density plotted is the number of type-$\frac{1}{2}$ disclination lines per unit area crossing a plane through the medium. This quantity was measured by counting the number of disclination lines crossing each of several planes, each measuring 1.36 $\times 0.45$ mm^2 (the visible width of the video frame by the cell depth). This quantity is related to the defect density, measured in line length per unit volume, via a geometrical factor that depends on the details of the statistics of the string tangle. At a time of 10 sec there are about 20 or 30 lines per mm^2, whereas there are only 2 or 3 lines per mm^2 after the nematic phase has annealed for 100 sec. The least-squares fit to these data show the defect density, ρ_s, to scale as $\rho_s \propto t^{-1.10}$, which is close to the expected scaling of $\rho_s \propto t^{-1}$.

In order to study the scaling of string density at times earlier than 1 sec, we measured the intensity I of the laser light transmitted through the cell. The data are shown, plotted several different ways in order to indicate the two separate scaling regimes for the transmitted light, in Figs. 4 through 6. The data shown are the averaged results taken from nine runs. For these runs the cell temperature was 38.7 °C and the pressure jump starting from atmospheric pressure was 6.9 MPa (1000 psi).

The late-time scaling of the light intensity that occurs when most of the light is transmitted through the cell without scattering can be understood by postulating that the intensity I of light leaving the cell is given by the Lambert-Beer law

$$I = I_0 e^{-\alpha L}, \tag{1}$$

where I_0 is the initial light intensity, α is the amount of light per unit length that is scattered out of the light beam, and L is the path length through the cell. The light attenuation per unit length α is proportional to the defect density ρ_s. It then follows that $-\ln(I/I_0)$ is a measure of ρ_s,

$$\rho_s \propto -\ln(I/I_0). \tag{2}$$

Figure 4 shows $-\ln(I/I_0)$ versus time on a log-log plot where I_0 is the transmitted light through the annealed nematic phase. A least-squares fit over the time interval from 3 to 30 sec gives a line with a slope of -0.93. This is close to the value of -1.0, expected if the defect density scales as t^{-1}, shown by the dotted line. The value of the slope and the apparent behavior of $-\ln(I/I_0)$ are sensitive to the measurement of I_0. The deviation of the data from the scaling behavior, seen for times greater than 50 sec, is due in part from errors in determining I_0 and in part from a crossover to two-dimensional scaling [14,18].

Sensitivity to I_0 is considerably reduced by plotting $d[\ln(I/I_0)]/dt$ as a function of time. This quantity is proportional to $d\rho_s/dt$ and is thus expected to scale as t^{-2}. Figure 5 shows $d[\ln(I/I_0)]/dt$ plotted as a function of time on a log-log plot. The data were obtained by numerically differentiating the data of Fig. 4. The measured scaling of this quantity, over the time interval of 3 to 30 sec, is $t^{-2.08}$, implying that the string density scales as $\rho_s \propto t^{-1.08}$.

At early times, immediately after the phase transition, the incident light diffuses via a random walk through the liquid-crystal sample. When the mean-free-path length is

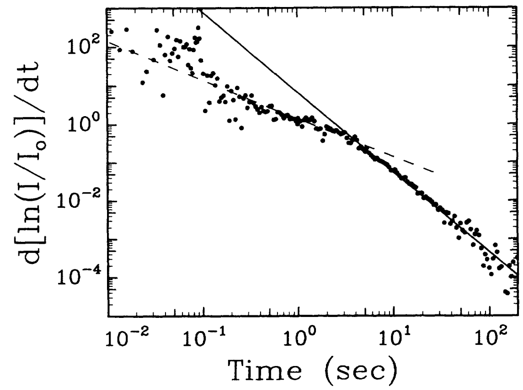

FIG. 5. The derivative of the data shown in Fig. 4, $d[\ln(I/I_0)]/dt$, vs time on a log-log scale. The solid line is a least-squares fit with a slope of -2.08 and the dashed line indicates the expected early-time coarsening, $I/I_0 \propto t$.

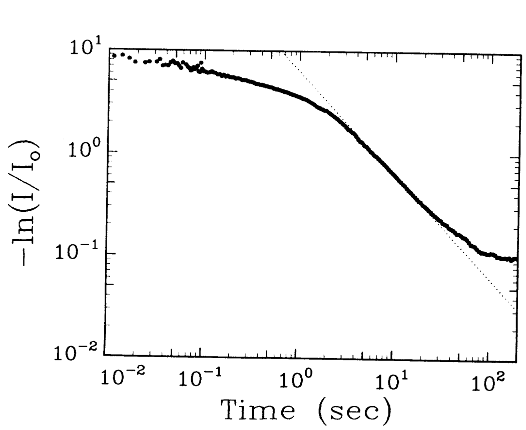

FIG. 4. The late-time coarsening of the defect density, measured by light transmission, shown as intensity vs time and plotted as $-\ln(I/I_0)$ vs time on a log-log scale. The dotted line has a slope of -1.0.

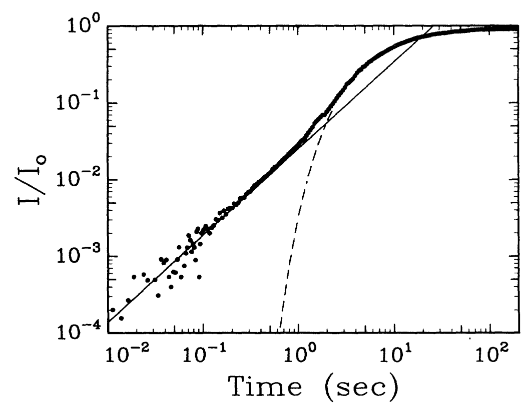

FIG. 6. The early-time coarsening, measured by light transmission, displayed as I/I_0 vs time on a log-log scale. The solid line is a least-squares fit with a slope of 1.13. The dashed line is a fit of $\ln(I/I_0)$ to the expected late-time dependence, $-at^{-1}$.

short compared to the sample thickness L most of the light diffuses back out through the entrance window. Assuming that there is negligible absorption of light by the medium, the intensity of the light that has diffused to the exit window is proportional to the mean-free-path length of the light in the medium [23]. Assuming that the mean free path is inversely proportional to the defect density, the intensity I of the light transmitted through the cell should be a measure of $1/\rho_s$ and should thus scale as t, that is,

$$I \propto 1/\rho_s \propto t \, . \tag{3}$$

This dependence is shown as the dashed line in Fig. 5 where $d[\ln(I/I_0)]/dt \propto t^{-1.0}$. This early-time dependence of light intensity can be seen more clearly in Fig. 6 in which I/I_0 is plotted as a function of time on a log-log plot. The solid line is a least-squares fit to the data over the time interval $10^{-2} < t < 1$ sec. The line has a slope of 1.13, indicating that the type-$\frac{1}{2}$ disclination line density scales as t^{-1} even at times within 10 msec of the phase quench. For comparison, the dashed line is a fit of the late-time coarsening, $\ln(I/I_0) = -at^{-1}$, with $a = 5.75$ a fitting parameter. The "crossover" region, from early- to late-time coarsening, is very short, spanning times from 1 to 3 sec.

We have studied the annealing of line defects by measuring the transmission of light through a uniaxial nematic liquid crystal. One of the cyclohexylcyclohexanes was chosen because of its small optical anisotropy. The defects were generated during the isotropic-to-nematic phase transition, induced by a pressure jump. The low optical anisotropy enabled us to detect the transmitted light even for times a few milliseconds after the phase transition. For early times the light intensity I scales as $I \propto t^{1.13}$, where $I \propto 1/\rho_s$ for a random-walk process and ρ_s is the string length per unit volume. For late times, with a mean free path large with respect to the cell thickness of 450 μm, the transmitted light intensity follows the Lambert-Beer absorption law, $\ln(I/I_0) \propto 1/\rho_s \propto t^{-0.93}$. These data show, for at least 4 orders of magnitude, that the scaling of the defect density with time is close to $\rho_s \propto t^{-1}$. Light transmission has thus proved to be a useful tool for probing the coarsening of nematic liquid crystals.

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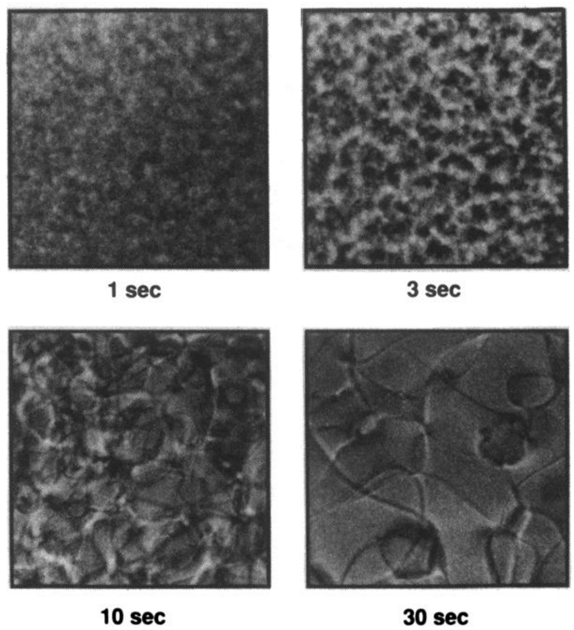

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