Scaling Behavior of Two-Time Correlations in a Twisted Nematic Liquid Crystal

N. Mason, A. N. Pargellis, and B. Yurke
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 11 September 1992)

We have measured the coarsening exponent ϕ and the nontrivial scaling exponent λ, which characterize how the two-time correlation function $C(t,t')$ scales with the correlation length $L(t)$, $C(t,t') \propto L(t)^{-\lambda}$ when $t \gg t'$, for a twisted nematic liquid-crystal system quenched from the isotropic phase to the nematic phase. This system is expected to be Ising-like with $\phi=0.5$ and $\lambda=1.25$. Our values, $\phi=0.515\pm0.026$, measured over two decades in time, and $\lambda=1.246\pm0.079$, measured over the late time period, 30 to 150 s, are in good agreement with theory.

PACS numbers: 64.70.Md

A system rapidly quenched from a high-temperature phase to a low-temperature phase via a symmetry-breaking phase transition generates topological defects. Following the phase quench, the density of these topological defects generally decays with time as the system anneals and, correspondingly, patches of the system become correlated over increasingly larger length scales. This coarsening process has been extensively studied for systems possessing a conserved scalar order parameter, such as systems exhibiting spinodal decomposition [1]. Recently there has been considerable activity in the study of coarsening processes in systems possessing nonconserved order parameters and stringlike or pointlike topological defects, both theoretical [2–6] and experimental [7–9]. The growth of the correlation length $L(t)$ as a function of the time t since the quench often asymptotically approaches a power law $L(t) \propto t^{\phi}$. The coarsening exponent ϕ is typically, although not always [10], $\frac{1}{2}$ for a system with a nonconserved order parameter. The exponent ϕ is known to be $\frac{1}{2}$ for the two-dimensional Ising model, as determined from theoretical arguments [11–13], numerical simulations [14], and experiment [11]. A second exponent of importance in characterizing the annealing is the exponent characterizing the asymptotic behavior of the two-time correlation function $C(t,t')=\langle\Phi(\mathbf{r},t)\Phi(\mathbf{r},t')\rangle$ where $\Phi(\mathbf{r},t)$ is the order parameter. When $t \gg t'$ this correlation function scales with the correlation length as $C(t,t') \sim L(t)^{-\lambda}$. This nontrivial dynamical scaling exponent was first studied by Fisher and Huse [15] in connection with spin glasses since it characterizes the decay of magnetization for such systems. It was argued by these authors, and supported with numerical work, that $\lambda=\frac{5}{4}$ for the two-dimensional Ising system. Recently there has been considerable theoretical activity directed toward calculating λ for systems other than the Ising system [16,17]. Most of this work has been restricted to the study of $O(n)$ models within the context of $1/n$ expansions. Work has also continued on the study of domain growth in random magnets [18] and Ising systems [19,20]. Bray and Humayun [21] argue that the two-time correlation function $C(t,t')$ should scale asymptotically as $L(t)^{-\lambda}$ even when the correlation length $L(t)$ does not follow a power-law growth, for example, due to the presence of pinning sites that slow down the growth of the correlation length. This scaling law is thus expected to be more generally valid than that for the growth of the correlation length. To our knowledge there have been no experimental measurements of λ reported in the literature. Here experimental data are presented which support a value of 1.25 for λ for two-dimensional Ising-like systems. Our data also support the expected value of 0.5 for ϕ.

Our system consists of a 20-μm-thick film of uniaxial nematic liquid crystal placed between two glass plates which have been treated to force the direction of molecular alignment (director field [22]) at the surface of a glass plate to lie parallel to the glass plate along a well-defined direction. The two glass plates are oriented such that the orientation of the director at one glass surface is orthogonal to the orientation of the director at the other glass surface. The director field must thus twist by $\pi/2$, either clockwise or counterclockwise, as one goes from one glass surface to the other. The clockwise or counterclockwise $\pi/2$ twist provides the two values of the order parameter that correspond to a spin pointing up or down in an Ising magnet. These two regions of the liquid crystal can be differentiated by their respective colors when viewed under crossed polarizers due to the circular dichroic nature of the liquid crystal [23]. The boundary or "domain wall" between adjacent patches of clockwise twisting and counterclockwise twisting liquid crystal consists of a type-$\frac{1}{2}$ disclination line [24]. The time evolution of the system, following a quench from the isotropic phase to the nematic phase, was recorded using video microscopy. The images were digitally processed to extract the two-time correlation function. The structure factor and the growth of the correlation length with time for this system has already been studied [24,25] using video microscopy techniques. Our work, however, presents the first experimental determination of the scaling exponent λ for any physical system.

The liquid crystal used was trans-(trans)-4-methoxy-4′-*n*-pentyl-1,1′-bicyclohexyl (Merck, CCH-501 or ZLI-3005). This material has an anisotropy in the index of refraction, $\Delta n=0.027$, that is low compared with values $\Delta n=0.05$ to 0.25, typically exhibited by thermotropic

liquid crystals. For sample cells of 20-μm thickness such a low anisotropy in the index is desirable to enhance the contrast between the domains of clockwise and counterclockwise twist when viewed through crossed polarizers [23]. The surfaces of the glass slides are treated by first degreasing and cleaning them. Then they are dipped in a 0.1% by weight solution of polyvinyl alcohol in distilled water. After drying, the slides are lightly buffed unidirectionally with a soft cloth to establish the direction of alignment of the liquid-crystal molecules at the glass boundary. Mylar spacers are used to define the thickness of the region of the sample cell where the liquid crystal is placed and the two (crossed) slides are epoxied together. The sample cell was placed in an oven composed of two aluminum plates with windows, resistive heaters, and a thin film thermocouple. The isotropic-to-nematic phase transition occurs at 37 °C and a nematic-to-smectic-A transition occurs at 29 °C. In a typical data run, the sample was heated to 41 °C and then quenched by directing air onto the oven, lowering the cell temperature through the transition temperature at a rate of 0.3 °C/s, and reaching a final temperature of 32 °C after about 50 s.

The time evolution of the clockwise twisting and counterclockwise twisting domains were recorded on videotape. The images were processed to enhance the contrast between the two types of domains. The procedure consisted of first plotting the distribution of pixel values, for a given video frame, in red-green-blue color space. The pixel density in the red-green-blue color space generally had two well-separated peaks. The images were then converted to black-and-white images by assigning the value "black" (-1) to those pixels associated with one peak in the pixel density and the value "white" $(+1)$ to the pixels associated with the other peak in the pixel density. Figure 1 shows a typical sequence of images, depicting coarsening, processed via the procedure described above. Each frame is labeled with the time since the phase transition. These images are of a sample region that is 1.56 mm on a side. The resolution is limited by the frame-grabbing software, which generates images that are (400 pixels)^2, each pixel representing a sample region 4 μm across. As can be seen, after having been quenched from the isotropic phase to the nematic phase, the liquid crystal consists of interpercolated domains which coarsen with time. As shown by Nagaya, Orihara, and Ishibashi [25], it is important that the two surfaces of the cell be aligned such that the director at the surface of one glass plate is orthogonal to the director at the surface of the other glass plate. The angle between the direction of alignment of the two glass surfaces in our samples was within 5° of 90°.

For a two-dimensional system with a nonconserved scalar order parameter one expects the characteristic length $L(t)$ to grow diffusively with time, $L(t) \propto t^{0.5}$. In order to determine the behavior of $L(t)$ with time for our system, data from five separate runs were analyzed and averaged to obtain the defect density (disclination line length per unit area) $\rho_l(t)$ as a function of time. This quantity should be inversely proportional to the correlation length, $\rho_l(t) \propto 1/L(t)$. The defect density was determined by measuring the total length of the disclination lines with a planometer and dividing this length by the area, 1.56 mm^2. Figure 2 is a log-log plot of the defect density, $\rho_l \propto 1/L(t)$, versus time. A least-squares fit through the data, obtained by minimizing the vertical residuals only since the error in the time after the quench is less than 0.2 s, gives a slope of -0.490 ± 0.006 for over two decades in time. Our value of the scaling exponent is

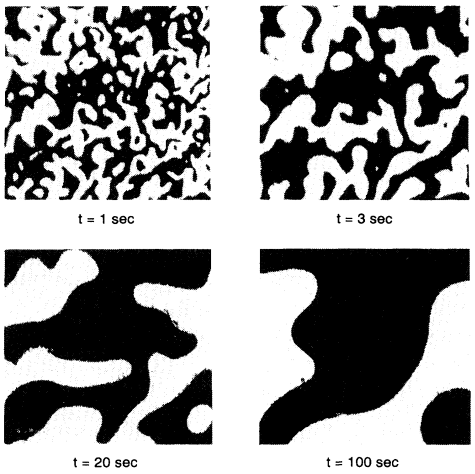

FIG. 1. A sequence showing the coarsening of domains of two types of twist in a twisted nematic liquid crystal as a function of time, in seconds, following an isotropic-to-nematic phase transition.

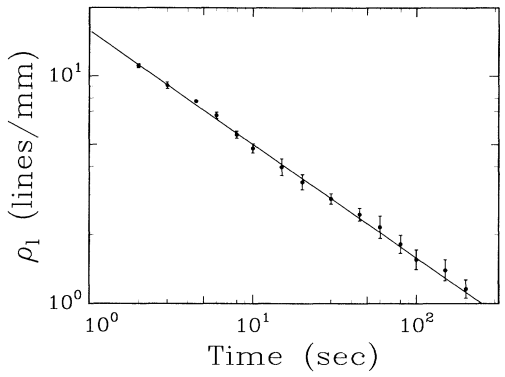

FIG. 2. A log-log plot of disclination line density, ρ_l, vs time. A least-squares fit gives a slope of 0.49, very close to the expected scaling of 0.5 for an Ising system, shown by the solid line. The error bars indicate the statistical uncertainty (standard deviation divided by the square root of the number of runs) determined from five runs.

larger than the value of 0.44 previously reported by Orihara and Ishibashi [24]. They suggest that the low value for ϕ is due to the fact that the sample cell continues to cool throughout the duration of a run. Dimensional analysis of the equation of motion [22,26] for the liquid-crystal director field shows that $K/\gamma \propto L^2/t$, where K and γ are the elastic and damping constants, respectively. A temperature dependent K/γ will thus affect the measured slope of $L(t)$ versus time. To determine the temperature dependence of K/γ we measured the collapse rate of circular disclination loops at different temperatures. From these data we conclude $\Delta\sqrt{K/\gamma}/\sqrt{K/\gamma}\Delta T = -0.01 \pm 0.01\ \mathrm{K}^{-1}$. In other words, $\sqrt{K/\gamma}$ varies by $(-5 \pm 5)\%$ over the 5 °C temperature change that occurs during the course of a given data run. Correcting our measured value for ϕ gives $\phi = 0.515 \pm 0.026$. This is close to the slope of 0.5, shown by the solid line, the behavior expected for a two-dimensional Ising system.

The two-time correlation $C(t,t')$ between the system at time t and at time t' was determined using the algorithm

$$C(t,t') = \langle \Phi(\mathbf{r},t)\Phi(\mathbf{r},t') \rangle = (1/N)\sum_j \Phi_j(t)\Phi_j(t') , \qquad (1)$$

where the sum is over $N = 400^2$ pixels and the parameters $\Phi_i(t)$ take on the values $+1$ and -1, depending on whether the pixel corresponds to a region of the sample with a clockwise or counterclockwise twist. In some of the runs, the temperature dependence of the circular dichroic effect made it impossible to analyze images occurring earlier than 2 s after the quench. During this early time, there is a thermal gradient across the field of view of 0.13 °C/mm as well as a rapid thermal change of 0.3 °C/s. The two-time correlation function with the initial time t' chosen to be 2 s will be denoted as $C(t,2)$. Figure 3 is a log-log plot of $C(t,2)$ versus time using data from the same five runs as before. A least-squares fit, again for vertical residuals, through the data of 30 through 150 s, gives an asymptotic slope of -0.582 ± 0.044. The solid line has a slope of -0.625, determined from the theory developed by Fisher and Huse [15] for the Ising model.

These data enable us to calculate the scaling exponent λ in two different ways. If the scaling for the growth of the correlation length as a function of time is $L \propto t^{\phi}$ and the scaling of the two-time correlation as a function of time is $C(t,2) \propto t^{-\nu}$, then the scaling exponent λ is given by $\lambda = \nu/\phi$. To be consistent, we obtained a value of $\phi = 0.475 \pm 0.028$ for a least-squares fit over the late time range, 30 to 150 s. Over this time interval, thermal corrections to ϕ are negligible. We did not take measurements at later times because the correlation length became comparable to the size of the largest blemish-free region. These blemishes provided pinning sites for the disclination lines which retarded the coarsening at late times. The measured values $\phi = 0.475$ and $\nu = 0.582$ thus yield $\lambda = 1.225 \pm 0.097$, which is consistent with theory. The exponent λ can also be extracted by directly plotting the logarithm of $C(t,2)$ against the logarithm of $\rho(t)$ as shown in Fig. 4. A least-squares fit to the data gives $\lambda = 1.246 \pm 0.079$. This result was obtained by calculating the slope and standard deviation of the slope for each individual run and then combining the results. The numerical data of Fisher and Huse [15] for $C(t,0)$ for the Ising model quenched from random initial conditions have been rescaled and drawn as the dashed curve in this figure in order to give a qualitative indication of how the approach to asymptotic behavior proceeds.

To summarize, we have measured the nontrivial dynamical scaling exponent λ for an Ising-like system. We observe the growth of the correlation length to closely follow that of an ideal Ising system and the value of 1.25 predicted by Fisher and Huse [15] is in good agreement with our measured value of λ.

We wish to acknowledge fruitful discussions with Da-

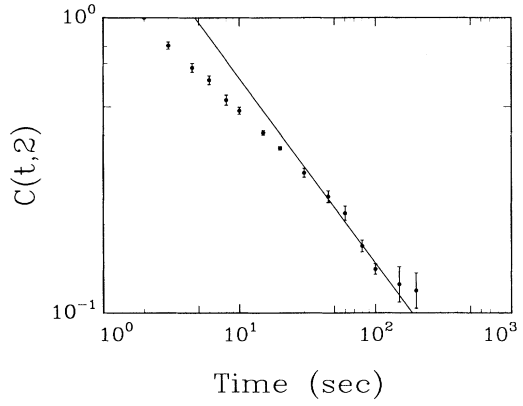

FIG. 3. A log-log plot of the two-time correlation function with time. The solid line has a slope of -0.625, the value obtained by Fisher and Huse [15]. The error bars indicate the statistical uncertainty determined from five data runs.

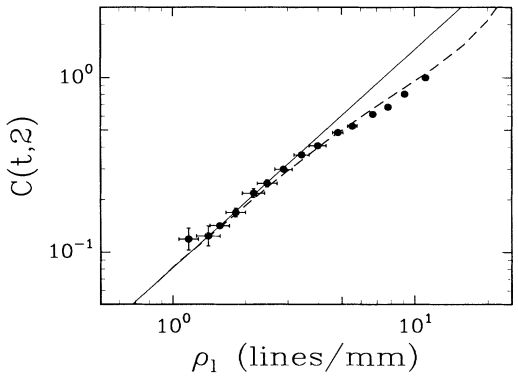

FIG. 4. A log-log plot of the two-time correlation function vs defect density, shown by the solid symbols. The solid line is the theoretically predicted asymptotic slope of 1.25 and the dashed curve is the (rescaled) numerical solution of Fisher and Huse [15] for $C(t,0)$.

vid Huse. We thank Pat Cladis for her technical assistance and Terry Kovacs for his assistance in developing the computer codes. We also thank Frank V. Allan for his help in obtaining the CCH-501 liquid crystal.

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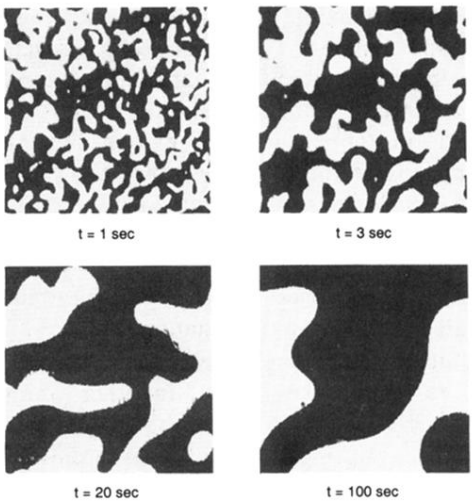

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