Annihilation rate and scaling in a two-dimensional system of charged particles

Won Gun Jang, Valeriy V. Ginzburg, Chris D. Muzny, and Noel A. Clark
Department of Physics, University of Colorado, Boulder, Colorado 80309-0390
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We study numerically the dynamic evolution of a two-dimensional system of charged particles which interact with a logarithmic potential, move with constant mobility, and annihilate on contact with an opposite charge. It is shown that in the diffusive regime, at high temperature where Brownian motion dominates, the number density decay is described by a power law with the exponent -0.55 ± 0.05, which is in agreement with theoretical result -0.5. In the deterministic regime, where motion is controlled by the interparticle forces, the exponent is -0.90 ± 0.05. A simple scaling hypothesis is suggested to explain this unusual exponent.

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INTRODUCTION

The study of topological defects has played an important role in liquid crystal (LC) physics, especially in the understanding of the symmetry and order of LC phases. This problem is particularly interesting in two dimensions. In freely suspended smectic-C LC films the director order is characterized by a two-dimensional orientation field, $\mathbf{c}(x,y)$ [1], and topological defects; in this case vortices ($\pm 2\pi$ point disclinations) in $\mathbf{c}$, can be visualized using depolarized reflected light microscopy (DRLM) [2] and, therefore, studied directly. The observation of spontaneous thermal fluctuations of such single vortices has been already carried out [3]. By transiently cycling a smectic film from the smectic-C phase to the smectic-A phase and back to the smectic-C phase, it is possible to generate arrays of such topological defects, which then coarsen under the influence of deterministic and Brownian forces [4]. In this paper we present a molecular dynamics computer simulation study of the coarsening process. In our simulation the topological defects are treated as dimensionless pointlike + or − charges. We take the deterministic interactions between defects to be those obtained from Frank elasticity: pair forces between particles i and j being $F_{ij} \propto 1/r_{ij}$, where $r_{ij}=|\mathbf{r}_i-\mathbf{r}_j|$ is the interparticle separation. The defects are assumed to move in a background medium with a constant mobility μ and are in contact with a thermal bath of temperature T.

There have been several recent theoretical and numerical studies of defect dynamics in related two-dimensional (2D) systems. Toussaint and Wilczek [5] studied annihilation kinetics of a 2D system with + and − pointlike defects, which annihilate in contact, but do not otherwise interact (pure diffusion regime). They showed that in the case of random particle distribution at the beginning, initial correlations slow down the annihilation process, and the particle density in the system decays accordingly to the power law $\rho(t) \propto t^{-\nu}$, with exponent $\nu=0.5$. This decay is significantly slower than the t^{-1} behavior, obtained for a system with random defect distribution in the mean-field approximation, where all spatial correlations are assumed to be insignificant at all times. At much longer times, when diffusion length becomes comparable with system size, and provided that not all particles annihilated at this time, initial correlations become insignificant, and mean-field decay rate, described by the power law with exponent $\nu=1$, is observed indeed. We will refer to these two regions as "intermediate" and "final" scaling regions, respectively.

In several studies [6–10] the time-dependent Landau-Ginzburg model was employed, analyzing defects (vortices in the XY model) as singularities of the orientation field. Mondello and Goldenfeld [6] analyzed the 2D XY system with continuous-symmetry conserved order parameter at zero temperature. They investigated the decay of defect density and found it to be described by the power law with exponent $\nu=0.75$. Yurke *et al.* [7] considered the same system and suggested that the topological singularities, which interact effectively with the $1/r$ force, are also subject to a drag mobility μ, dependent on the overall particle configuration, that is obtained by integration of the spin dissipation over the entire system area. The result (for a single vortex with core radius r_c on a film of radius r) is

$$1/\mu = 2\pi\eta_\phi \ln(r/r_c) , \tag{1}$$

where η_ϕ is the rotational viscosity.

This dependence of the drag mobility on system size r leads, upon replacing r with defect separation, in a multiparticle annihilating system, in which positional correlations are ignored, to the following time dependence of particle density [7]:

$$\rho(t) \propto t^{-\nu} \ln t, \quad \nu=1 . \tag{2}$$

The logarithmic factor in (2) is due to the effective distance dependence of the mobility for a pair of interacting defects; an exponent $\nu=1$ is the mean-field prediction for annihilation rate, obtained if the correlations are neglected. Thus, according to Ref. [7], in the case of a two-dimensional XY system, the logarithmic correction due to screening of viscous drag on a particle by neighboring particles is large enough to account for deviation from mean-field behavior in the case when deterministic forces

prevail over thermal diffusion. As was mentioned earlier, in our simulations we considered particles of two different signs having constant mobility, so no variation of the drag on a defect is introduced in the simulation, and the logarithmic corrections of Ref. [7] ought not appear.

In the present work, two particles of opposite signs were considered to have annihilated, if distance between them becomes less than r_c. Thus, the kinetic equation for particle density is:

$$\frac{d\rho}{dt} = -2\pi r_c g(t,r_c) v_c \rho^2 , \tag{3}$$

where $v_c = 2K\mu/r_c$ is the relative velocity of two particles separated by r_c, $g(t,r_c)$ is a pair correlation function for particles of opposite charge at time t at a distance r_c, K is the force constant, and μ is the single particle mobility. Equation (3), of course, suggests that at large t, when finite dimensions of the system become comparable with diffusion path and initial correlations are no longer a decisive factor (see Ref. [5]), $\rho(t) \propto t^{-\nu}$, $\nu = 1$. Nevertheless, this region (we will refer to it as a final scaling region) is not in fact observed in our simulations, because it happens at much later stage of the process.

The aim for the simulations described in this paper, therefore, is to analyze in detail the region of the intermediate scaling, when the power-law behavior with $\nu \neq 1$ occurs. Different conditions ranging from zero force constant and finite temperature to zero temperature and finite force constant, are analyzed numerically by use of Brownian dynamics.

SIMULATION DETAILS

The equation of defect motion is a Langevin equation;

$$\mathbf{r}_i(t+\delta t) - \mathbf{r}_i(t) = \mu \sum \mathbf{F} \delta t + \delta \mathbf{r}_{iG} = (D/k_B T) \left[\sum q_i q_j K \mathbf{r}_{ij} / r_{ij}^2 \right] \delta t + \delta \mathbf{r}_{iG} , \tag{4}$$

where $\mathbf{r}_i$ locates defect i, r_{ij} is a distance between defects i and j, $D = k_B T \mu$ is the defect diffusion constant, $\delta \mathbf{r}_{iG}$ is a random Brownian displacement of the ith defect, and q_i is the ith defect's charge (either $+1$ or -1). If the distance between two defects of opposite sign becomes less than r_c, these defects are considered to have annihilated and are removed from the system.

The simulation results are given in physical units relevant to the freely suspended film system. For example, r_c was assigned the value of 0.001 cm, the microscope resolution in the DRLM experiments. The initial particle distribution was random. The time step δt is chosen to be the time interval over which diffusive and deterministic displacement of a defect in an annihilating pair together amount to a maximum of r_c or less;

$$2\sqrt{2k_B T \mu \delta t} + K\mu \delta t / r_c < r_c .$$

As discussed above in the liquid crystal film experiments [1–4], the topological defect drag coefficient is related to the rotational viscosity of LC molecules, and molecular rotational viscosity itself is a function of temperature. However, for simplicity, we have ignored this T dependence so that defect mobility μ (which in the case of liquid crystals is an inverse function of their rotational viscosity) remains unchanged as T and D both approach zero. The elastic constant $K = 1.5 \times 10^{-12}$ erg for a three-layer-thick p-decyloxy benzylidene, p'-amino 2-methylbutyl cinnamate (DOBAMBC) film with a director tilt angle $\theta = 23.5°$, typical for freely suspended films well into the smectic-C phase [8]. For a smectic-C phase, $K \propto n \sin^2(\theta)$, where n is the number of layers in the film, so that K can vary over a wide range of values in a given film as θ changes. The defect mobility is taken to be $\mu = (D/k_B T) = 3.9 \times 10^6$ (cm/dyn sec), the value obtained from Eq. (1) for a three-layer-thick DOBAMBC film with LC rotational viscosity of 4.4×10^{-9} dyn sec/cm and system radius $R = 1$ cm [3].

Each run consisted of 20 000 time steps. The particles moved in a 0.1×0.1 cm^2 square area with periodic boundary conditions. The minimum image principle was used (each particle interacts only with those particles or particle images that are within the 0.1×0.1 cm square area with center at the particle position).

The dynamics can be expected to show a characteristic dependence on the parameters K, T, and μ in the diffusive and deterministic limits. In diffusive limit, e.g., we can produce two dimensionless combinations of particle density ρ, time t, initial density ρ_0, and diffusion constant D: $\Lambda_1 = (\rho/\rho_0)$ and $\Lambda_2 = (\rho_0 D t)$. We expect, therefore, to have some functional dependent $\Lambda_1 = f_{\text{dif}}(\Lambda_2)$ describing scaling properties of the system in this limit. For some region of Λ_2, this dependence can be a power law, and, according to Ref. [5], the exponent of this power law equals 0.5. In deterministic limit, the "effective diffusivity" $K\mu$ replaces D to yield $\Lambda_1 = (\rho/\rho_0)$, $\Lambda_2 = (\rho_0 K \mu t)$, and $\Lambda_1 = f_{\text{det}}(\Lambda_2)$. Again, for some region of Λ_2, this functional dependence is a power law. Specific features of this dependence are discussed in the next section.

RESULTS

The annihilation run configurations were analyzed for a variety of dynamical characteristics and statistical measures of defect motion and correlation. Figure 1(a) shows the number of remaining defects $N(t)$ vs t for initial numbers $N_0 = 200$, 500, and 2000 of randomly placed defects for force constant $K = 10^{-11}$ erg and temperature $T = 0$. These and subsequent $N(t)$ data are obtained from averaging over 30 runs of 20 000 time steps. For this choice of parameters, there is no thermal diffusion and, as was discussed previously, the characteristic system time scales as $1/N_0$. Figure 1(b) shows this scaling, $N(t)$ exhibiting similar behavior for all N_0 values and the three curves overplotting in scaled coordinates (N/N_0) vs $(N_0 t)$. This scaling behavior indicates that there is little dependence on system size in the simulations. The straightline, having a slope of 0.90 indicates the asymptotic power-law exponent of $N(t)$, $\nu = 0.90 \pm 0.05$.

Figure 2(a) gives $N(t)$ for $N_0 = 500$ and varying values

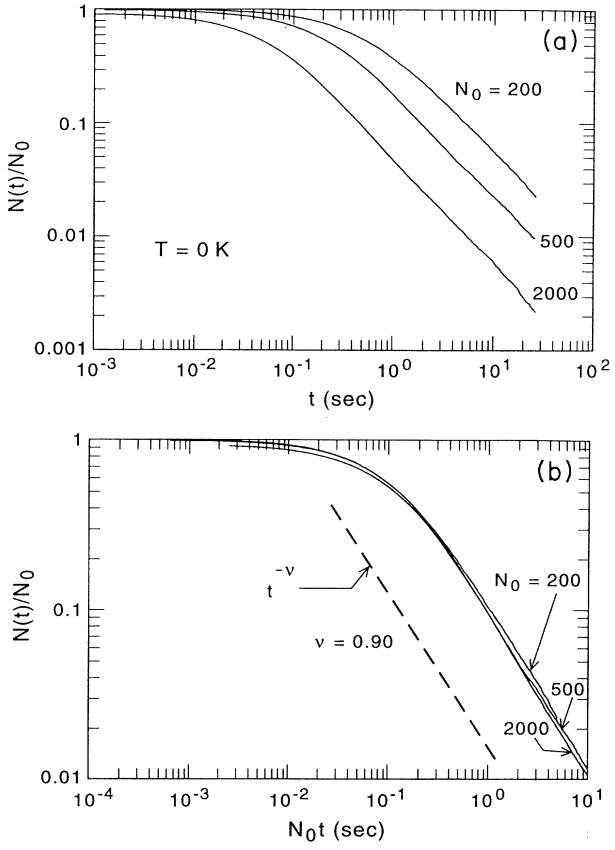

FIG. 1. Particle number as function of time for different initial densities ($N_0=200$, 500, and 2000) at $T=0$ K, $K=10^{-11}$ erg, unscaled (a) and scaled (b) plots. Decay power exponent $\nu=-d(\log N)/d(\log t)=0.90\pm0.05$. Dashed straight line has slope 0.90.

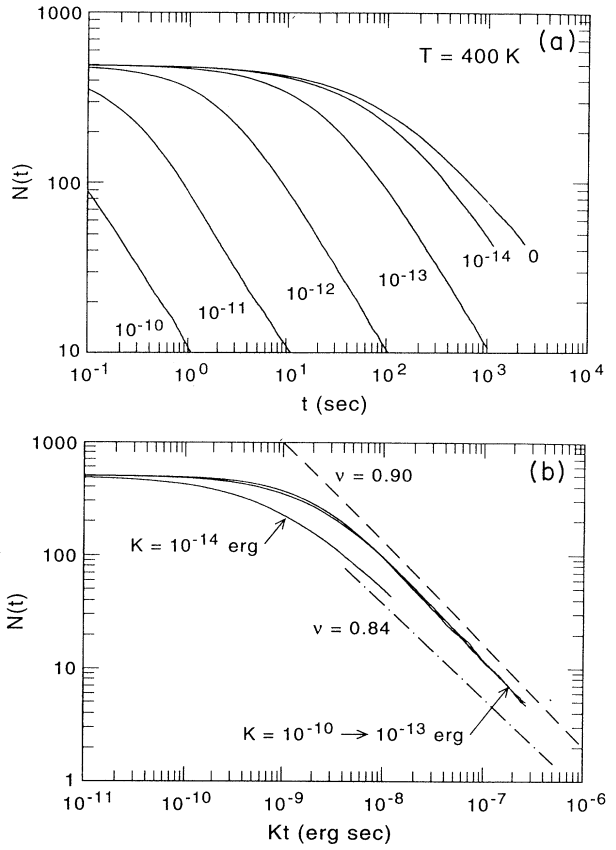

FIG. 2. Particle number as function of time for different force constants ($K=10^{-10}$, 10^{-11}, 10^{-12}, 10^{-13}, 10^{-14}, and 0 erg) at $T=400$ K and $N_0=500$, unscaled (a) and scaled (b) plots. Decay power exponents $\nu=-d(\log N)/d(\log t)=0.84\pm0.05$ for $K>10^{-13}$ erg, $\nu=0.84\pm0.05$ for $K=10^{-14}$ erg, and $\nu=0.55\pm0.05$ for $K=0$. Dashed-dotted line has slope 0.84 and dotted line has slope 0.90.

of the force constant K at fixed temperature $T=400$ K, obtained by averaging over 30 runs of 20 000 time step for each K. As suggested by the scaling rule for the deterministic regime (as discussed in the previous section), all these data lay perfectly onto one master curve (N/N_0) vs Kt, showing that $N(t)$ exhibits the expected scaling behavior for large $K>10^{-13}$ erg. However, for smaller K, diffusion plays an important role and this scaling is lost, i.e., at small or zero K, diffusion begins to prevail over long-range deterministic interaction, thus changing the driving mechanism of number decay.

In Fig. 3, the same dependence on K is analyzed for $T=0$ K. Again, data show same scaling behavior as in the previous case.

All of the N vs t data, plotted in double-logarithmic scale, show two distinct regions. At the early time, decay is rather slow, and $d(\log N)/d(\log t)\approx 0$. At later times, density exhibits a power-law decay, with $-1<d(\log N)/d(\log t)<0$. We observe two distinct cases for this intermediate scaling region: If deterministic forces are much stronger than diffusion, then $d(\log N)/d(\log t)=-0.90\pm0.05$; if diffusion is dominant in the system, then $d(\log N)/d(\log t)=-0.55\pm0.05$. In Fig. 4, N vs t for several different temperatures in the case $K=0$ are shown, with evident scaling behavior and the same exponent 0.55 in the intermediate scaling region. While this result is in agreement with other simulations of two-dimensional Brownian systems [5], the exponent measured for the deterministic case ($\nu=0.90$) has not been reported in the literature. The origins of this exponent will be discussed in the next section.

We have also calculated $g(r)$ for the + and − defects as a function of r at temperature 400 K and force constant 10^{-11} erg at time $t=0.6$ sec. The correlation function for particles of opposite sign, $g_{+-}(r)$, is shown in Fig. 5, and the correlation function for particles of the same sign, $g_{++}(r)$ or $g_{--}(r)$, is shown in Fig. 6. Both correlation functions were produced by averaging over 60 runs. The data in Fig. 5 shows that $g_{+-}(r)$ has a small peak near r_c; at the same time, as it can be seen from Fig. 6, $g_{++}(r)$ vanishes rapidly near r_c. These results are also in agreement with earlier simulations [6].

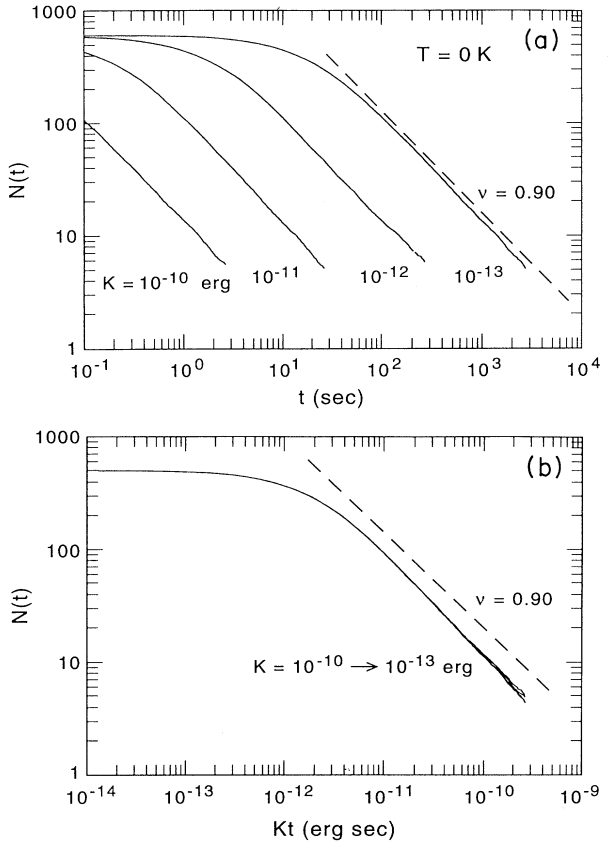

FIG. 3. Particle number as function of time for different force constants ($K=10^{-10}$, 10^{-11}, 10^{-12}, and 10^{-13} erg) at $T=0$ K and $N_0=500$, unscaled (a) and scaled (b) plots. Decay power exponent $\nu=-d(\log N)/d(\log t)=0.90\pm0.05$. Dashed line has slope 0.90.

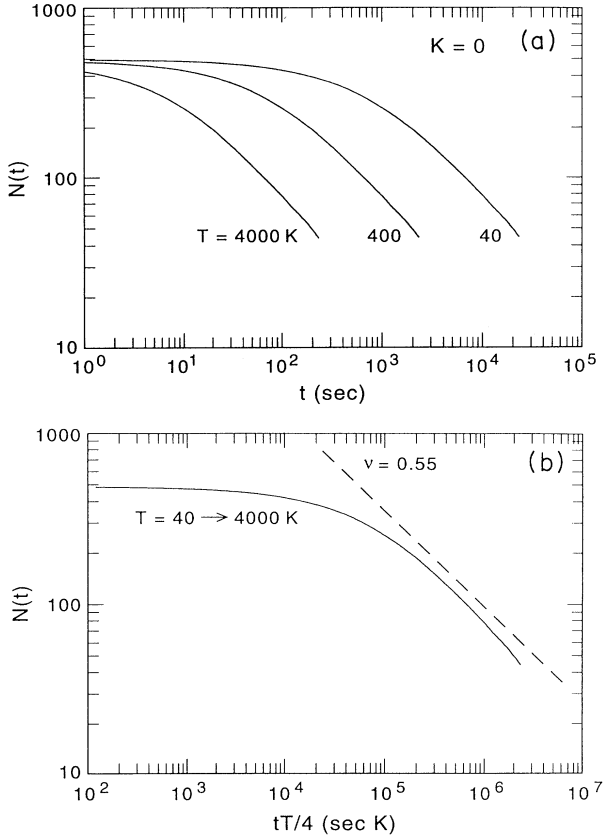

FIG. 4. Particles number as function of time for different temperatures ($T=40$, 400, and 4000 K) at $K=0$ and $N_0=500$, unscaled (a) and scaled (b) plots. Decay power exponent $\nu=-d(\log N/)d(\log t)=0.55\pm0.05$. Dashed line has slope 0.55.

To provide better insight to the annihilation dynamics, we carried out the following calculation. For each pair of defects, annihilating at time T_a (actual time), the time T_p (pair time) is determined. The pair time T_p is the time required for this pair to annihilate *in the absence of other particles, starting from their position in the initial configuration,* driven only by their mutual attraction. If the initial particle separation in the pair is L, then $T_p=L^2/(4K\mu)$. In Fig. 7, pair time is plotted against actual time. The data show that T_p is usually larger than T_a, so the large-scale charge density fluctuations seem to accelerate annihilation of pairs of remote particles.

THEORY

It is known that for systems of charged particles allowed to annihilate, the annihilation rate, as determined by the mean-field approach, is given by $\rho\propto\tau^{-\nu}$, $\nu=1$ (if system dimensionality d is two or more). Nevertheless, simulation results do not seem to support this prediction. According to Ref. [5], in the case of charged particles with no long-range forces, when only diffusive motion governs the rate of annihilation, particle density scales as $\rho\propto t^{-0.5}$ (for $d=2$). This result is in agreement also with our simulations. In Ref. [6], it is suggested that for $d=2$ and with long-range logarithmic potential with no thermal diffusion in the system, particle density scales as $\rho\propto t^{-0.75}$. The results of our simulations suggest slightly different exponent, $\rho\propto t^{-0.90\pm0.05}$. The following scaling hypothesis explains this exponent and predicts the theoretical value of $\nu=6/7$.

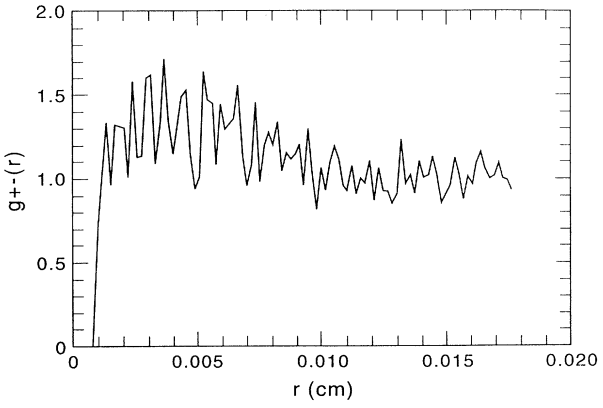

FIG. 5. Correlation function $g(r)$ for + and − defects for $T=400$ K, $N_0=500$, and $K=10^{-11}$ erg at time 0.6 sec.

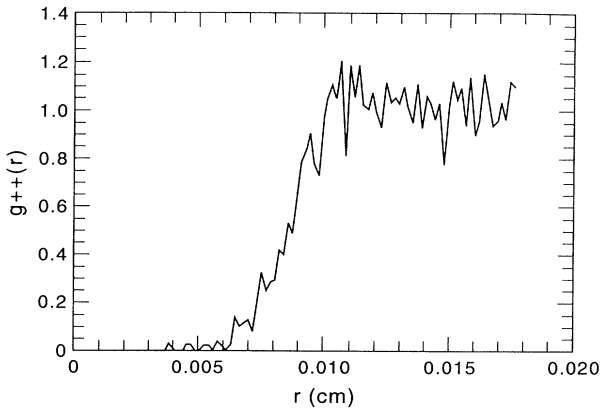

FIG. 6. Correlation function $g(r)$ for $+,+$ or $-,-$ defects for $T=400$ K, $N_0=500$, and $K=10^{-11}$ erg at time 0.6 sec.

Since deviations from mean-field behavior are caused by large-scale fluctuations, it is necessary to consider large region of characteristic size L. At the very beginning, particle density fluctuation $\delta\rho_0(L)$ is proportional to $(\rho_0 d_0)^{1/2}L^{-3/2}$ (see Ref. [6]). Indeed, since long-range potential forces act between particles, a Gaussian theorem holds for this system

$$\int E_n dl = 2\pi\Delta Q \ , \tag{5}$$

where E_n is a component of the field, normal to the contour of integration, and ΔQ is net charge in a region inside this contour. Let us now divide the region under consideration into small domains of size $d_0\times d_0$, $d_0=(\rho_0)^{-1/2}$ being the average distance between particles. In this case we can say that only particles that are in the border area contribute to the left-hand side of Eq. (4). The average number of particles in this area is proportional to $\rho_0 L d_0$, and fluctuations of this number are, therefore, proportional to $(\rho_0 L d_0)^{1/2}$. According to Eq. (4), charge fluctuations in the domain as a whole are proportional to particle number fluctuation in the domain boundary region. That brings us to the scaling relation $\delta\rho_0(L)\sim\Delta Q/L^2\propto(\rho_0 d_0)^{1/2}L^{-3/2}$, as pointed above.

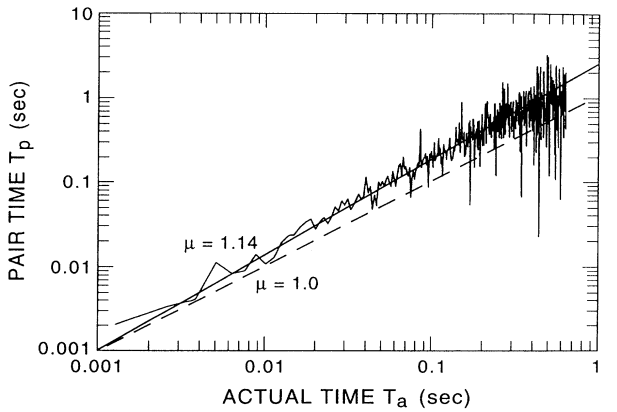

FIG. 7. Plot of pair annihilation time vs actual annihilation time, $T=0$ K, $K=10^{-11}$ erg, and $N_0=500$. Power exponent $\mu=d/(\log T_p)/d(\log T_a)=1.14\pm0.05$. Dashed line has slope 1, solid line is the best linear fit for the data and has slope 1.14.

According to the ideas of the scaling approach, when all particle-antiparticle pairs in the area $L\times L$ have annihilated, particle density at this moment is close to $\delta\rho_0(L)$, and particles of only one sign are present in the area. Let us denote τ_L the time at which this annihilation is complete. Then, most of the particles annihilating at $t\sim\tau_L$, had traveled a mean-square distance of the order of L, otherwise annihilation would not be complete.

The Langevin equation for a particle's motion in a viscous medium with deterministic force $\mathbf{F}$ acting on it, is

$$m\mathbf{R}_{tt}+(1/\mu)\mathbf{R}_t=\mathbf{F} \ , \tag{6}$$

where m is particle's mass, μ is particle mobility, and $\mathbf{R}$ is absolute displacement of the particle. We can multiply by $\mathbf{R}$ both sides of Eq. (6) and average to get (see Ref. [11])

$$(R^2)_{tt}+(1/\mu m)(R^2)_t=D+\langle\mathbf{F}\cdot\mathbf{R}\rangle/m \ , \tag{7}$$

where $\langle\ \rangle$ means average over all particles and $\mathbf{F}\cdot\mathbf{R}$ is a scalar product of force and displacement vectors.

Here we make an assumption that the motion of each particle is not chaotic, but deterministic, and that average $\langle\mathbf{F}\cdot\mathbf{R}\rangle$ is not zero, as it is in diffusive motion. In this case, we can estimate $\langle\mathbf{F}\cdot\mathbf{R}\rangle$ by using the following dimension arguments:

(i) F should be of the order of K/d; recalling that $d=\rho^{-1/2}$ and substituting $\rho=(\rho_0 d_0)^{1/2}L^{-3/2}$, we obtain the estimate for F;

$$F\propto K(\rho_0 d_0)^{1/4}(L^2)^{-3/8} \ , \tag{8}$$

$$\text{(ii)}\quad R=(L^2)^{1/2} \ . \tag{9}$$

Therefore, after substituting in Eq. (7) $\langle\mathbf{F}\cdot\mathbf{R}\rangle$ with FR [F,R given by Eqs. (8) and (9)], neglecting the second time derivative in the left-hand side and diffusion constant D in the right-hand side of Eq. (7), we obtain

$$(L^2)_t=[K\mu](\rho_0 d_0)^{1/4}(L^2)^{1/8} \ . \tag{10}$$

It immediately follows from Eq. (10) that

$$(L^2)\propto[K\mu(\rho_0 d_0)^{1/4}t]^{8/7} \ , \tag{11a}$$

$$\rho\propto(\rho_0 d_0)^{1/2}(L^2)^{-3/4}\propto\rho_0[(K\mu)\rho_0 t]^{-6/7} \ , \tag{11b}$$

which is in very good agreement with our simulations data.

Equation (11b) accounts also for the scaling behavior of normalized density ρ/ρ_0 as a function of time and force constant K [see Figs. 2(a) and 2(b)].

It is worth mentioning that, because particles are driven by the field instead of moving diffusively, the annihilation time for pair of opposite charge particles in the presence of other particles (actual time T_a) is less than in the system with no other particles (pair time T_p). According to Eq. (11a), if the initial distance between particles was L, the time before annihilation is proportional to $L^{7/4}$. On the other hand, for the same pair in the absence of other charges, the Langevin equation is

$$(1/\mu)R_t + K/(2R) = 0 , \quad (12)$$

where $2R$ is the distance between particles. After integration we obtain

$$L^2 = (4K\mu)t . \quad (13)$$

Therefore, for large L, the actual time of annihilation T_a is proportional to $L^{7/4}$, while the isolated pair annihilation time T_p is proportional to L^2. This means that $T_p \propto T_a^{8/7}$. In Fig. 7(b), the log-log plot for T_p vs T_a is shown; in the region corresponding to the intermediate scaling regime ($T_p > 0.01$ sec), the average slope is 1.14 ± 0.05, which is in perfect agreement with the scaling prediction 1.143.

Since annihilation kinetics can be described in terms of densities and pair correlation function, we can also speculate about the time behavior of the correlation function for particles with opposite charges. If density time dependence is described by Eq. (3), then by substituting ρ with its scaling time dependence, we can obtain such a dependence for $g(t,r_c)$: $g(t,r_c) \propto t^{-1/7}$, therefore decreasing with time until becoming asymptotically close to one in the final region.

DISCUSSION

The model used in the simulations, was slightly different from the typical XY model. Because defects in a two-dimensional system are considered as point particles moving in liquid, not as field singularities, one could expect some differences in these two models. These differences could, in principle, lead to some corrections in predicted density decay kinetics—corrections that, most likely, should be logarithmic, like in Ref. [7]. Nevertheless, our approach suggests several important features in defect behavior that will not be changed even if these corrections are taken into account.

Since defects or particles interact strongly in this system, the charge density fluctuations in a region of size L are proportional to its perimeter $2\pi L$, not its area πL^2. This, in turn, means that, unlike the case of pure diffusion (as in Ref. [5]), only small part of charged particles could find themselves close to particles of the same sign—but these particles will be affected more strongly than in the case of diffusion because of long-range interaction now in effect. So, while most defects annihilate rather quickly in the bulk of this region (with mean-field exponent 1), these extra particles will have to travel larger distances before annihilating. It seems to account for paradox, described by Mondello and Goldenfeld [6], that although annihilation exponent $\nu > 0.5$ (annihilation is quicker than in case of pure diffusion), mean-square displacement of a particle $\langle r^2(t) \rangle \propto t^{\lambda} < 1$. This paradox seems to be explained by the fact that, though a small fraction of particles do travel larger distances and account for annihilation decay exponent, the majority of particles are rather immobile and do not need to travel in order to annihilate—a situation that somewhat resembles normal and superfluid fractions in liquid helium. Although we cannot for now explain quantitatively the $\langle r^2(t) \rangle$ dependence, since it requires thorough account of motion of "immobile" particles, qualitatively this concept seems to be in agreement with all results.

We believe that the same approach could be used for other similar problems—systems with particles with nonlogarithmic interaction and/or three dimensional instead of two dimensional.

CONCLUSIONS

The model was developed and simulations were performed to investigate behavior of two-dimensional system with charged particles of two opposite signs, immersed into medium with constant viscosity and interacting with logarithmic interaction. It served to describe kinetics and dynamics of disclination-type defects in freely suspended films of liquid crystals, but can also model other two-dimensional systems.

In our simulations the region of intermediate scaling behavior was investigated. In the intermediate region, the kinetics of particle annihilation is determined mostly by the initial density fluctuations, whereas in the final scaling region at later time, it is determined only by current density and strength of interaction. In the final scaling region, where initial density is not a parameter, scaling requires $\rho \propto L^{-2} \propto (Kt\mu)^{-1}$. In the intermediate region, though, another length parameter d_0 exists, so different exponents may be obtained for particle density decay in systems with different mechanisms of particle motion (like pure deterministic or pure diffusive). It also depends on the behavior of the particle mobility, which is constant in our model, but becomes a function of the distance between particles in all continuum models, thus giving a logarithmic (nonpower) factor to particle decay law in the intermediate region.

Our simulations seem to suggest that long-range forces significantly accelerate annihilation in comparison with pure diffusive regime. Fluctuations are smaller and particles can move quicker because of net local effective forces. All this leads to exponent $\nu \approx 0.90 \pm 0.05$, which was obtained in the course of the simulations. The scaling hypothesis, proposed for this case by the authors, suggests exponent $\nu = 0.843$—in a very good agreement with simulation value.

The results of our scaling estimates are valid for the systems with predominant deterministic forces, giving a value of $\nu = 0.90 \pm 0.05$; when temperature becomes comparable with force constant, crossover occurs to $\nu = 0.5 \pm 0.05$ as can be seen in Fig. 2 [9,10].

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