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Using a hydrodynamic lattice-gas model, we study interface growth in a binary fluid with various concentrations of surfactant. We find that the interface is smoothed by small concentrations of surfactant, while microemulsion droplets form for large surfactant concentrations. To assist in determining the stability limits of the interface, we calculate the change in the roughness and growth exponents \( \alpha \) and \( \beta \) as a function of surfactant concentration along the interface.

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The study of rough interfaces is of important experimental and theoretical interest and has wide interdisciplinary applications, including fluid imbibition experiments, flow in porous media, and fluid-fluid displacement [1]. Rough interfaces have been extensively studied by direct integration of continuum equations and using simple discrete lattice models. The development of lattice-gas models makes it possible to study interface roughening in hydrodynamic systems. Lattice-gas models have evolved from simple one-component Navier-Stokes fluids [2] to two-component immiscible lattice-gas (ILG) models [3], and, most recently, to a model including amphiphilic particles [4]. Lattice-gas models can reproduce the dynamics on mesoscopic scales, allowing for the investigation of nonequilibrium behavior over a much broader range of length and time scales than is possible with molecular dynamics [5]. The simplicity of the collision rules, the exact conservation of mass and momentum, and the natural underlying kinetic fluctuations suggest that lattice-gas models are an appropriate choice for studying the scaling behavior of interfaces in complex hydrodynamic systems.

In this Letter, we study interfacial roughening in the presence of a surfactant by including amphiphilic particles that tend to reside at the binary fluid interface [6]. Flekkøy and Rothman recently studied fluctuating interfaces in an ILG model without surfactant and described the scaling properties of these interfaces [7]. When surfactant is added to an initially flat interface, we find that the roughness exponent \( \alpha \) and growth exponent \( \beta \) decrease for small concentrations of surfactant [8]; however, for larger concentrations of surfactant, \( \beta \) increases while \( \alpha \) continues to decrease. The continued addition of surfactant causes the interface to spontaneously break up into a microemulsion phase. We also find that the saturated width of the interface has a minimum when the surfactant concentration along the interface matches the binary fluid concentration in the bulk.

Far from any interface, the present model approaches the one-component FHP-II model [2,9], which employs a triangular lattice with each lattice site occupied by up to seven particles, each of which is either at rest or has unit velocity directed along one of the six lattice vectors toward neighboring sites. No two particles may have the same velocity at a lattice site. The rules dictate that a particle propagates along a lattice direction until a collision occurs. When no surfactant is present, the current model reduces to the two-component ILG model [3], which employs two types of particles and assigns each of the two species a color, “red” or “blue.” Each collision must conserve red particles, blue particles, and the total momentum. Additional collision rules are included that give rise to the aggregate behavior of an immiscible fluid. These rules can be described using an electrostatic analogy [4], where the color flux \( \mathbf{J}(x,t) \) of an outgoing state is the difference between the red and blue momenta. The color field \( \mathbf{E}(x,t) \) is the gradient of color between neighboring sites. At each time step, the color work—defined by the inner product \( \mathbf{J} \cdot \mathbf{E} \)—is minimized at each lattice site, which has the effect of inducing phase separation.

With the addition of surfactant molecules, the ILG becomes a three-component model having the potential to model amphiphilic systems [4]. To make the behavior of the surfactant consistent with a molecule composed of hydrophobic and hydrophilic ends, the surfactant is represented as a “color dipole” with a dipole vector \( \mathbf{\alpha}(x,t) \) and produces a dipolar field \( \mathbf{P}(x,t) \) at neighboring sites. A term proportional to \( \mathbf{\alpha} \cdot \mathbf{E} \), representing the interaction of colored particles with dipoles, is added to the Hamiltonian. Similarly, we add a term proportional to \( \mathbf{\alpha} \cdot \mathbf{P} \) to include the dipole-dipole interaction. Minimization of the modified Hamiltonian favors the surfactant particles lining up along the interface between red and blue particles and makes it unfavorable for surfactant particles to align with each other at neighboring sites, modeling the expected behavior of an amphiphilic molecule [10].

We perform simulations of the three-component model using a sequence of two-dimensional lattices of edge \( L = 16, 32, 64, 128 \) and 256 with periodic boundary conditions in both directions. Initially, red and blue particles are placed on the lattice separated by two flat interfaces which are evenly spaced and oriented perpen-
interface mediates the roughening process. The system then tends to align perpendicular to the interface with varying concentration and angular orientation randomly selected between 0 and $2\pi$. (After a single collision step, the angular orientation of the surfactant molecules tends to align perpendicular to the interface, as this is energetically favorable.) The system then evolves according to the collision rules of the model. Our interest focuses on how the addition of surfactant to the interface mediates the roughening process.

We first calculate the average interface width $W(L, t) = \langle h^2(x, t) - h(x, t) \rangle^{1/2}$ at logarithmically spaced time intervals with no surfactant present. We find that $W(L, t) \sim t^\beta$ until a crossover time $t_c(L)$, whereupon it saturates at a value $W_{sat}(L) \sim L^\alpha$ [1, 7], with $\beta = 0.33 \pm 0.02$ and $\alpha = 0.50 \pm 0.02$ (Fig. 2). Hence $z = \alpha/\beta = 1.50 \pm 0.03$, where $z$ is the dynamical exponent, defined by $t_c(L) \sim L^z$. In Fig. 2(b), we plot the rescaled width $W(L, t)/L^\alpha$ against the rescaled time $t/L^z$ and confirm the scaling hypothesis, $W(L, t) \sim L^\alpha f(t/L^z)$ [1]. Our results match the scaling exponents $\alpha = 1/2$ and $\beta = 1/3$ for the Kardar-Parisi-Zhang (KPZ) universality class [13], and are comparable to earlier results obtained by Flekkøy and Rothman [7].

Next we consider interface roughening with surfactant present. In the three-component model, for surfactant concentrations below the critical concentration for spontaneous emulsification, the surfactant particles tend to line up along the interface between the two fluids. Since we are interested in the role that the surfactant plays in the kinetic roughening process, we start with an initial configuration where the fluids are separated and we replace red and blue particles with surfactant particles in a single row along the interface [Fig. 1(b)]. We define the surfactant concentration $\rho_s = N_s/L$, where $N_s$ is the number of surfactant particles placed along the interface. To obtain $\rho_s > \rho$, we replace red and blue particles with surfactant particles on multiple rows [14].

In Fig. 3(a), we plot $W(L, t)$ for all system sizes and $\rho = \rho_s = 0.5$. We still see a growth regime that crosses over to a saturated regime. Compared to Fig. 2, the case without surfactant, the growth rate of the width is slower and $W_{sat}$ is smaller. We find $\beta = 0.27 \pm 0.02$ and $\alpha = 0.43 \pm 0.02$. Figure 3(b) confirms the scaling hypothesis using $z = \alpha/\beta = 1.59 \pm 0.03$. These values for $\alpha$ and $\beta$ are not the accepted values for the roughness and growth exponents of the KPZ equation in $1 + 1$ dimensions. The KPZ exponents also obey the sum rule, $z + \alpha = 2$, a consequence of Galilean invariance [1]. We obtain $z + \alpha = 2.02 \pm 0.04$ for this case. This result is surprising since lattice-gas models are
not Galilean invariant, due to the fact that the lattice constitutes a preferred Galilean reference frame [2]. On the other hand, the flow equations of the lattice-gas model do contain a nonlinear inertial term, as does the KPZ equation.

We next consider the effect of the surfactant concentration on the growth of the interface and the saturated width. In Fig. 4(a), we show the dependence of $\beta$ on $\rho_s$ for a lattice of size $32 \times 32$. We observe a dramatic reduction in the growth exponent upon the addition of any surfactant to the interface. Additional surfactant causes little change in $\beta$ up to $\rho_s = \rho$, where $\beta = 0.22 \pm 0.02$. For $\rho_s > \rho$, the growth exponent increases but never returns to the value of the pure case. For $\rho_s \approx 1.5$, the interface spontaneously breaks up, giving rise to a micellar microemulsion phase where the width is no longer well defined [Fig. 1(c)]. The effect of $\rho_s$ on $W_{\text{sat}}$ is similar to its effect on $\beta$. For $\rho_s < \rho$ we see that the interface width saturates at a value that decreases for increasing $\rho_s$ [Fig. 4(b)]. However, for $\rho_s > \rho$, $W_{\text{sat}}$ increases until we reach the microemulsion-droplet phase.

We can understand the behavior of $W_{\text{sat}}$ by considering the effect the surfactant has on the binary fluid interface. From the Hamiltonian, we know that the color-dipole energy, $\mathbf{\sigma} \cdot \mathbf{E}$, required for surfactant to be removed from the interface is large compared to the energy for the surfactant to remain on the interface. As a result, the surfactant particles are effectively “anchored” at the interface. For $\rho_s < \rho$, the surfactant reduces the fluctuations of the interface because it is anchored in position between red and blue particles. As a result, $W_{\text{sat}}$ is smaller. For $\rho_s > \rho$, the color-dipole interaction still forces surfactant particles to remain on the interface, but now surfactant particles are more likely to neighbor each other. Since the dipole-dipole interaction, $\mathbf{\sigma} \cdot \mathbf{P}$, makes it energetically unfavorable for dipole molecules to align with other dipole molecules at neighboring sites, the surfactant creates additional surface by roughening the interface to reduce the number of neighboring surfactant molecules, thereby increasing $W_{\text{sat}}$. To demonstrate that the turning point in $W_{\text{sat}}$ depends on our choice of $\rho$, we measure $W_{\text{sat}}$ for various $\rho_s$ with a binary fluid concentration $\rho = 0.7$ [Fig. 4(c)]. We again see $W_{\text{sat}}$ decreasing for $\rho_s \leq \rho$ and increasing for $\rho_s > \rho$.

To investigate the effect of $\rho_s$ on $\alpha$, we consider system sizes $L = 16, 32, 64$, and 128 for $\rho_s = 0.8$ and 1.0. At $\rho_s = 0.8$ we find $\beta = 0.27 \pm 0.02$ and $\alpha = 0.36 \pm 0.02$, while for $\rho_s = 1.0$ we obtain $\beta = 0.30 \pm 0.02$ and $\alpha = 0.23 \pm 0.02$. Using the exponents for these concentrations, we do not find $z + \alpha = 2$. In Table I we summarize the behavior of the exponents $\alpha$, $\beta$, $z$, and $z + \alpha$ as a function of the surfactant concentration. We
TABLE I. Roughness, growth and dynamic exponents for various surfactant concentrations measured from the simulations. Also included are the number of samples that have been averaged.

<table>
<thead>
<tr>
<th>ρ_s</th>
<th>α</th>
<th>β</th>
<th>z</th>
<th>z + α</th>
<th>Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.50 ± 0.02</td>
<td>0.33 ± 0.02</td>
<td>1.50 ± 0.03</td>
<td>2.00 ± 0.04</td>
<td>170</td>
</tr>
<tr>
<td>0.5</td>
<td>0.43 ± 0.02</td>
<td>0.27 ± 0.02</td>
<td>1.59 ± 0.03</td>
<td>2.02 ± 0.04</td>
<td>170</td>
</tr>
<tr>
<td>0.8</td>
<td>0.36 ± 0.02</td>
<td>0.27 ± 0.02</td>
<td>1.33 ± 0.03</td>
<td>1.69 ± 0.04</td>
<td>150</td>
</tr>
<tr>
<td>1.0</td>
<td>0.23 ± 0.02</td>
<td>0.30 ± 0.02</td>
<td>0.77 ± 0.03</td>
<td>1.00 ± 0.04</td>
<td>150</td>
</tr>
</tbody>
</table>

notice that α decreases as we increase the surfactant concentration.

In summary, we have found that a three-component lattice gas can be used to test the effect of surfactant on the scaling properties of hydrodynamic interfacial growth and roughening. In particular, we have found that surfactant alters the growth and roughness exponents, with α decreasing and β increasing as we increase ρ_s above the concentration of the binary fluid. These results may assist in understanding the stability limits of interfacial growth in the presence of a surfactant and the conditions for the formation and breakup of microemulsion droplets.

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[8] The roughness exponent α describes the dependence of the saturated width on system size, W_{sat}(L) \sim L^{\alpha}, while the growth exponent β describes the initial increase in interface width, W(L,t) \sim t^{\beta}.
[10] The relative strength of each term in the Hamiltonian can be controlled by coupling constants. Using the nomenclature of Ref. [4], we have chosen α = 1.0, μ = 0.05, ε = 8.0, and ζ = 0.5. Here α is not to be confused with the roughness exponent.
[12] This is equivalent to 3.5 particles per site on average. This choice is not crucial for a binary fluid provided ρ is above the critical concentration needed for phase separation.
[14] For ρ_s \leq ρ, we replace the red and blue particles with surfactant particles on only one row of the interface. For ρ < ρ_s \leq 2ρ, we use two rows. Similarly, we use three rows for 2ρ < ρ_s \leq 3ρ.