A Letters Journal Exploring the Frontiers of Physics

January 2010

EPL, **89** (2010) 10006
doi: 10.1209/0295-5075/89/10006

www.epljournal.org

Lateral transport of thermal capillary waves

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received 15 October 2009; accepted in final form 9 December 2009
published online 19 January 2010

PACS 05.40.-a – Fluctuation phenomena, random processes, noise, and Brownian motion
PACS 05.50.+q – Lattice theory and statistics (Ising, Potts, etc.)
PACS 68.05.Cf – Liquid-liquid interface structure: measurements and simulations

Abstract – We demonstrate that collective motion of interfacial fluctuations can occur at the interface between two coexisting thermodynamic phases. Based on computer simulation results for driven diffusive Ising and Blume-Capel models, we conjecture that the thermal capillary waves at a planar interface travel along the interface, if the lateral order parameter current $j(y)$ is an odd function of the distance y from the interface and hence possesses opposite directions in the two phases. Such motion does not occur if $j(y)$ is an even function of y. A discrete Gaussian interface model with effective dynamics exhibits similiar transport phenomena but with a simpler dispersion relation. These findings open up avenues for controlled interfacial transport on the nanoscale.

Understanding the motion of interfaces is important in areas such as multiphase flow, dendritic and crystal growth, microchip fabrication, combustion, blood flow, and cell dynamics. Pronounced changes to the microscopic interfacial structure can occur as a result of the motion, *e.g.*, the development of kinetically enhanced self-affine roughness [1] of the driven interface between a growing material and its environment. In striking contrast, driving the interface along the plane of its average position can lead to interfacial smoothening, as has been observed experimentally for a colloidal gas-liquid interface under shear flow [2], and in computer simulations of interfaces in driven lattice gas models [3,4]. Coherent lateral transport of large-scale interfacial structures is well known in macroscopic systems that are far from equilibrium, such as migrating sand dunes and ripples, or ocean waves. Finding thermal analogues can be of relevance in micro- and nanofluidic devices, *e.g.* due to the possibility of transport of nanoparticles at a liquid-liquid interface by controlled motion of thermal capillary waves.

Consider two distinct equilibrium phases at thermodynamic phase coexistence, organised into two different regions of space that are separated by a planar interface. The local order parameter Φ varies along the axis y perpendicular to the interface plane and for large enough systems reaches its respective bulk value far from the interface. The (scalar) order parameter profile $\Phi(y)$ can represent the density near a gas-liquid interface, the relative concentration at the interface separating two coexisting liquid phases, or the magnetization near an Ising domain wall. On a coarse-grained scale, the interface can be characterized by its local departure (height) $h(x,t)$ from a reference plane $y = h(x,t) = 0$, where x indicates the coordinate(s) parallel to the interface and t is time. At temperatures T above the roughening transition, the interface exhibits large spatial fluctuations. Phenomenological capillary wave theory [5], as well as rigorous results [6], indicate that in the absence of external fields that couple to the order parameter, the length scale of such fluctuations diverges with system size, *i.e.*, the interface thickness becomes infinite in the thermodynamic limit in spatial dimensions $d \leqslant 3$.

In this letter we consider external driving that creates a steady state with non-vanishing current of the order parameter, $j(y)$, parallel to a planar interface. We investigate the effects on the dynamics of the capillary waves via the two-point correlation function $C(x,t) = \langle h(0,0)h(x,t)\rangle$, where the angles denote an average in the steady state. Based on computer simulation results for $C(x,t)$ for various simple microscopic models, we conjecture that the

lateral flux of the order parameter at a planar interface induces lateral motion of the thermal capillary waves, provided $j(y)$ is an odd function of the distance y from the interface. The spatial symmetry at equal times, $C(x,0) = C(-x,0)$, is broken for times $t > 0$, such that $C(x,t) \neq C(-x,t)$. For general forms of $j(y)$ it is the odd component, $[j(y) - j(-y)]/2$, that induces the motion. No motion occurs when $j(y)$ is even. We shed light on the transport mechanism by constructing a corresponding (Gaussian) effective interface model that displays similar transport phenomena.

As a microscopic approach we use kinetic lattice models with dynamics that conserve the order parameter locally. The Ising and the Blume-Capel [7] models of binary mixtures possess the Hamiltonian $\mathcal{H} = -J \sum_{\langle i,l \rangle} \sigma_i \sigma_l$, where $\langle i,l \rangle$ denotes nearest-neighbor pairs and the spin-spin coupling constant $J > 0$. Contributions to $\mathcal{H}$ from static external fields are omitted, as we will work in an ensemble of fixed numbers of spins σ_i of each type. For the Ising model $\sigma_i = \pm 1$, while for the Blume-Capel model $\sigma_i = -1, 0, +1$, which in lattice fluid language corresponds to occupancy of site i by particles of species "−1", vacancy, or species "+1" of the mixture, respectively. We initially consider two-dimensional square lattices of dimensions $L_x \times L_y$ with coordinates x (horizontal) and y (vertical). The interface with mean orientation in the x-direction is established and localized by assuming boundary conditions $\sigma_i = +1$ at the upper $(y = (L_y + 1)/2)$ and $\sigma_i = -1$ at the lower $(y = -(L_y + 1)/2)$ edges of the lattice. Periodic boundary conditions are applied in the x direction. As a consequence, $\Phi(y) = \langle \sum_x \sigma_i \rangle / L_x$, $i = (x,y)$ crosses over from $\Phi < 0$ for $y < 0$ to $\Phi > 0$ for $y > 0$. We use Kawasaki spin exchange dynamics [8], where the elementary move consists of swapping the values of the spin variables σ_i and σ_l of two nearest-neighbor sites i and l. The system is driven by a force field $F(y)$ that acts in the x-direction (parallel to the interface) and varies with distance y from the interface. The acceptance rates for the trial moves are assumed to be of modified Metropolis type, $\min\{1, \exp(-(\Delta H + \Delta W)/(k_B T))\}$, where ΔH is the change in internal energy, ΔW is the work due to the driving field, and k_B is the Boltzmann constant. Hence the work performed by the field is dissipated into a heat bath, which is kept at $T = \text{const}$. We have carried out extensive Monte Carlo (MC) simulations using multispin [9] coding techniques extended for Kawasaki dynamics to include drive. Results are presented for $L_x = 200$ and $L_y = 20$ at fixed total magnetization $\sum_i \sigma_i = 0$ for $T/T_c = 0.75$ (where $T_c = 2.2692 J/k_B$) and run lengths of the order of $N_{\text{MC}} = 10^8$ MC steps ($L_x \times L_y$ trial moves).

We first discuss the Ising lattice gas where the drive creates a work term for an exchange in the x-direction, $\Delta W = -JF(y)(\sigma_i - \sigma_l)/2$, where $i = (x,y)$ and $l = (x+1, y)$; exchanges in the y-direction occur with normal equilibrium rates, $\Delta W = 0$. In the case of odd symmetry upon spatial reflection, $F(y) = -F(-y)$, the field acts in opposing directions in both halves of the system, *e.g.* with linear variation across the slit, $F(y) \equiv \gamma y$, where γ is the (scaled) field difference between rows. We define the particle current profile $j_\sigma(y)$ as the average net number of $\sigma = \pm 1$ particles that move from x to $x+1$ at given height y per unit time. $j_\sigma(y)$ is independent of x due to translational symmetry in the x-direction. Simulation results indicate that the order parameter current profile $j(y) \equiv j_+(y) - j_-(y)$ possesses the same direction and the same symmetry as the driving field. For odd driving fields, we find $j(y) = -j(-y)$. We define the instantaneous interface position via a coarse-graining method based on the (scaled) column magnetization, $h(x,t) = -(2m_{\text{b}})^{-1} \sum_y \sigma_i$, where site $i = (x,y)$ possesses the value σ_i at time t, and m_{b} is the spontaneous equilibrium magnetization in bulk. Note that while, *e.g.*, in the solid-on-solid (SOS) model bubbles and overhangs at the interface are forbidden, here the underlying configurations are those of the Ising model.

In fig. 1(a) we plot $C(x,t)$ as a function of x for several fixed values of time difference t for the case of linear drive, $\gamma = 1$. At $t = 0$, a cusp is apparent at $x = 0$ and $C(x,0)$ exhibits long-ranged decay with distance x, characterizing the (equal-time) spatial correlations of the interfacial fluctuations. Upon increasing time, $t > 0$, the position of the peak moves towards negative values of x, its height decays and its width increases. This behaviour clearly indicates the existence of damped propagating modes that move in the negative x-direction. The position of the maximum of $C(x,t)$ varies linearly with time. The inferred velocity is $v_{\text{peak}} = 0.009$ (in units of lattice constant per MC step); for not too large values of γ the velocity v_{peak} grows linearly with increasing γ. For step-like drive of strength f, *i.e.*, $F(y) \equiv f \,\text{sgn}(y)$, where $\text{sgn}(\cdot)$ is the sign function, we observe very similar behaviour of $C(x,t)$ (data not shown), from which we conclude that the occurrence of the interfacial motion is not tied to the specific functional form of the (odd) driving field. Qualitatively different behaviour occurs for even symmetry of the drive, $F(y) = F(-y)$, such that the drive acts in the same direction throughout the system. We find, for the cases considered, that the order parameter current profile also attains even symmetry, $j(y) = j(-y)$. For a V-shaped spatial dependence, $F(y) \equiv \gamma |y|$, simulation data, shown in the inset of fig. 1(a), show that with increasing time the peak of $C(x,t)$ decreases in magnitude but remains stationary at $x = 0$, which indicates the absence of propagating modes. This observation holds also for the case of uniform drive, $F(y) \equiv f = \text{const}$. The intercept of the equal-time correlation function, $C(0,0) = \langle h(0,0)^2 \rangle$, provides a measure of the (squared) interfacial width [4]; we find for all cases considered that $C(0,0)$ decreases under drive. Comparison of the results for linear and V-shaped drive indicates that the suppression of roughness [3] is less strong in cases where interfacial motion occurs. The interfacial transport is intimately related to a broken symmetry under space reflection,

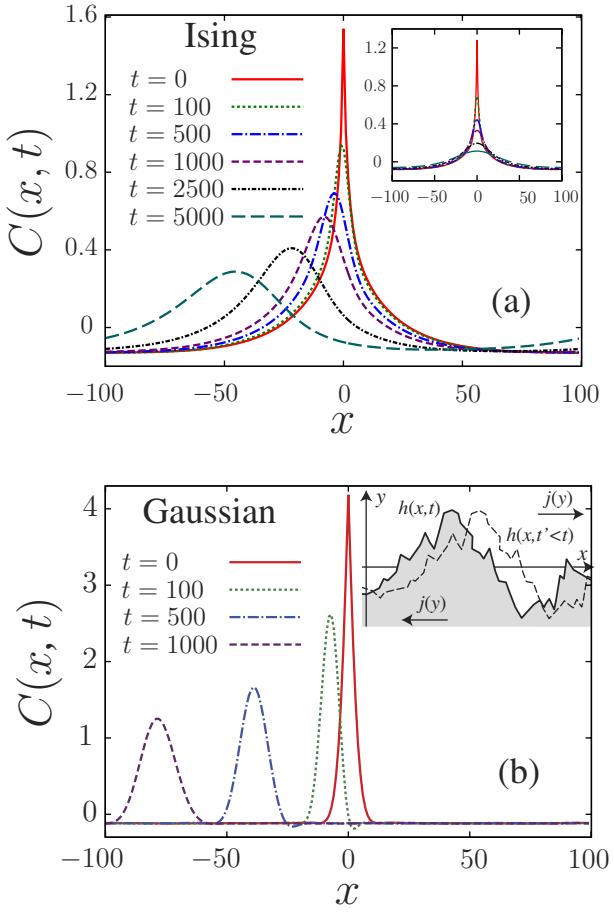

Fig. 1: (Color online) (a) Height-height correlation function $C(x,t)$ of the Ising model as a function of the distance x at constant time t (as indicated, in units of MC steps) for temperature $T/T_c = 0.75$, and system size $L_x = 200$, and $L_y = 20$. The system is subject to linearly varying drive with strength $\gamma = 1$. Inset: $C(x,t)$ for the case of V-shaped drive for the same parameters. (b) Same as (a), but for the discrete Gaussian interface model subject to linear h-dependent drive. Inset: Illustration of the motion of the fluctuating interface. The height function $h(x,t)$ is depicted at times t' (dashed line) and $t > t'$ (solid line). The arrows indicate the direction of the order parameter current $j(y)$.

$y \to -y$ and $x \to -x$, and species inversion, $\sigma_i \to -\sigma_i$, that occurs for cases of odd driving. As a consequence, on average -1 spins in the region with $y > 0$ move in the *same* (negative) x direction as $+1$ spins do in the region with $y < 0$. Hence "intruders" of the other phase move in the same direction throughout the system. (Intruders are $+1$ spins in a region with negative average magnetization and -1 spins in a region with positive average magnetization.) This direction is opposite to that of the "velocity profile" of the order parameter, $v_\Phi(y) = j(y)/\Phi(y)$, which is $0 \leqslant v_\Phi(y) \leqslant 0.022$ for all y. See the inset of fig. 1(b) for an illustration of the transport phenomenon.

The discrete Gaussian model [10] provides a reduced description with interfacial degrees of freedom only. Nevertheless these are known to exhibit long-wavelength equilibrium fluctuations that are characteristic of real fluid interfaces. In a one-dimensional system the interface is represented by integer height variables $h(x)$ with $x = 1, \ldots, L_x$ and periodic boundary conditions. The Hamiltonian is $\mathcal{H}_{\rm DG} = (J/2)\sum_{x=1}^{L_x}(h(x+1) - h(x))^2$. In our (conserved) dynamics a site x and one of its neighbours $x' = x \pm 1$ are chosen at random, and the heights are changed as $h(x) \to h(x) + 1$ and $h(x') \to h(x') - 1$, with probability given by the modified Metropolis rate. We have performed MC simulations for the same parameters as for the Ising model. First, the work varies linearly with height, $\Delta W = -(x' - x)(h(x) + h(x'))J\gamma/2$. Results for $C(x,t)$, shown in fig. 1(b), exhibit characteristics very similar to those found in the Ising model with capillary wave motion. The velocity of the position of the maximum is $v_{\rm peak} = 0.08$ (in units of lattice constants per MC step, where one MC step consists of L_x exchange moves), significantly larger than that of the Ising model. Biasing the moves to the (say) left irrespective of the height variable, $\Delta W = -(x' - x)Jf$, corresponds to uniform drive in the Ising model. As in that model, no transport is observed. In cases where transport occurs, the combined symmetry $h \to -h$, $x \to -x$, and exchange of x and x', is broken.

The short-time dynamics of the capillary waves can be characterized by a dispersion relation of frequency ω as a function of the wave number $q = 2\pi k/L_x$, obtained from the (average) phase shift of mode $k = 1, 2, \ldots, L_x$ in unit time as $\omega(q) = \arg\langle \tilde{h}^*(q,t)\tilde{h}(q,t+{\rm d}t)\rangle/{\rm d}t$, where $\tilde{h}(q,t)$ is the spatial Fourier transform of $h(x,t)$, and ${\rm d}t$ is a small time interval ($L_x/10$ spin exchanges). Surprisingly, for small wave numbers q the driven Ising lattice gas possesses, besides a leading linear term, strong quadratic and cubic contributions to the dispersion relation, see fig. 2(a) where results are shown for the case of linear drive with $\gamma = 1$. In contrast, $\omega(q)$ for the discrete Gaussian model remains nearly linear over a much larger range of values of q, see fig. 2(b). Modelling the dynamics by a simple linear transport operator[1] $\partial_t - v\partial_x$, with the continuous partial time derivative ∂_t, discrete spatial derivative $\partial_x h(x,t) = [h(x+1,t) - h(x-1,t)]/2$, and plane wave modes $\exp({\rm i}(\omega t + qx))$ yields $\omega(q) = v\sin(q)$. This describes the simulation data very well, see fig. 2(b). For small q, the behaviour $\omega(q) = vq$, with $v = 0.0760(1)$, is in good agreement with the value of $v_{\rm peak}$ obtained from analysis of $C(x,t)$.

[1] From the evolution equation for the mean change in height per unit time, $\partial_t h(x,t) = [w(x-1 \to x) - w(x \to x+1) + w(x+1 \to x) - w(x \to x-1)]/2$, where $w(x' \to x)$ is the (Metropolis) probability for the simultaneous update $h(x') \to h(x') - 1$ and $h(x) \to h(x) + 1$, one obtains for strong drive, $|\Delta W| \gg |\Delta H|$, and *e.g.* away from the midline $h = 0$ the result $\partial_t h = \alpha\exp(-\alpha|h| - \alpha\,{\rm sgn}(h)\partial_x^2 h/4)\partial_x h/2$, where $\alpha = \gamma J/(k_B T)$. Linearization yields $\partial_t h = (\alpha/2)\partial_x h$.

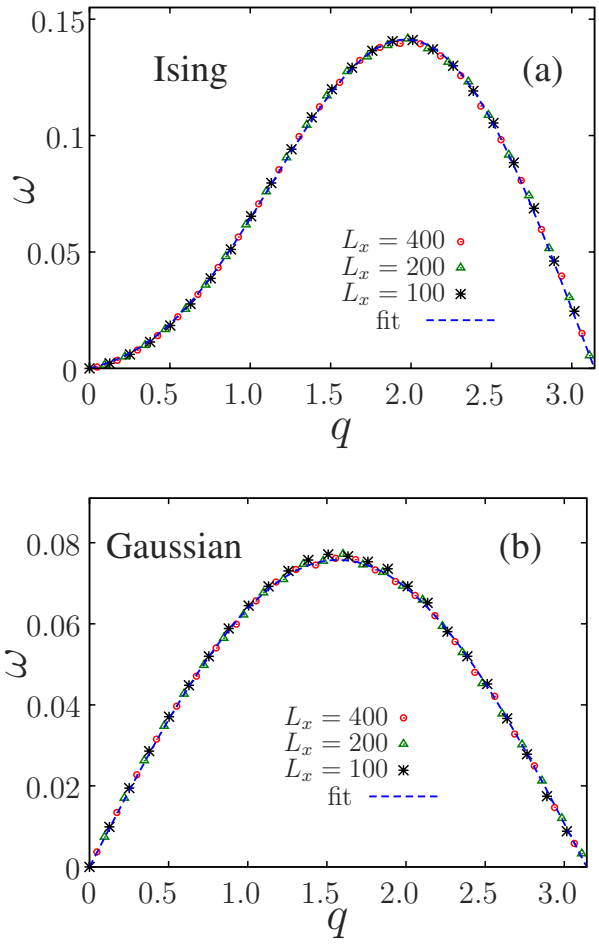

Fig. 2: (Color online) Dispersion relation $\omega(q)$ for the lateral propagation of capillary waves in the two-dimensional Ising model driven by a linear field for $\gamma = 1$ (a), and in the one-dimensional kinetic discrete Gaussian model with height-dependent driving (b). The spectral variable is $q = 2\pi k/L_x$, where k is the wave number. Parameters are the same as in fig. 1. Data for different system sizes $L_x = 100, 200, 400$ collapse onto each other. Also shown is the analytical formula (line) described in the text.

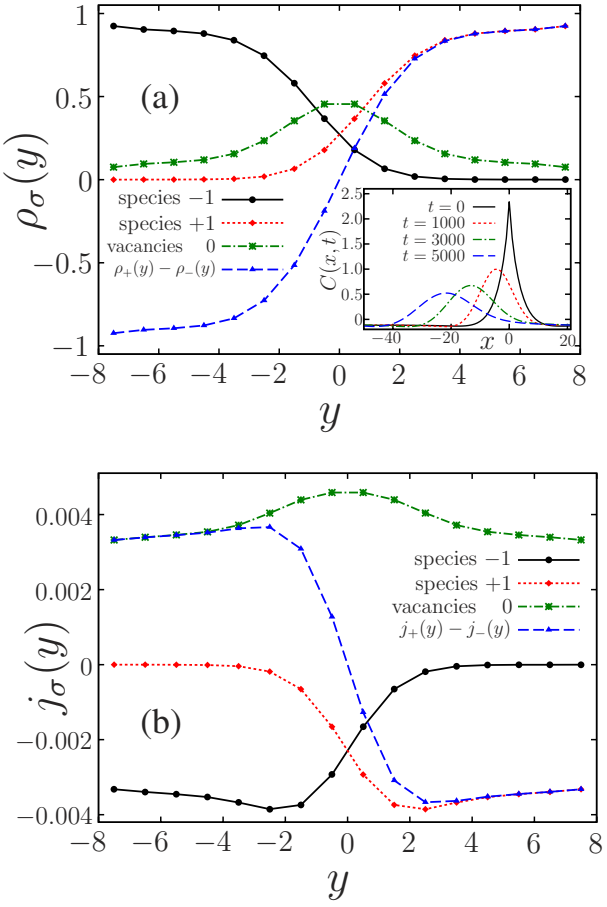

Fig. 3: (Color online) (a) Density profiles $\rho_\sigma(y)$ of species -1, $+1$, and vacancies (0) for the Blume-Capel model under homogeneous drive of strength $f = 0.25$ and $T = 0.75J/k_B$ as a function of the distance y from the mean interface position. Also shown is the order parameter profile $\Phi(y) = \rho_+(y) - \rho_-(y)$. Inset: $C(x,t)$ as a function of x for various values of t (as indicated). (b) Particle current profiles $j_\sigma(y)$ for species $\sigma = -1, +1, 0$ and order parameter current profile, $j(y) = j_+(y) - j_-(y)$, as a function of y.

The much richer behaviour of the Ising model can be fitted to $\omega(q) = (v + 2u)\sin(q) - u\sin(2q) + s\sin^2(q)$ with small-q expansion $\omega(q) = vq + sq^2 + (u - v/6)q^3$, and $v = 0.0172(7), u = 0.0402(2), s = 0.0263(13)$. Assuming $\partial_t - v\partial_x + u\partial_x^3$ (5-point stencil for ∂_x^3) yields the first and second terms of the fit function, but the third term cannot be obtained from a linear transport equation with real coefficients[2]. We have checked that these results do not depend significantly on the method of coarse-graining to obtain $h(x,t)$ from the microscopic spin configurations. Using the procedure of ref. [11] yields results for $\omega(q)$ that can be fitted with $s = 0$ and a differing value for v. The qualitative features of $C(x,t)$, cf. fig. 1(a), remain unchanged. Moreover, the bare spin-spin correlation function $\langle \sigma_i \sigma_j \rangle$, calculated along the midline of the system, displays similar transport features as $C(x,t)$ does.

In the kinetic Blume-Capel model the presence of vacancies (sites i with $\sigma_i = 0$) creates richer microscopic dynamics. In contrast to the Ising model, where driving $+1$ particles is intimately related to counter-driving -1 particles in the opposite direction, the Blume-Capel model offers the possibility to *co-drive* the ± 1 species in the same direction. The corresponding driving fields $F_\sigma(y)$ acting on species $\sigma = \pm 1$ then obey $F_+(y) = F_-(y) \equiv F(y)$. The work term for an exchange of spins i and l with coordinates x and $x' = x + 1$, respectively, is $\Delta W = -JF(y)(\sigma_i^2 - \sigma_l^2)$. Simulation results for a vacancy concentration of 20%, where we find the confined interface to be stable, indicate that the symmetries of the driving field $F(y)$ and of $j(y)$ are no longer the same, *e.g.*, that an even field can

[2]Including an imaginary operator is$(\partial_x^2 + \partial_x^4/4)$ with 3- and 5-point stencils, respectively, yields the third term, albeit at the expense of taking $h(x,t)$ as a complex field.

give rise to an odd order parameter current profile, see fig. 3 for results of the density profile, $\rho_\sigma(y)$, and current profile, $j_\sigma(y)$, of each species $\sigma = -1, 0, +1$ for the case of uniform drive, $F(y) = \text{const}$. Indeed motion of capillary waves is observed, see the inset of fig. 3(a) for results for $C(x,t)$. We do not find wave motion when counter-driving, *i.e.* for $F_+(y) = -F_-(y)$, where $j(y)$ turns out to be even. All these findings are fully consistent with the proposed scenario that the interfacial motion is caused by the symmetry of $j(y)$ and not by that of the driving fields.

For general cases where the driving field and hence the order parameter current profile do not possess unique but rather mixed spatial symmetry, we find, for the Ising model, that it is the presence of an odd component that leads to interfacial motion. We have also checked that the transport phenomenon is not specific to two-dimensional systems, but also occurs in the three-dimensional Ising model, where simulation results indicate that the direction of motion is parallel to that of the drive and that the second coordinate parallel to the interface plays a mere spectator role. It is well known that the microstructure and mobility of driven interfaces depends significantly on the detailed form of the stochastic transition rates [1]. We have used (exchange) dynamics with rates that factorize into internal and external contributions, *i.e.* "soft Glauber" $[1+\exp(\beta\Delta H)]^{-1}[1+\exp(\beta\Delta W)]^{-1}$ and "soft Metropolis" $\min\{1, \exp(-\beta\Delta H)\}\min\{1, \exp(-\beta\Delta W)\}$. Preliminary results for sheared driving suggest that $C(x,t)$ remains qualitatively unchanged, *i.e.* possesses features characteristic of interfacial motion, albeit with a reduced value of v_{peak}. In view of possible microfluidic applications, it would be very interesting to test the validity of the proposed scenario in experiments (*e.g.*, using colloids [2]) and in molecular-dynamics simulations [12]. It would also be very valuable to investigate motion of thermal capillary waves using models based on the order parameter [13].

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We thank D. Abraham, S. Dietrich, A. Gambassi, and T. Fischer for useful discussions. We acknowledge financial support by the EPSRC under Grant No. EP/E065619 and by the DFG via SFB840/A3.

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