École doctorale 209 : Sciences physiques et de l'ingénieur

THESIS

to obtain a PhD from

Université de Bordeaux

Spécialité doctorale “Laser, Matière, Nanosciences“

presented by

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Confinement and driving effects on continuous and discrete model interfaces

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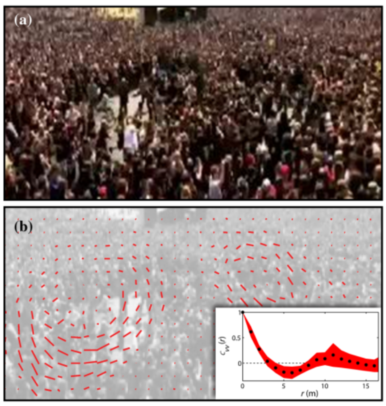
Content

Statistical systems

Collective behavior from a large number of degrees of freedom

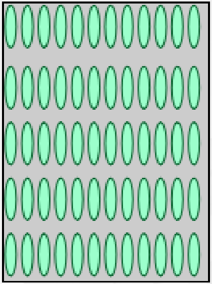


Collective Motion of Humans in Mosh and Circle Pits at Heavy Metal Concerts [Silverberg et al. 2013]

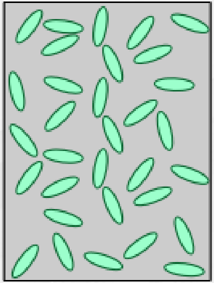
Order parameter : mean people's density (conserved)

Statistical systems

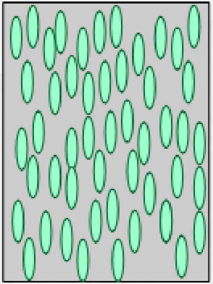
Collective behavior from a large number of degrees of freedom



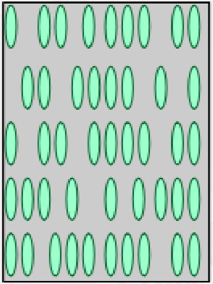
Solid phase: orientation and periodicity



Liquid phase: no orientation or periodicity



Nematic phase: orientation, no periodicity



Smectic phase: orientation with some periodicity

Schematics of liquid crystals [Chem.libretext.org]

Order parameter : mean liquid crystal's orientation (non-conserved)

Statistical field theory

Continuous field $\phi(\mathbf{x},t)$: density, magnetization, orientation... [Hohenberg et Halperin 1977 (18),Bray 1994 (22)]

Total order parameter : $\Phi(t) = \int d\mathbf{x}\phi(\mathbf{x},t)$

Brownian field dynamics

$$\frac{\partial\phi(\mathbf{x})}{\partial t} = -L\frac{\delta H}{\delta\phi(\mathbf{x})} + \eta(\mathbf{x},t)\ ,\ \langle\eta(\mathbf{x},t)\eta(\mathbf{x}',t)\rangle = \delta(t-t')\Gamma(\mathbf{x},\mathbf{x}') \quad (1)$$

$$p(\phi) = \exp(-\beta H[\phi])\ ,\ \Gamma(\mathbf{x},\mathbf{x}') = 2TL(\mathbf{x},\mathbf{x}') \quad (2)$$

Grand Canonical ensemble : **model A**

$\Phi(t) \neq cte$
$L(\mathbf{x},\mathbf{x}') = \alpha\delta(\mathbf{x}-\mathbf{x}')$,

$$\frac{\partial\phi(\mathbf{x})}{\partial t} = -\alpha\frac{\delta H}{\delta\phi(\mathbf{x})} + \eta(\mathbf{x},t) \quad (3)$$

Discrete : **Glauber** dynamics

Canonical ensemble : **model B**

$\Phi(t) = cte$
$L(\mathbf{x}-\mathbf{x}') = -D\nabla^2\delta(\mathbf{x}-\mathbf{x}')$

$$\frac{\partial\phi(\mathbf{x})}{\partial t} = \nabla\cdot\left[D\nabla\frac{\delta H}{\delta\phi(\mathbf{x})} + \boldsymbol{\eta}(\mathbf{x},t)\right] \quad (4)$$

Discrete : **Kawasaki** dynamics

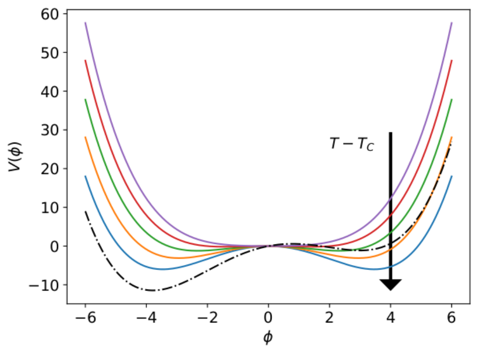
Phase separation

Ginzburg-Landau Hamiltonian [Landau et Lifschitz 1990 (1)]

$$H[\phi] = \int d\mathbf{x} \quad \underbrace{\frac{\kappa}{2}[\nabla\phi]^2}_{\text{cost of changing } \phi} \quad + \quad \underbrace{V(\phi)}_{\text{phase separating potential}} \tag{5}$$

ϕ^4 potential

$$V(\phi) = \frac{1}{2}(T - T_C)\phi^2 + \frac{\lambda}{4!}\phi^4 \tag{6}$$



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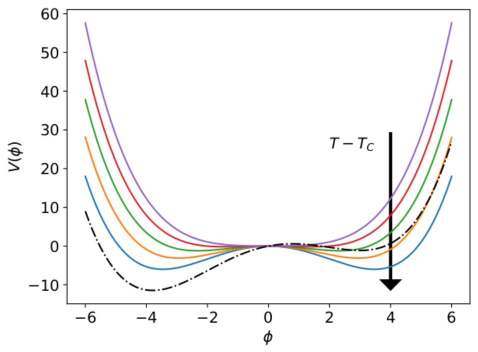
ϕ^4 potential

$$V(\phi) = \frac{1}{2}(T - T_C)\phi^2 + \frac{\lambda}{4!}\phi^4 + \mathbf{B}\phi \tag{7}$$



Interface between two coexisting phases

Mean-field equilibrium system (dynamic independent)

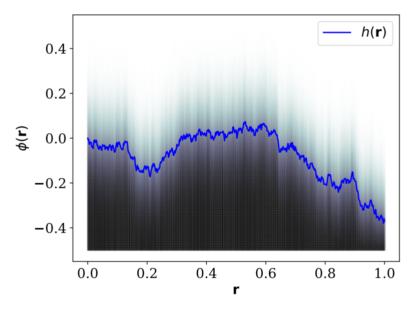
$$\cancel{\frac{\partial \phi(\mathbf{x})}{\partial t}} = -L \frac{\delta H}{\delta \phi(\mathbf{x})} + \cancel{\eta(\mathbf{x}, t)} \implies \frac{\delta H}{\delta \phi(\mathbf{x})} = 0 \tag{8}$$

ϕ^4 mean-field kink solution ($h = 0$)

$$\phi_K(z) = \tanh\left(\frac{z}{\xi_\perp}\right) \tag{9}$$

Interface SFT approximation

$$\phi(\mathbf{x}, t) = f(z - h(\mathbf{r}, t)) \tag{10}$$
$$\phi_K(z) = f(z) \tag{11}$$



Interface schematics

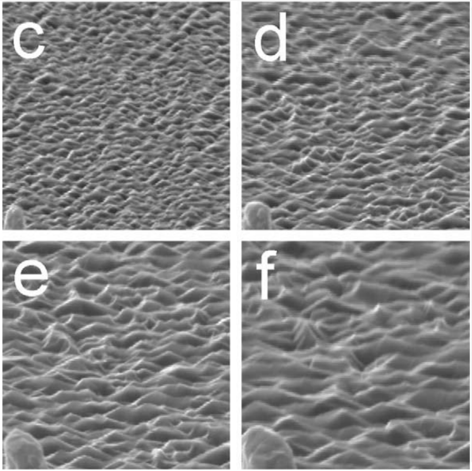
Interface examples

Equilibrium (boundary conditions), grand-canonical (discrete model A - Glauber)

2D Ising Monte Carlo simulation, $T = 0.85T_C$

Interface examples

Out-of-equilibrium (initial conditions) surface growth, grand-canonical (model A)



SEM movie showing a gallium arsenide film grown on silicon [Finnie et al. 2000]

Interface examples

Equilibrium, canonical (model B)

Water-oil interface

Interface examples

Out-of-equilibrium (initial conditions), canonical (model B)

Pigment concentration in cocktails [Cocktailmag.fr]

Surface tension

Definition by the free energy [Abrahams et Reed 76 (46)]

$$\sigma = f_{interface} - f_{homogeneous}[E \cdot L^{-2}] \tag{12}$$

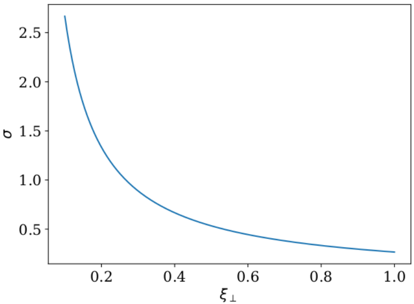
Cahn-Hilliard estimate [Cahn et Hilliard 58 (25)]

$$\sigma = \int dz\ \kappa \left(\frac{d\phi_K(z)}{dz}\right)^2 \tag{13}$$

Limits

- $\xi_\perp \to 0$: sharp interface
- $\xi_\perp \to \infty$: ultra-low surface tension, critical systems

Surface tension with respect to interface width $\xi_\perp$



Confinement forces [chapter 3]

Excess free energy and Casimir force [Gambassi 09 (3)]

$$F(t,h,L) = L'^2 \left(L f_{bulk} + \beta^{-1} f_{ex} \right) \quad (14)$$

$$f_{casimir} = -\beta^{-1} \frac{\partial f_{ex}}{\partial L} \quad (15)$$

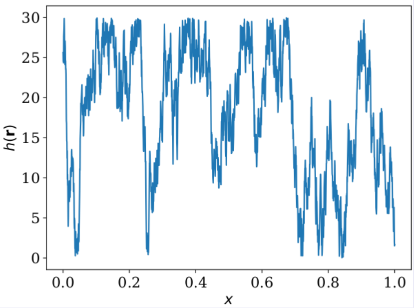
1D Confined elastic line $0 < h(\mathbf{r}, t) < L$

$$f = \frac{T^2 \pi^2}{2\sigma L^2} \quad (16)$$

Finite-size effects surface tension [Privman 1988 (16)]

$$\sigma(L) \simeq \sigma_{bulk} + \frac{Ta}{L} \quad (17)$$

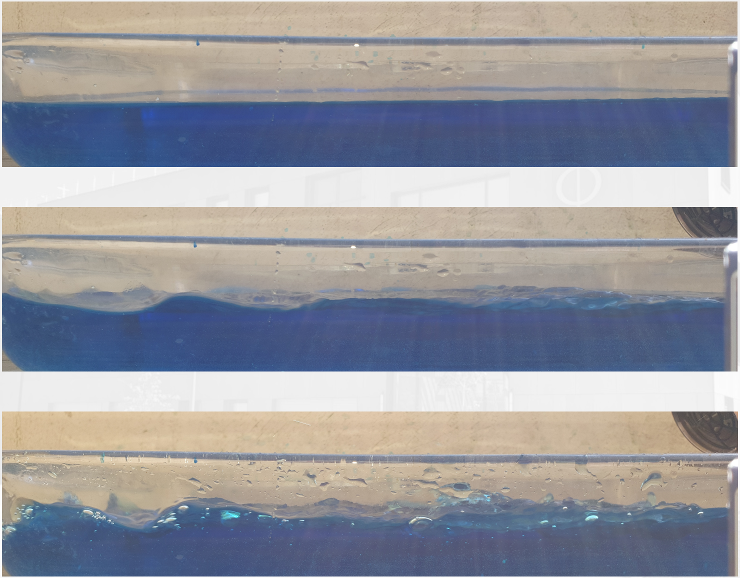
$$f_c = \frac{T\pi^2}{2aL} \quad (18)$$



Confined interface for $L_Y = 30$ schematics

How do out-of-equilibrium steady-states affect interfaces ?

Driving at the interface



Home-made wind generated waves (from a hair dryer)

The Solid-On-Solid model

Solid-On-Solid model

Lattice model with interface height h_i

$$H = J \sum_{i=0}^{L'} |h_i - h_{i+1}| + \mu \sum_{i=0}^{L'} \frac{h_i + h_{i+1}}{2} \quad (19)$$

Metropolis algorithm

- Glauber

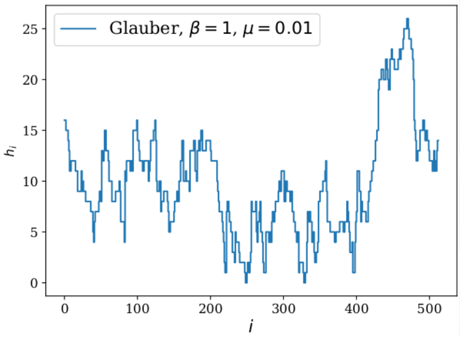
$$\{h_i\} \to \{h_i \pm 1\} \quad (20)$$

- Kawasaki ($\sum h_i = cte$)

$$\{h_i, h_{i\pm1}\} \to \{h_i - 1, h_{i\pm1} + 1\} \quad (21)$$

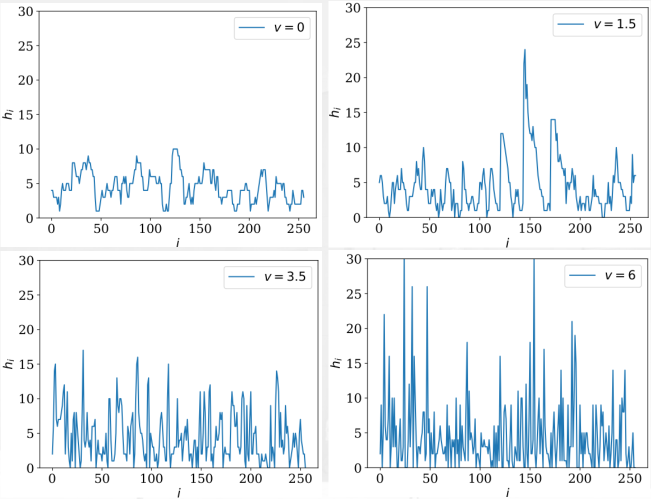
- New configuration with probability

$$A(C \to C') = \exp(-\beta \Delta H) \quad (22)$$



Driving at the interface

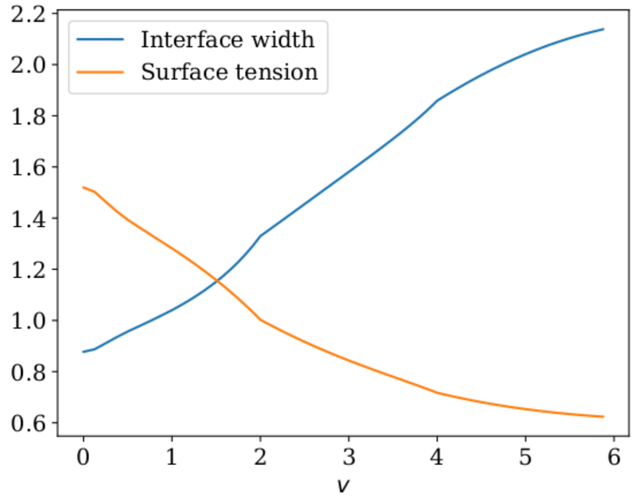
Driving : $\Delta E_d = \Delta E_{eq} \pm v$ in Kawasaki dynamics



$$\beta = 1, L_Y = 200, \frac{1}{L'} \sum h_i = 4.51$$

Driving at the interface

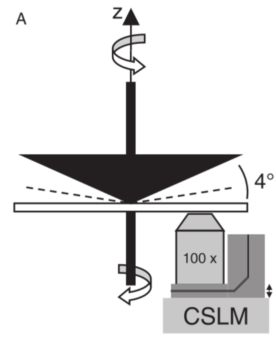
Driving : $\Delta E_d = \Delta E_{eq} \pm v$ in Kawasaki dynamics



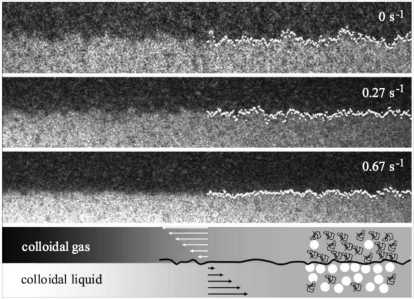
Monte Carlo driving of SOS interface at $\beta = 1$ and $\frac{1}{L'} \sum h_i = 4.51$

What happens if we shear not only the interface, but the whole system ?

PMMA polymers in polystyrene solvent under shear [Derks et al. 2006]

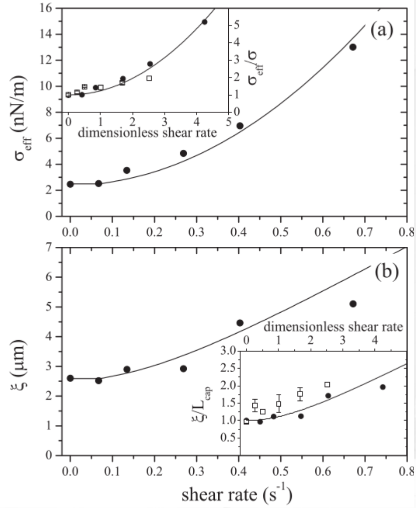


Experimental setup
[Derks et al. 2004]



Interface snapshots under shear
[Derks at al. 2006]

PMMA polymers in polystyrene solvent under shear [Derks et al. 2006]



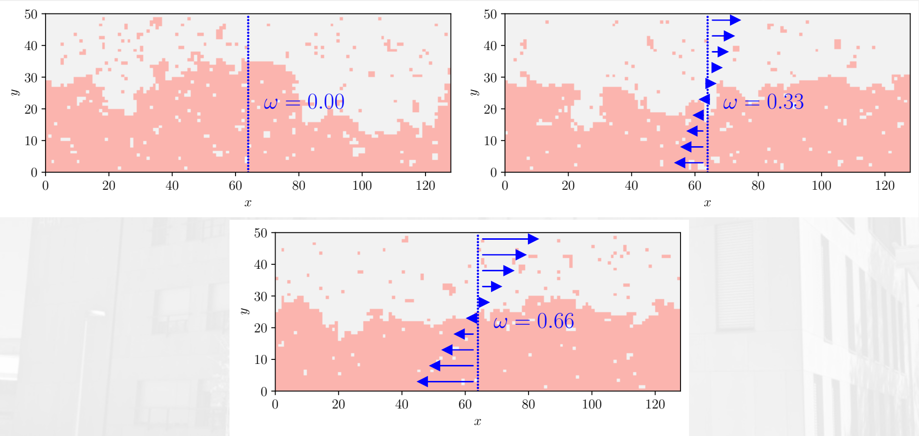
Effective interfacial tension and correlation length along the interface

Phenomenolocial model

Modes slower than shear rate no longer contribute to the entropy of the interface

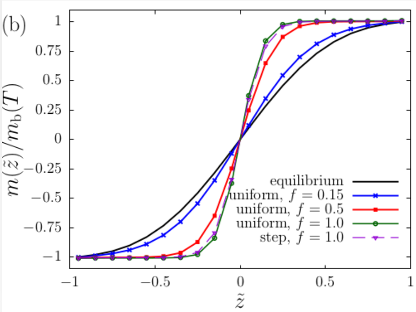
$$\sigma_{eff}(\dot{\gamma}) = \sigma + \frac{3k_BT}{4\pi}\frac{\dot{\gamma}\tau_{cap}}{L_{cap}^2}\sqrt{(\dot{\gamma}\tau_{cap})^2 - 1} \tag{23}$$

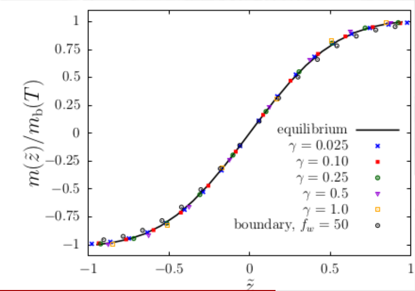
2D sheared or driven Ising model [T.H.R. Smith 2010 (71)]

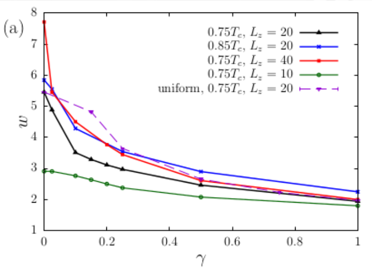


Snapshots of 2D Ising driven systems for $T = 0.9T_C$

2D sheared or driven Ising model [T.H.R. Smith 2010 (71)]

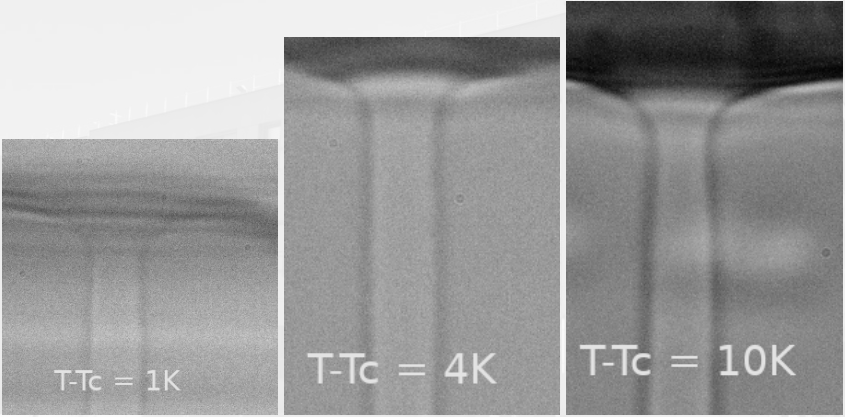






Magnetisation profiles, rescaled magnetisation profiles, interfacial width with respect to shear for different L_Y

Binary mixture under laser radiation pressure [Delville et al. 08 (J. Opt. A)]

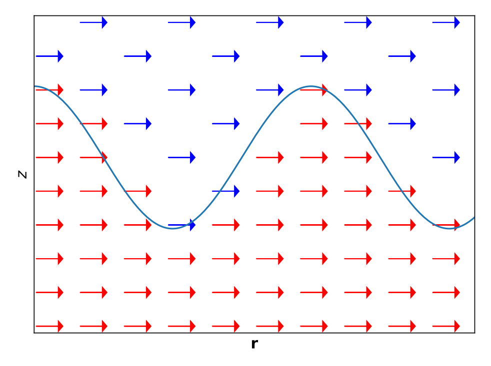


Quaternary liquid mixture made of toluene, sodium dodecyl sulfate (SDS), n-butanol and water gives 2 separate micellar phases

Imposed hydrodynamic flow

Advecting the field $\psi(\mathbf{x}, t)$ with a flow $\mathbf{v}(\mathbf{x})$ gives

$$\frac{\partial \psi(\mathbf{x}, t)}{\partial t} + \nabla \cdot (\mathbf{v}\psi(\mathbf{x}, t)) = D\nabla^2 \frac{\delta H}{\delta \psi(\mathbf{x})} + \sqrt{2DT}\nabla \cdot \boldsymbol{\eta}_1(\mathbf{x}, t) \tag{24}$$



Uniform advecting flow $\mathbf{v}(\mathbf{x}, t) = v\mathbf{e}_x$ in the **sharp interface approximation**, galilean invariance $\mathbf{x} \to \mathbf{x} + vt$

The effect of driving on model C interfaces [Dean, Gersberg,Holdsworth 2020 (17)]

ψ : colloid field under model B dynamics
ϕ : passive solvent under model A dynamics

$$H[\psi,\phi] = H_1[\psi] + H_2[\psi,\phi] \tag{25}$$

$$H_1[\psi] = \int d\mathbf{x} \left[\frac{\kappa}{2}[\nabla\psi(\mathbf{x})]^2 + V(\psi(\mathbf{x})) - gz\psi(\mathbf{x})\right] \tag{26}$$

$$H_2[\psi,\phi] = \int d\mathbf{x} \frac{\lambda}{2}(1-\psi(\mathbf{x})-\phi(\mathbf{x}))^2 \tag{27}$$

κ : interface energy
V : phase separating potential
g : gravitational term introducing finite size correlations
λ : coupling parameter

Model C dynamics

Equations

$$\frac{\partial\psi(\mathbf{x},t)}{\partial t} + \mathbf{v}\cdot\nabla\psi(\mathbf{x},t) = D\nabla^2\frac{\delta H}{\delta\psi(\mathbf{x})} + \sqrt{2DT}\nabla\cdot\boldsymbol{\eta}_1(\mathbf{x},t) \quad (28)$$

$$\frac{\partial\phi(\mathbf{x},t)}{\partial t} = -\alpha\frac{\delta H}{\delta\phi(\mathbf{x})} + \sqrt{2\alpha T}\eta_2(\mathbf{x},t) \quad (29)$$

are coupled by H_2, with a closed form on ψ

$$\underbrace{\frac{\partial\psi(\mathbf{x},t)}{\partial t}}_{\text{evolution}} \underbrace{- \lambda D\nabla^2\int_{-\infty}^{t} dt' \exp(-\alpha\lambda(t-t'))\frac{\partial\psi(\mathbf{x},t')}{\partial t}}_{\text{coupling term}} + \underbrace{\mathbf{v}\cdot\nabla\psi(\mathbf{x},t)}_{\text{advection}} = \underbrace{D\nabla^2\mu(\mathbf{x},t')}_{\text{model B}} + \underbrace{\zeta(\mathbf{x},t)}_{\text{thermal noise}} \quad (30)$$

where

$$\mu(\mathbf{x},t) = \frac{\delta H_1}{\delta\psi(\mathbf{x},t)} \quad (31)$$

and

$$\tilde{\zeta}(\mathbf{x},\omega) = \frac{\sqrt{2\alpha TD\lambda}}{i\omega+\alpha\lambda}\nabla^2\tilde{\eta}_2(\mathbf{x},\omega) + \sqrt{2DT}\nabla\cdot\tilde{\boldsymbol{\eta}}_1(\mathbf{x},\omega). \quad (32)$$

Model C interface dynamics

Interface approximation $\psi(\mathbf{x},t) = f(z - h(\mathbf{r},t))$ using method of Bray et al. 2001 (9,29)] at first order in h

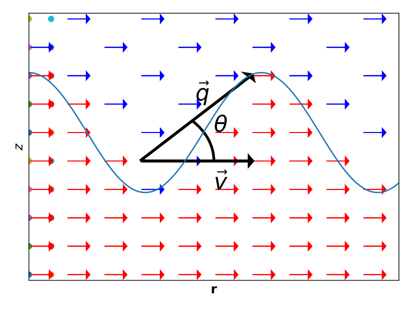
$$\Delta\psi^2 \int d\mathbf{r} G(0,\mathbf{r}-\mathbf{r}')[\frac{\partial h(\mathbf{r},t)}{\partial t} + \mathbf{v}\cdot\nabla h(\mathbf{r},t)] = \sigma[\nabla^2 h(\mathbf{r},t) - m^2 h(\mathbf{r},t)] + \xi(\mathbf{r},t)$$
$$+\frac{\sigma\lambda D}{\kappa}\int_{-\infty}^{t} dt' \exp(-\alpha\lambda(t-t'))\frac{\partial h(\mathbf{r},t')}{\partial t'} \tag{33}$$

where $G = -\nabla^{-2}$, $\xi(\mathbf{r},t) = \int_{-\infty}^{\infty} dz f'(z-h(\mathbf{r},t))\nabla^{-2}\zeta(\mathbf{x},t)$, $m^2 = \Delta\psi g/\sigma$

Model C interface correlation function

Correlation function

$$\tilde{C}_s(\mathbf{q}) = T\frac{\left(2D\sigma q(\kappa[q^2+m^2]+\lambda)+\alpha\kappa\lambda\Delta\psi^2\right)^2+\kappa^2\Delta\psi^4(\mathbf{q}\cdot\mathbf{v})^2}{\sigma[q^2+m^2]\left(2Dq\sigma(\kappa[q^2+m^2]+\lambda)+\alpha\kappa\lambda\Delta\psi^2\right)^2+\kappa\left(\kappa\sigma[q^2+m^2]+\lambda\sigma\right)\Delta\psi^4(\mathbf{q}\cdot\mathbf{v})^2} \tag{34}$$



No driving $v \to 0$

$$\tilde{C}_s(\mathbf{q}) = \frac{T}{\sigma[q^2+m^2]} \tag{35}$$

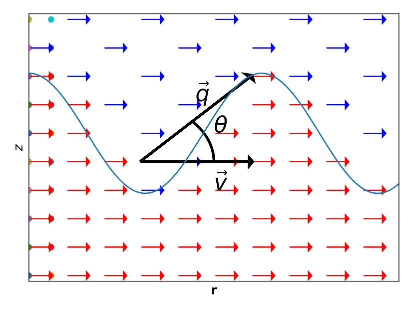
Infinite driving $v \to \infty$

$$\tilde{C}_s(\mathbf{q}) = \frac{T}{\sigma[q^2+m^2+\frac{\lambda}{\kappa}]} \tag{36}$$

Model C interface correlation function

Correlation function

$$\tilde{C}_s(\mathbf{q}) = T \frac{\left(2D\sigma q(\kappa[q^2+m^2]+\lambda)+\alpha\kappa\lambda\Delta\psi^2\right)^2+\kappa^2\Delta\psi^4(\mathbf{q}\cdot\mathbf{v})^2}{\sigma[q^2+m^2]\left(2Dq\sigma(\kappa[q^2+m^2]+\lambda)+\alpha\kappa\lambda\Delta\psi^2\right)^2+\kappa\left(\kappa\sigma[q^2+m^2]+\lambda\sigma\right)\Delta\psi^4(\mathbf{q}\cdot\mathbf{v})^2} \tag{34}$$



Small q approximation

- Angle-dependent surface tension

$$\sigma_s(\theta) = \sigma\left(1+\frac{v^2\cos^2(\theta)}{{v_0}^2}\right) \tag{37}$$

- Angle-depentent correlation length

$$\xi_s = \xi_{eq}\sqrt{1+\frac{v^2\cos^2(\theta)}{{v_0}^2}} \tag{38}$$

- Intrinsic velocity

$$v_0 = \sqrt{\alpha^2\lambda\kappa} \tag{39}$$

Presentation's conclusion

Driven Solid-On-Solid model

- Explanation of the model
- Driven does increase interface width, as in wind generated waves

Driven Model C

- Similarities between shearing and driving
- Steady-state out-of-equilibrium driving needs a coupling between two fields due to Galilean invariance (model C)
- Increase of the effective surface tension in the direction of driving and also an increase in the correlation length of the height fluctuations with respect to a non-driven equilibrium interface
- Uniform driving leads to effective equilibrium statistics

Thesis' conclusion

Chapter 3 : Equilibrium Interface models and their finite size effects

- General method to compute free energy and probability distribution functions of continuous gaussian interfaces with path integral method
- Generalization the Lopes Cardozo-Jacquin-Holdsworth method to compute free energy in numerical Monte Carlo lattice systems for any type of external potentials, useful for Kawasaki dynamics
- Exact diagonalization of the finite Solid-On-Solid transfer matrix

Chapter 4 : Beyond Solid-On-Solid : the Particles-Over-Particles model

- New model generated from SOS with entropic term
- Multi-particles formulation
- Numerical Monte Carlo issues : corner case of Metropolis algorithm
- Actually is the model that gaves us the computational idea for model C (the paper)

Chapter 5 : Driven interfaces

- Presentation's conclusion

Overall conclusion

- Relationship between continuous gaussian and discrete solid-on-solid models (Eq. 3.84 and 3.184), with surface tension computations
- Casimir-type effect have an interesting manifestation in interface physics,
- Interface models have no bulk free energy, only excess free energy due to confinement
- Casimir-like effect is seen in interface models in the critical regime $\sigma \to 0$
- POP systems allow for particles coupling with different temperatures or thermodynamical ensembles (model C) and different hydrodynamical flows
- SFT explanation as to why driving increases surface tension and correlation length along the interface
- Driven steady-states do have equilibrium properties

Perspectives

- Thanks to the generalized free energy computation method, study the difference between thermodynamical ensembles on finite-size critical systems
- Get deeper on the relationship between critical and interface systems
- Solve POP's numerical problems. Study coupled systems as presented model C through it
- Better understand the difference between driving at interface and bulk driving
- Do out-of-equilibrium confined interfaces have different physics than equilibrium confined ones ?