Microscopic fluctuations in the spreading fronts of wetting liquid droplets

CompPhys24

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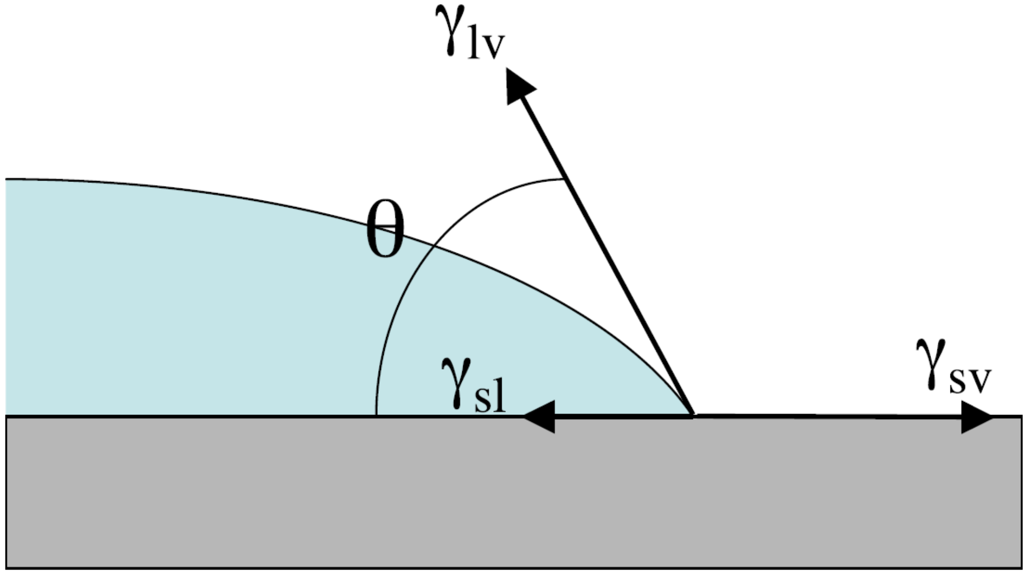
Outline of the talk

- Introduction: Wetting and Spreading phenomena
- Model: modified Ising lattice gas
- How we perform our kMC simulations
- Geometry of the system
- Numerical results
- Conclusions

Wetting phenomena



$$\gamma_{SV} = \gamma_{SL} + \gamma \cos \theta_{eq}$$

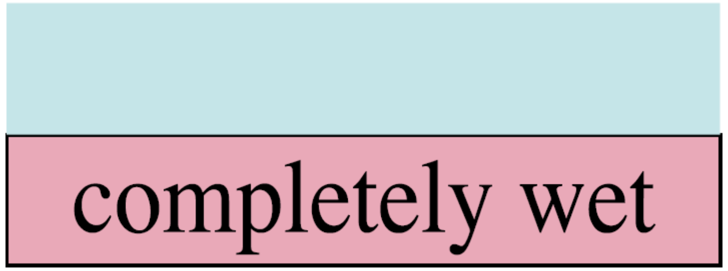
D. Bonn, J. Eggers, J. Indekeu, J. Meunier, and E. Rolley, *Wetting and Spreading,* Rev. Mod. Phys. **81**, 739 (2009)

Wetting phenomena

$$\gamma_{SV} < \gamma_{SL} + \gamma$$

Water

$$\gamma_{SV} = \gamma_{SL} + \gamma$$



Most oils

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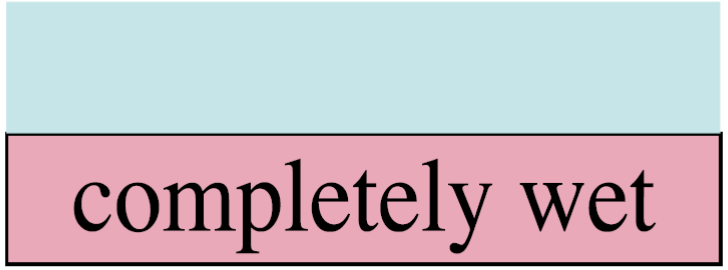

Most oils

Our case

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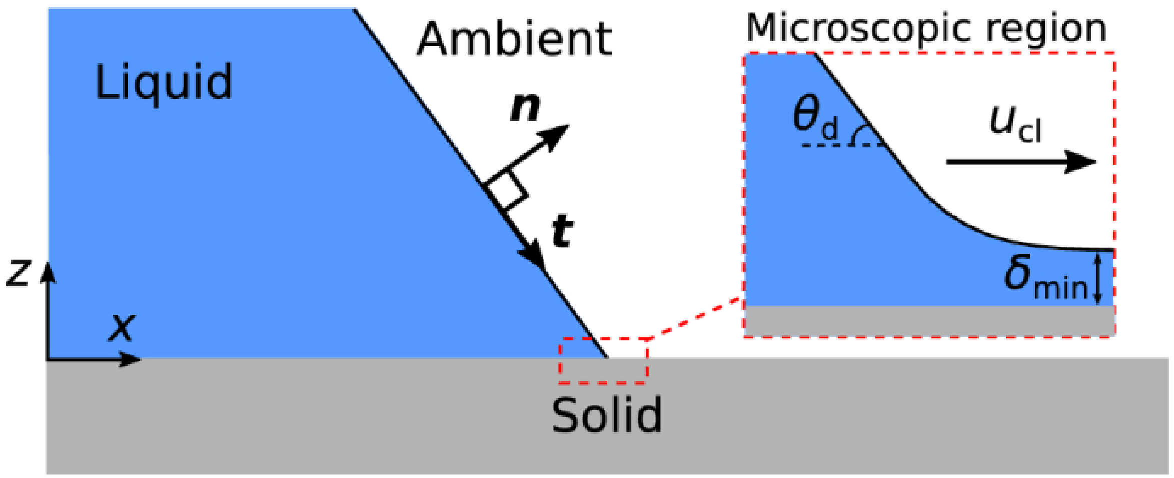
Spreading phenomena



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D. Beaglehole, Profiles of the precursor of spreading drops of siloxane oil on glass, fused silica, and mica. J. Phys. Chem. **93** 893 (1989)

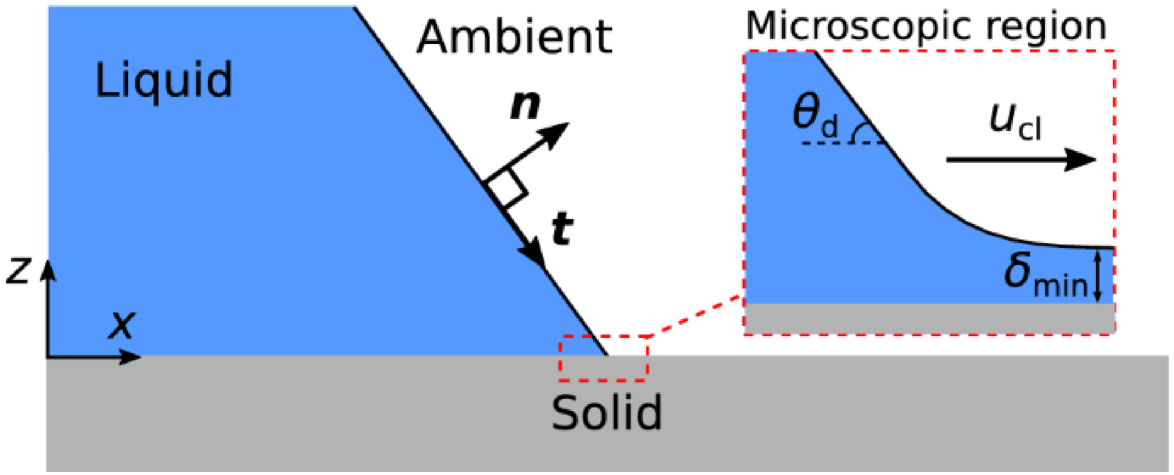
Spreading phenomena

Macroscopic Spreading

$$R_\mathrm{d} \sim t^{\delta_\mathrm{d}}$$

$$\delta_\mathrm{d} = 1/10$$

Tanner's Law



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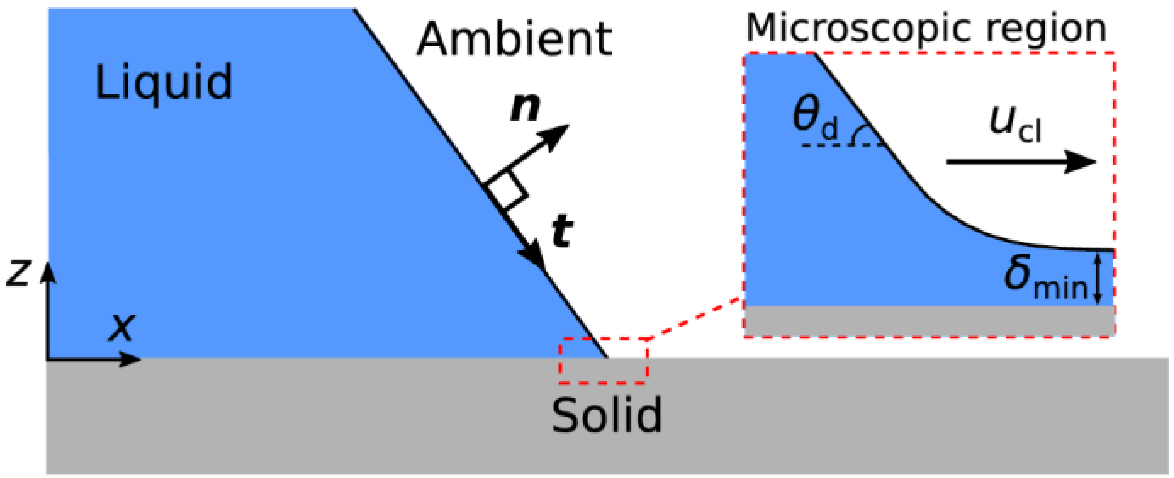

Thin Film Spreading

$$R \sim t^{\delta}$$

$$\delta \approx 1/2$$

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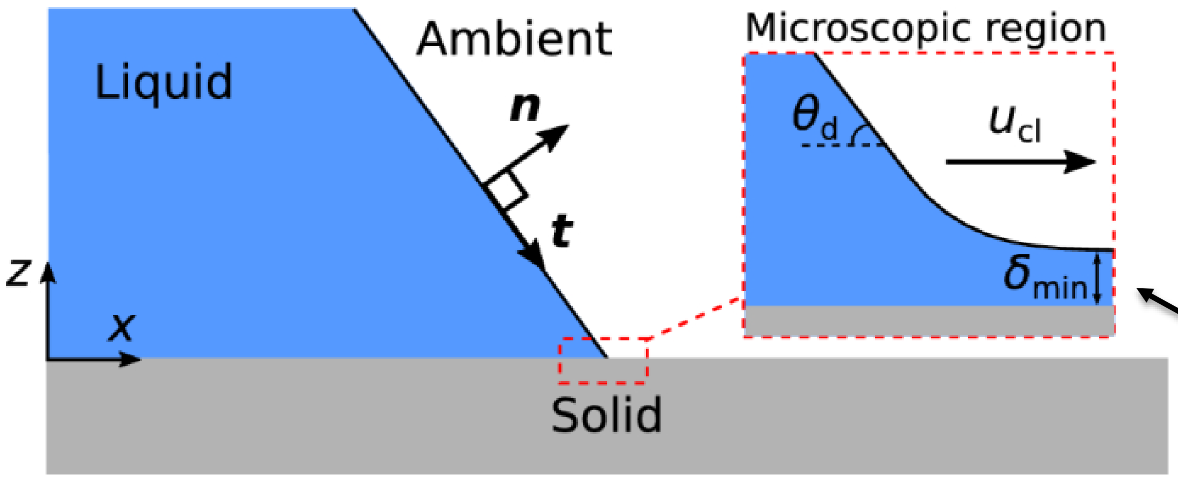

Thin Film Spreading

$$R \sim t^{\delta}$$

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Precursor film
(verified by ellipsometry)
Polydimethylsiloxane
(Silicone oil)

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Model: microscopic driven Ising lattice gas

$$\mathcal{H} = -J \sum_{\langle \mathbf{r},\mathbf{s} \rangle} n(\mathbf{r},t) n(\mathbf{s},t) - A \sum_{\mathbf{r}} \frac{n(\mathbf{r},t)}{Z^3}$$

D. B. Abraham, R. Cuerno, and E. Moro, *Microscopic Model for Thin Film Spreading*, Phys. Rev. Lett. **88**, 206101, 2002.

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Nearest neighbors interaction

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Nearest neighbors interaction — Interaction with substrate

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Nearest neighbors interaction

Interaction with substrate

$Z = 1$ Precursor layer

$Z = 2$ Supernatant layer

D. B. Abraham, R. Cuerno, and E. Moro, *Microscopic Model for Thin Film Spreading*, Phys. Rev. Lett. **88**, 206101, 2002.

kMC simulation details

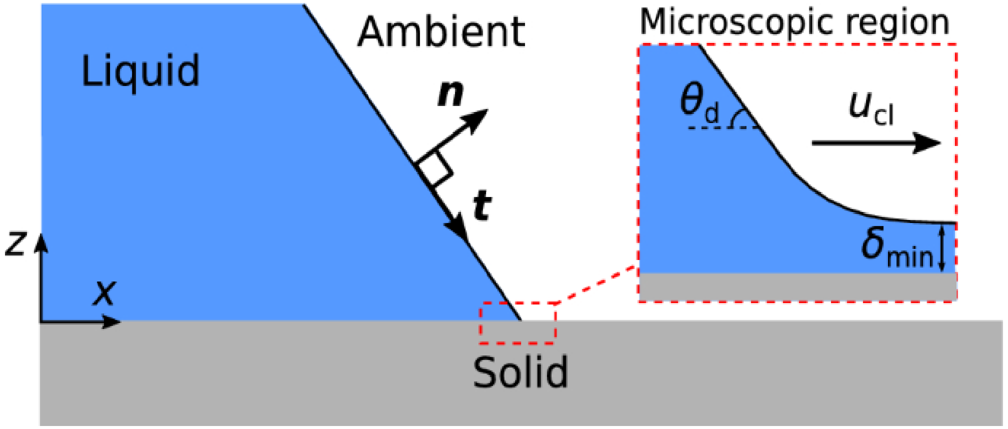
- Two overlapping 2D lattices $L_x \times L_y$
  $Z = 1$ Precursor layer
  $Z = 2$ Supernatant layer

- Kawasaki local dynamics:
  (spin exchange dynamics as in the **COP Ising Model**)
  COP Ising: constant Magnetization → constant Number Particles. **Growth?**

- Continuous time. We fix the number of exchanges performed:
  We do not reject exchanges

Droplet reservoir

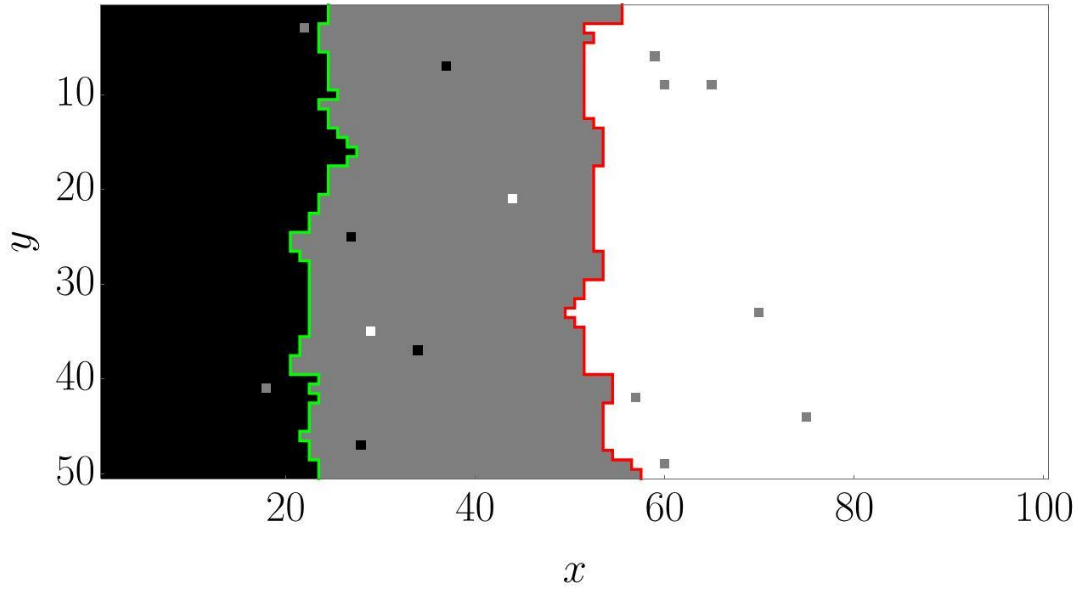
- Reservoir: represents the macroscopic droplet that feeds the film growth
- Boundary condition:
  - Initially the only cells that are filled in our system
  - Cells that are always filled:
  - If any exchange leads to a vacancy
    in the reservoir they are automatically refilled



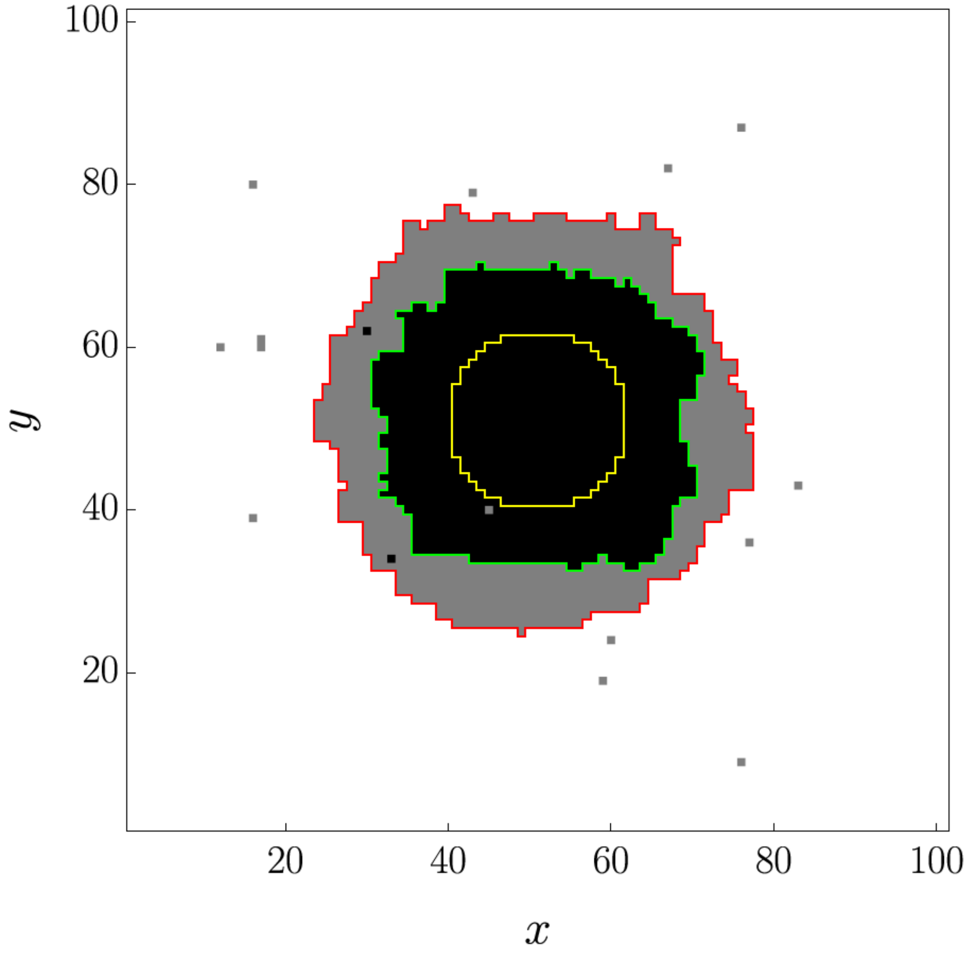
- Necessary for the system to grow as the dynamics only exchanges cells.

Geometry: layers and fronts

Band geometry



Radial geometry



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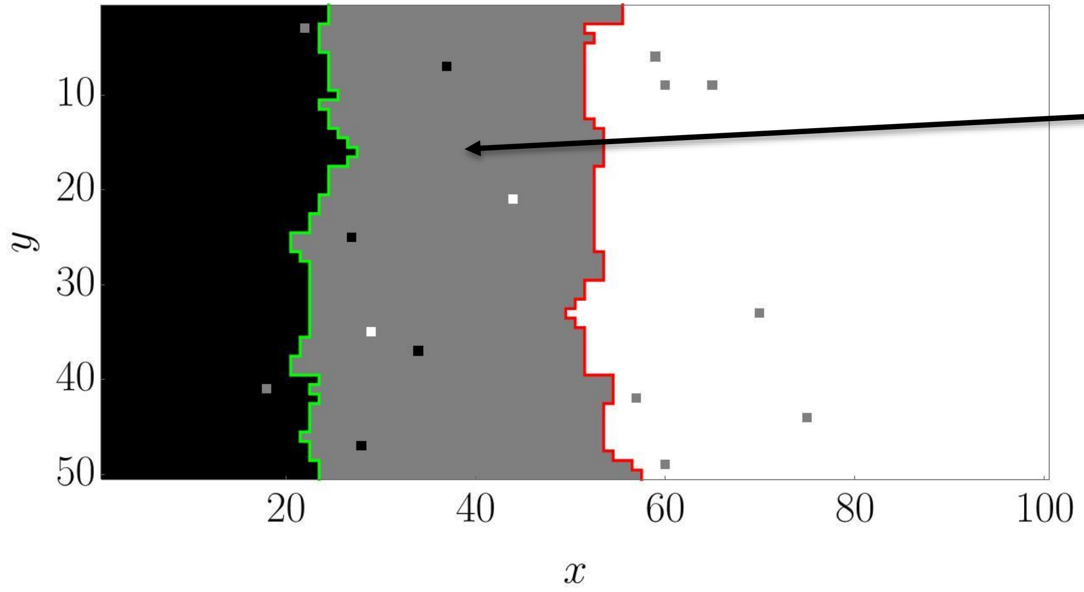
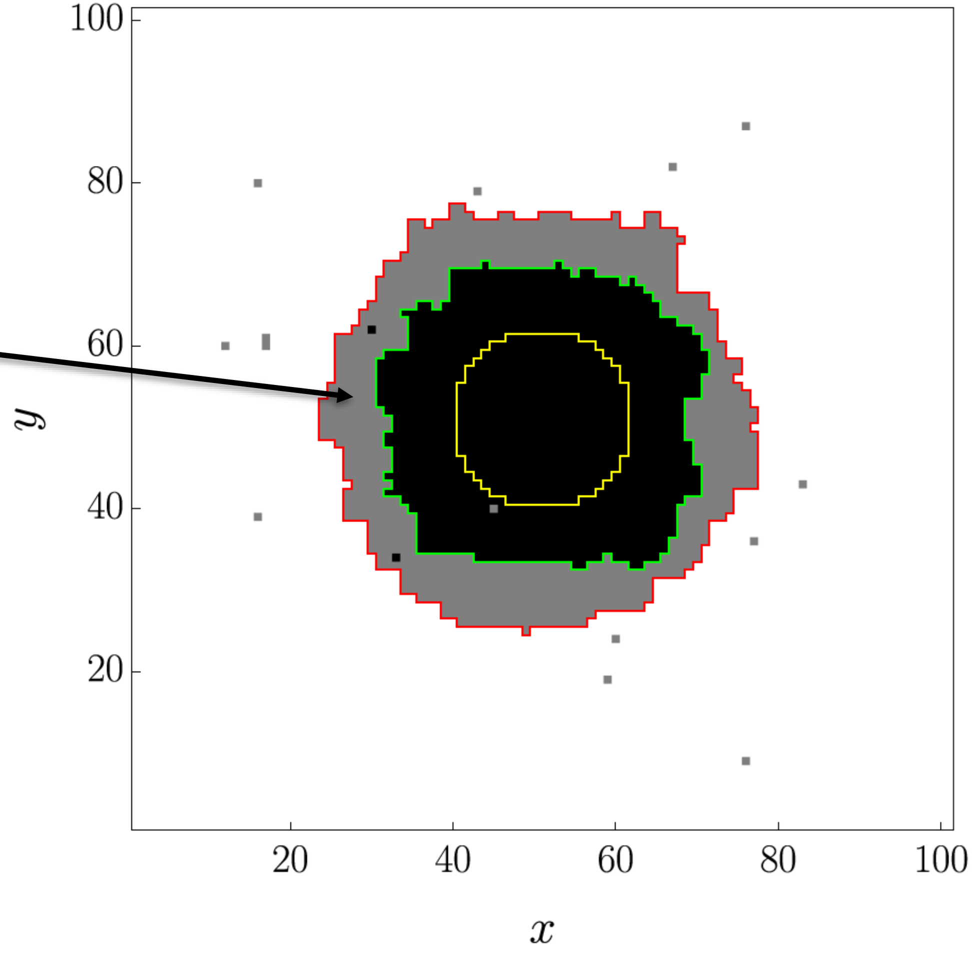


Layers

Radial geometry



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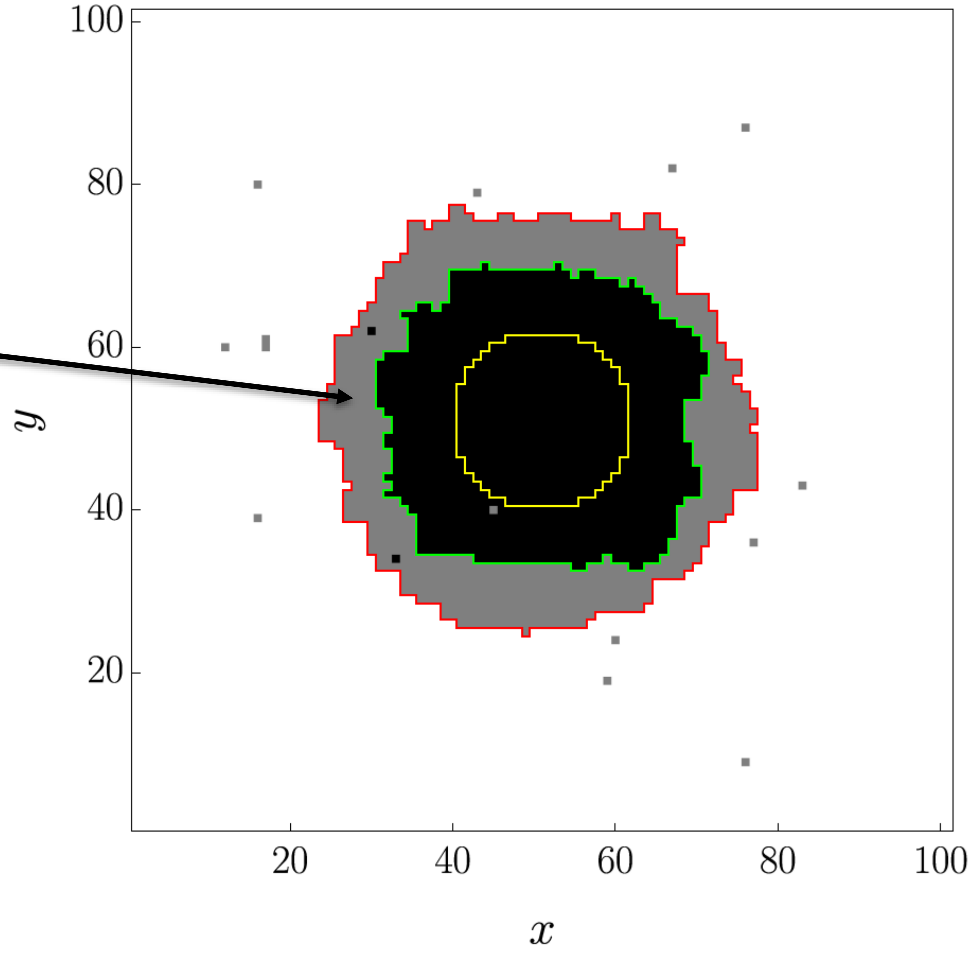

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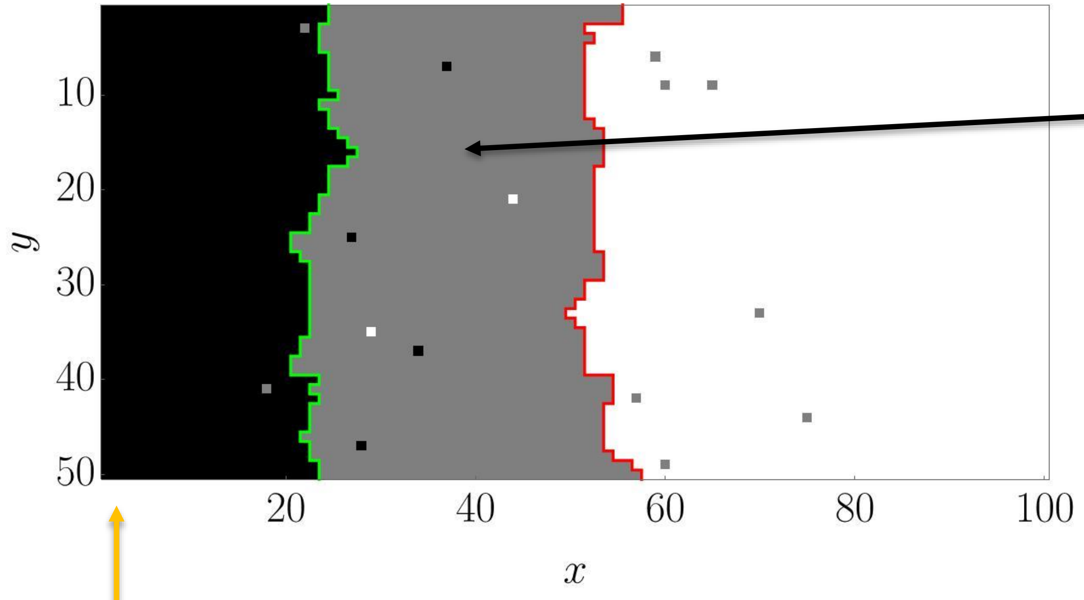
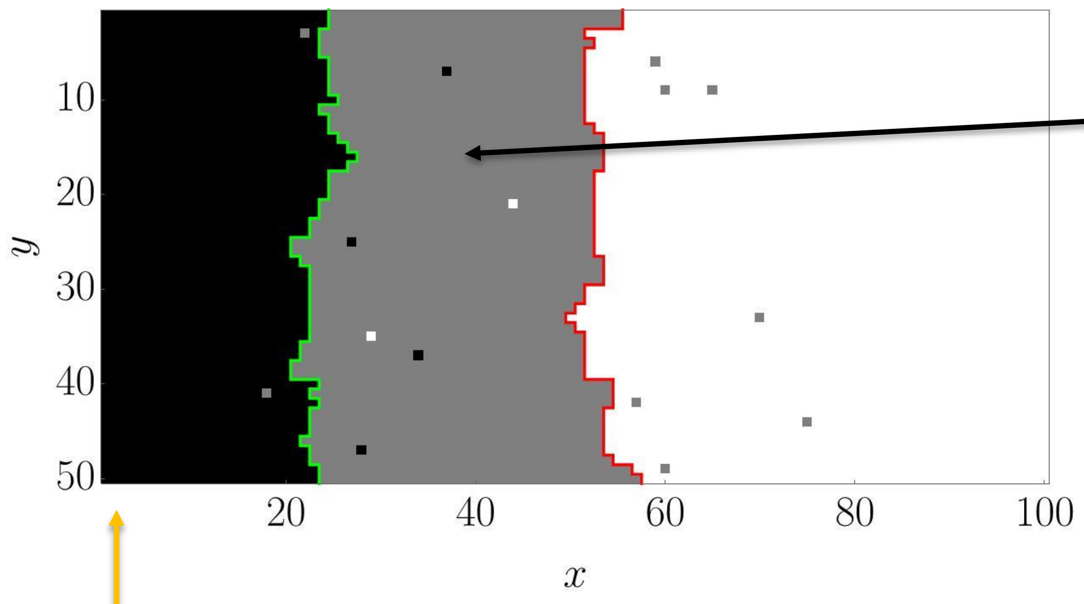


Layers

Droplet reservoir is
the first column

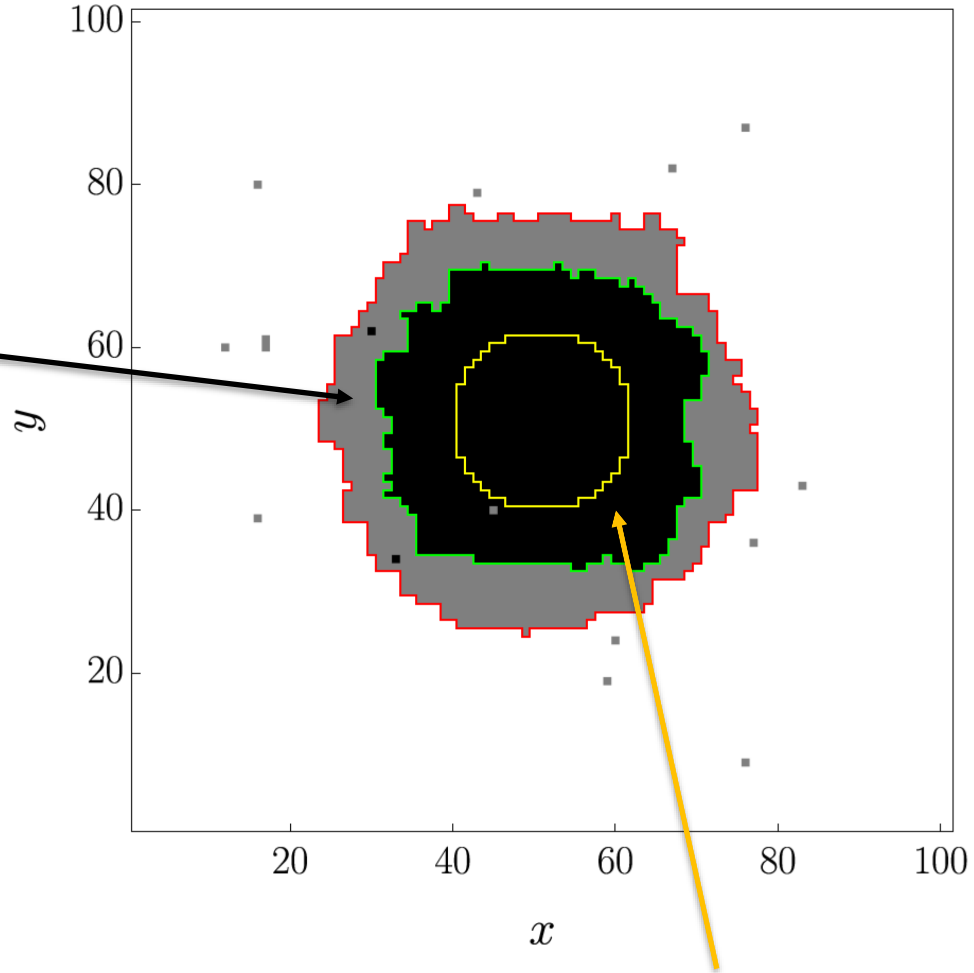
Geometry: layers and fronts

Band geometry



Layers

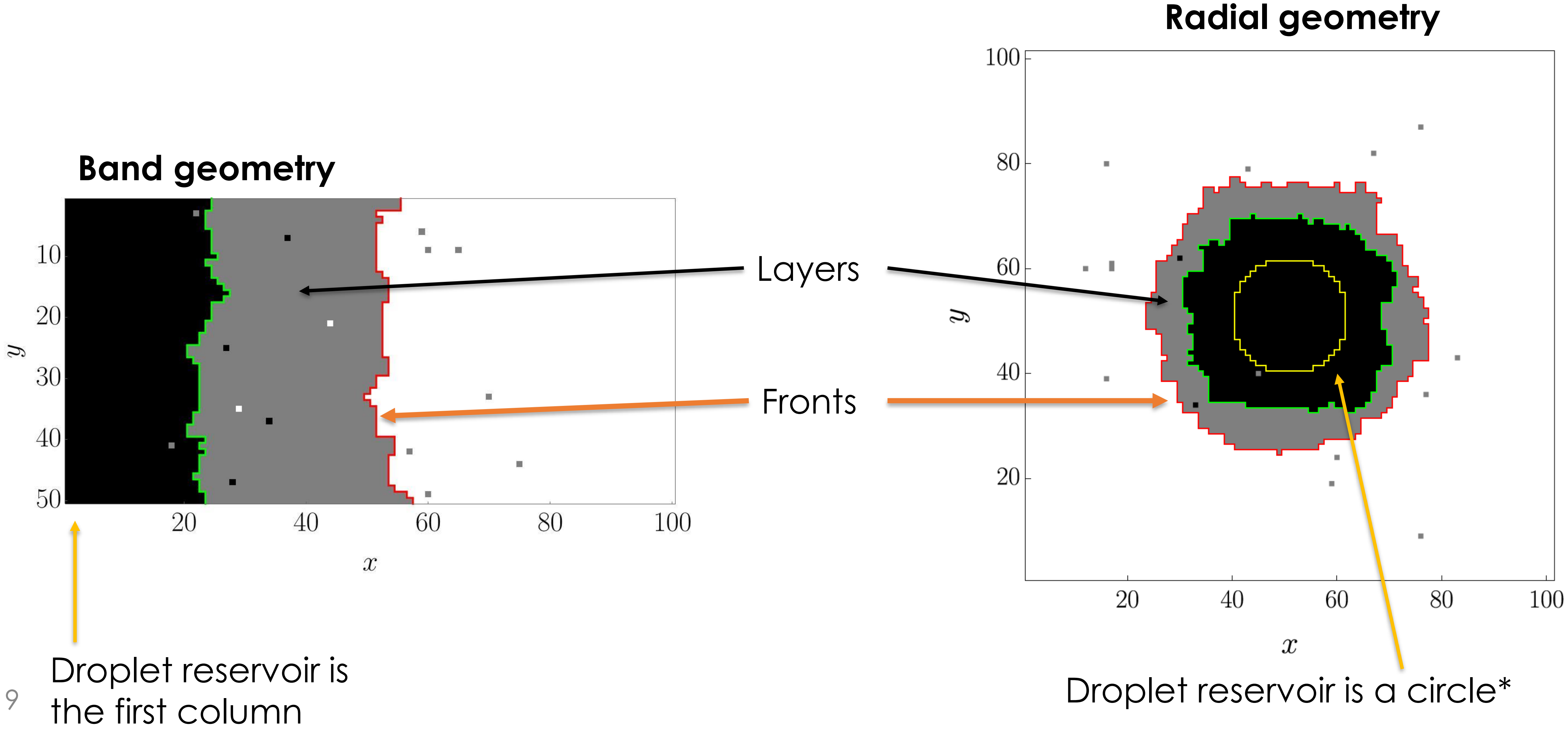
Radial geometry



Droplet reservoir is the first column

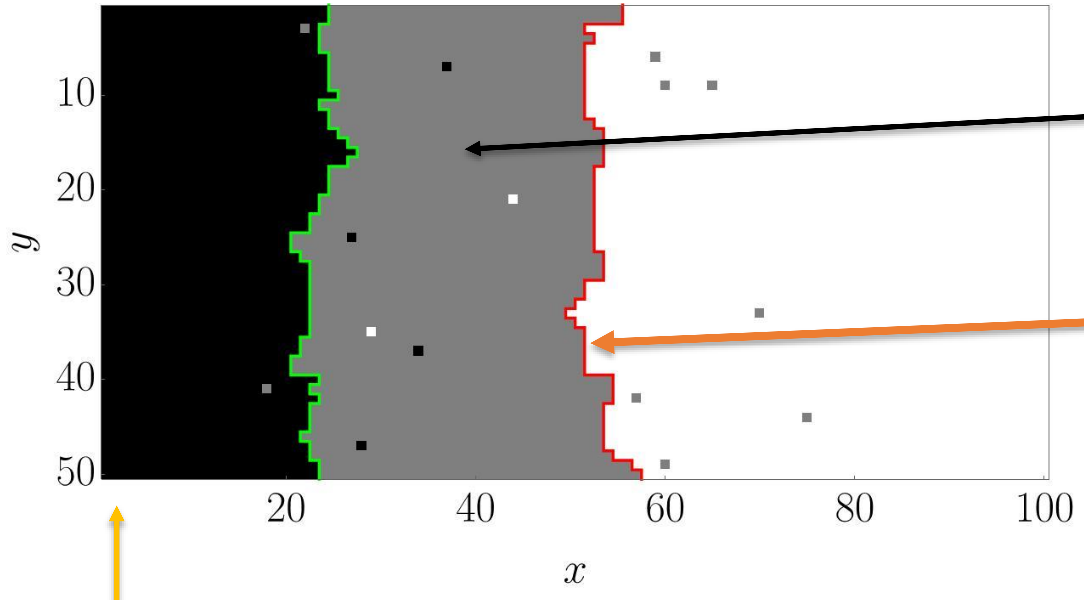
Droplet reservoir is a circle*

Geometry: layers and fronts

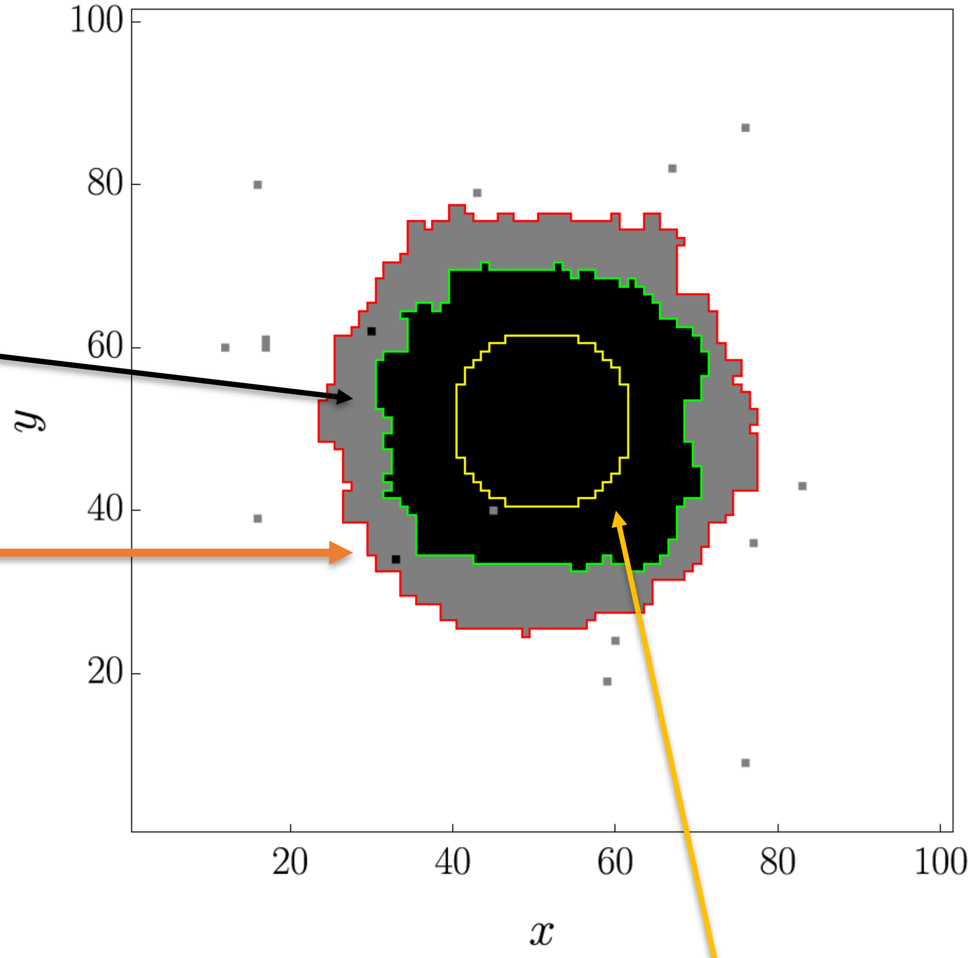


Geometry: layers and fronts

Band geometry



Radial geometry



Layers

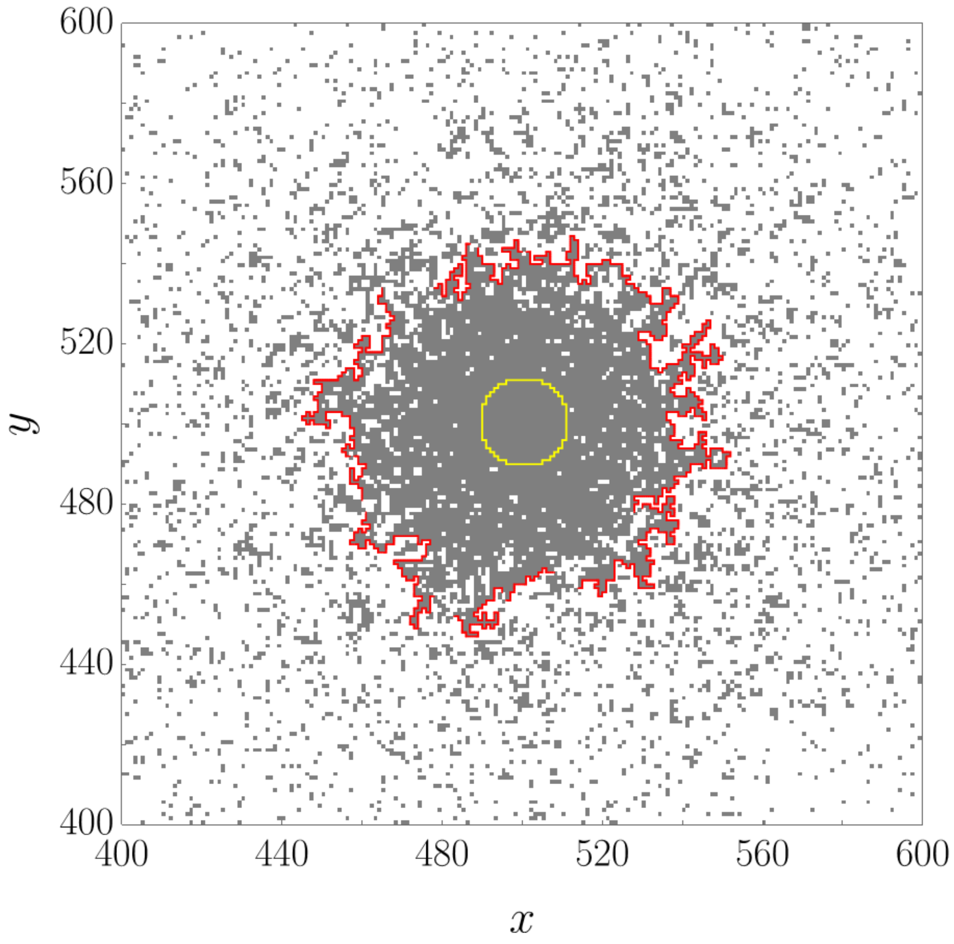
Fronts

Clustering method definition

Droplet reservoir is the first column

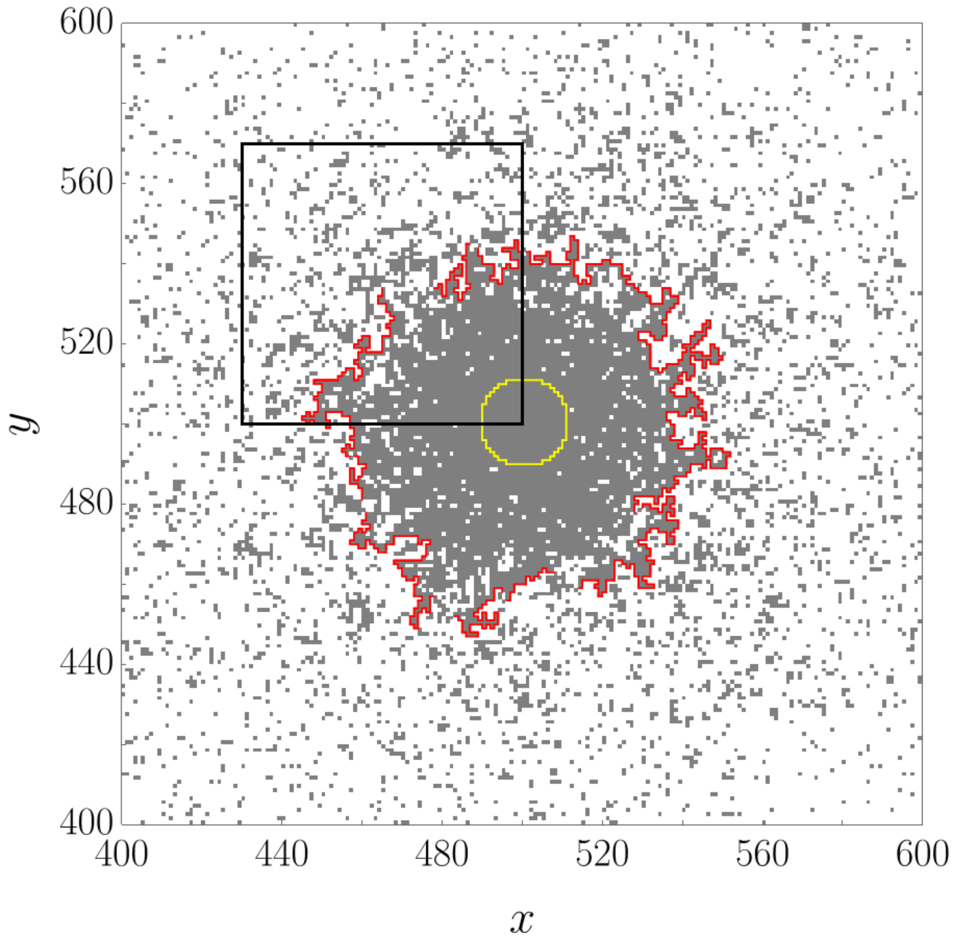
Droplet reservoir is a circle*

Measuring the fronts: clustering method



Here, we only show the bottom layer.

Measuring the fronts: clustering method



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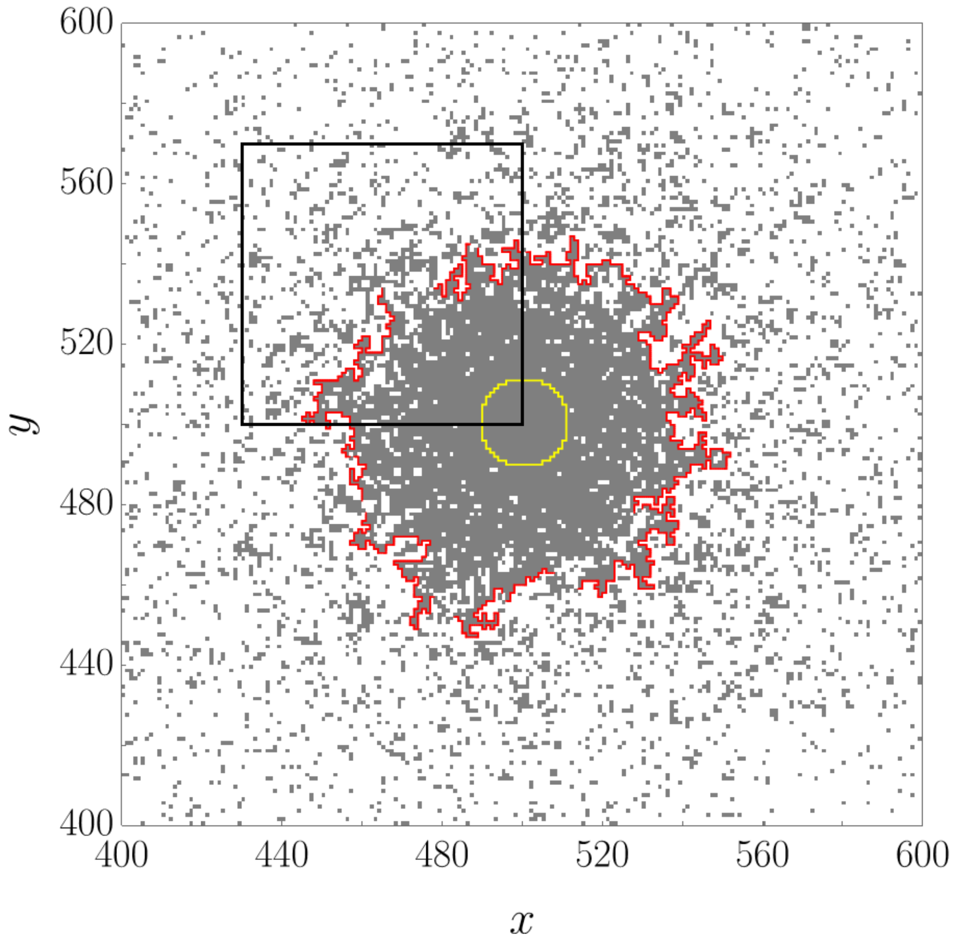

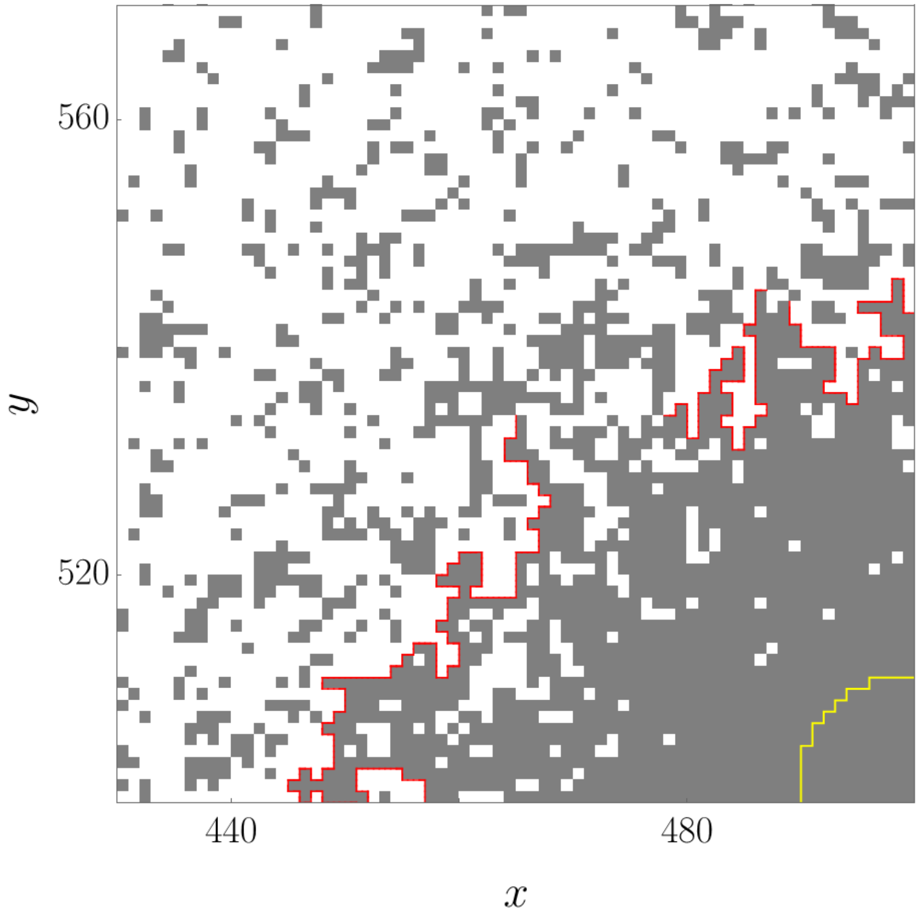

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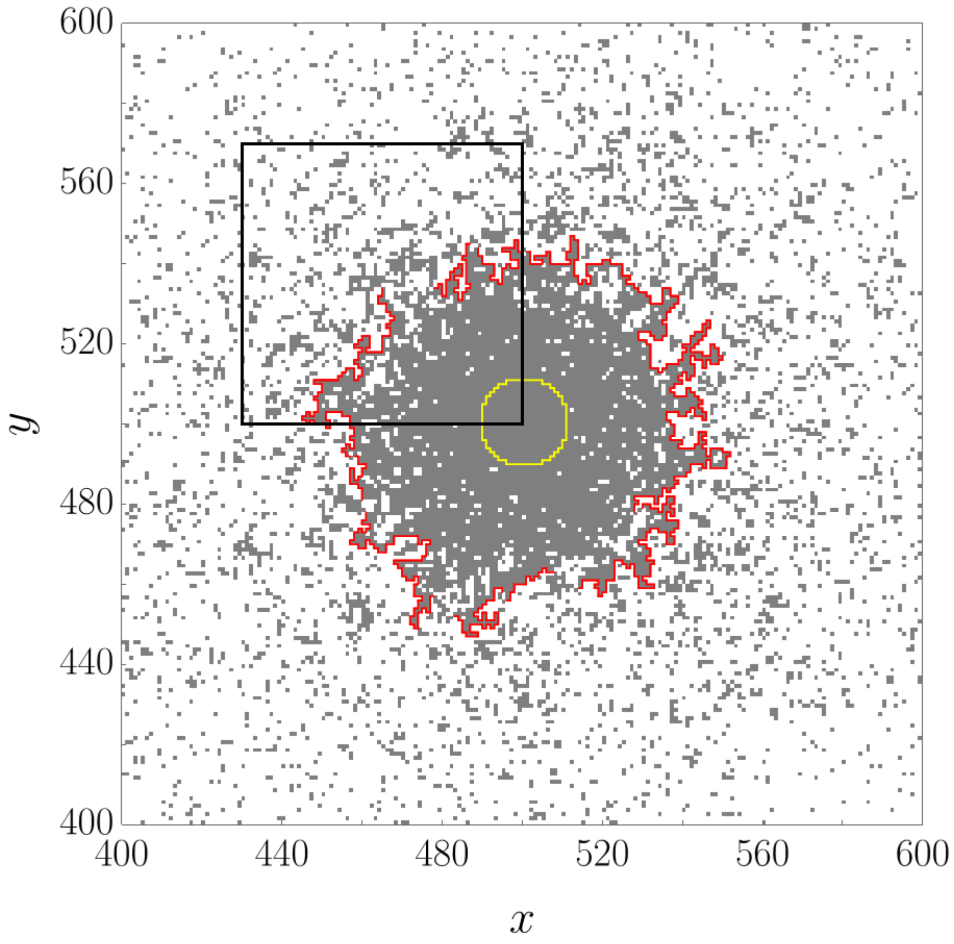

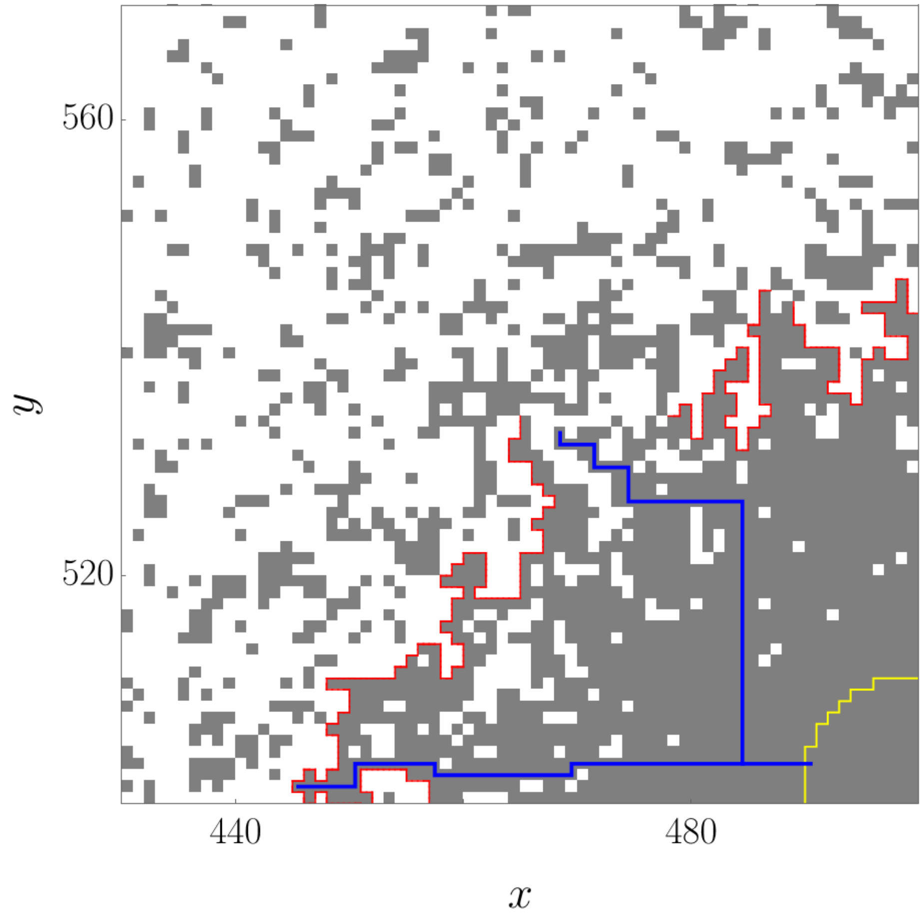

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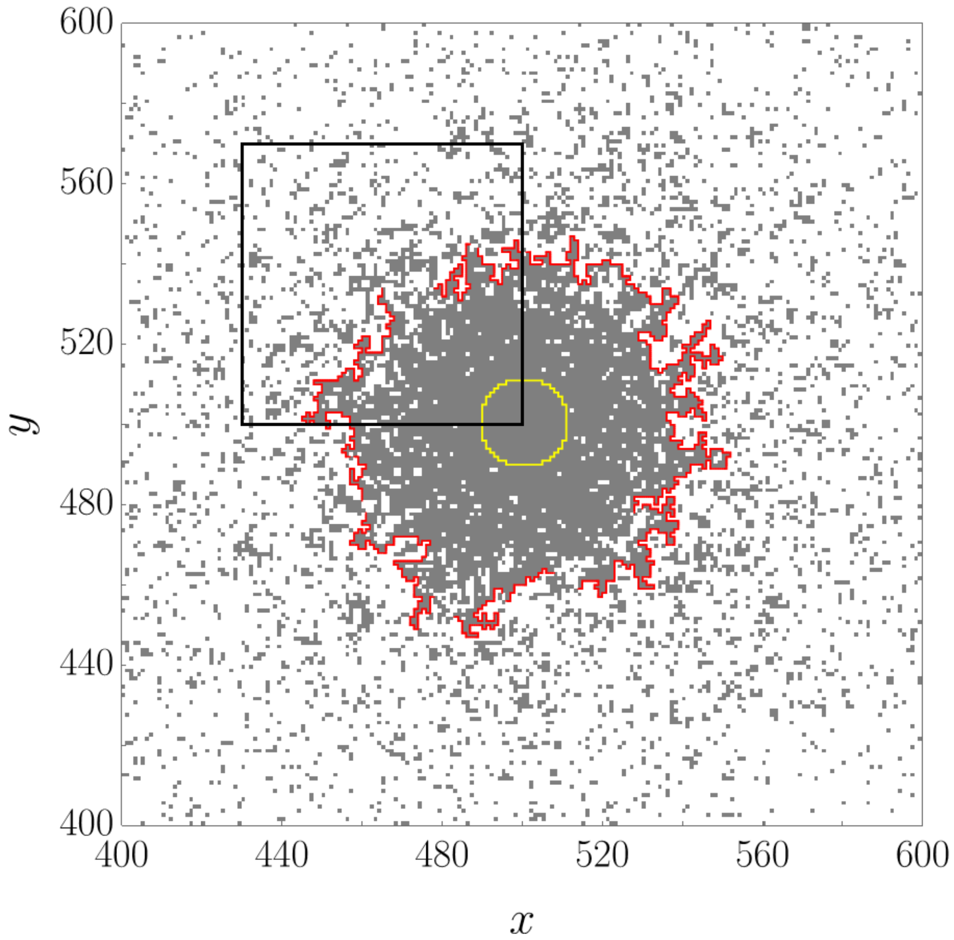

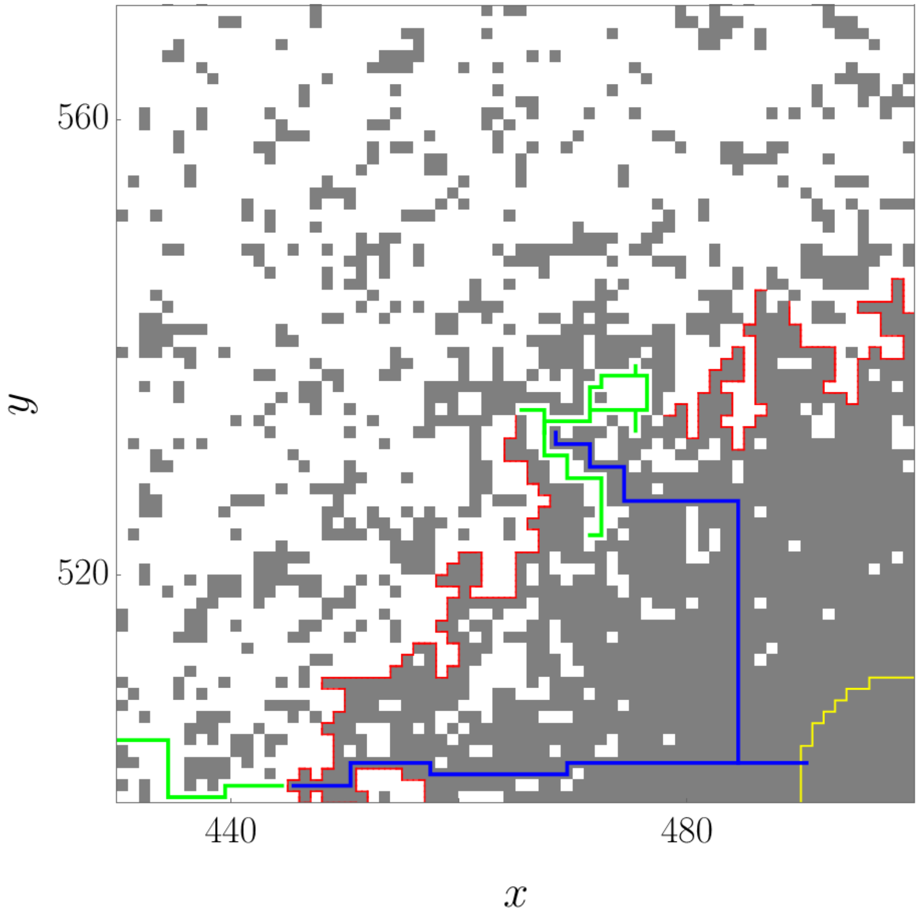

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Monte Carlo simulations: 3 Parameters, only 2 independent

$$A/k_B T \qquad J/k_B T$$

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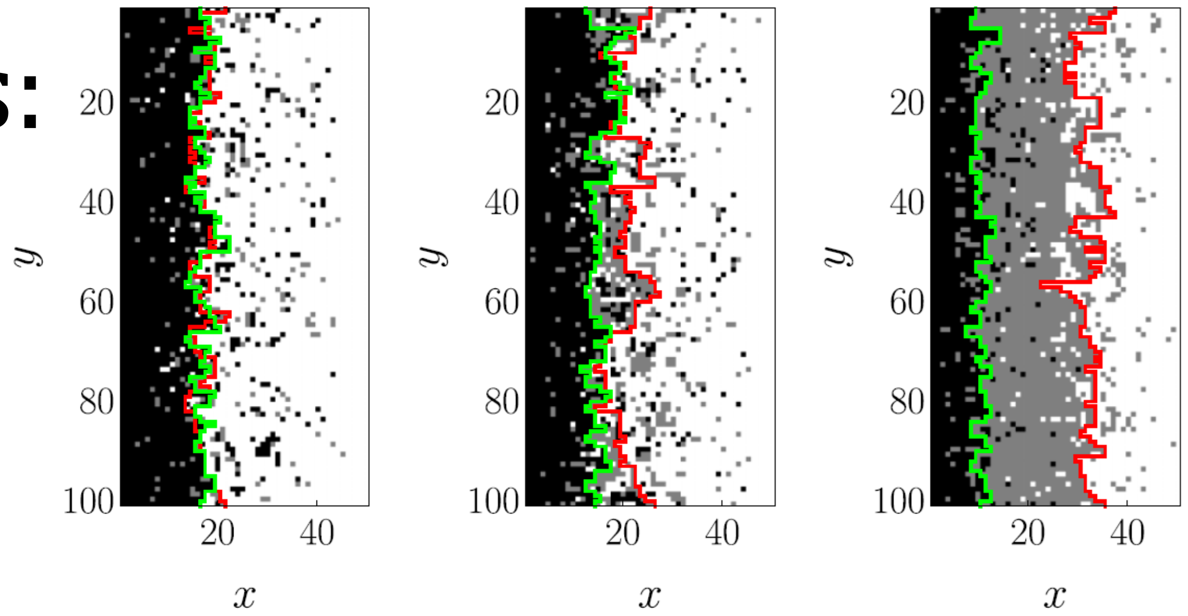

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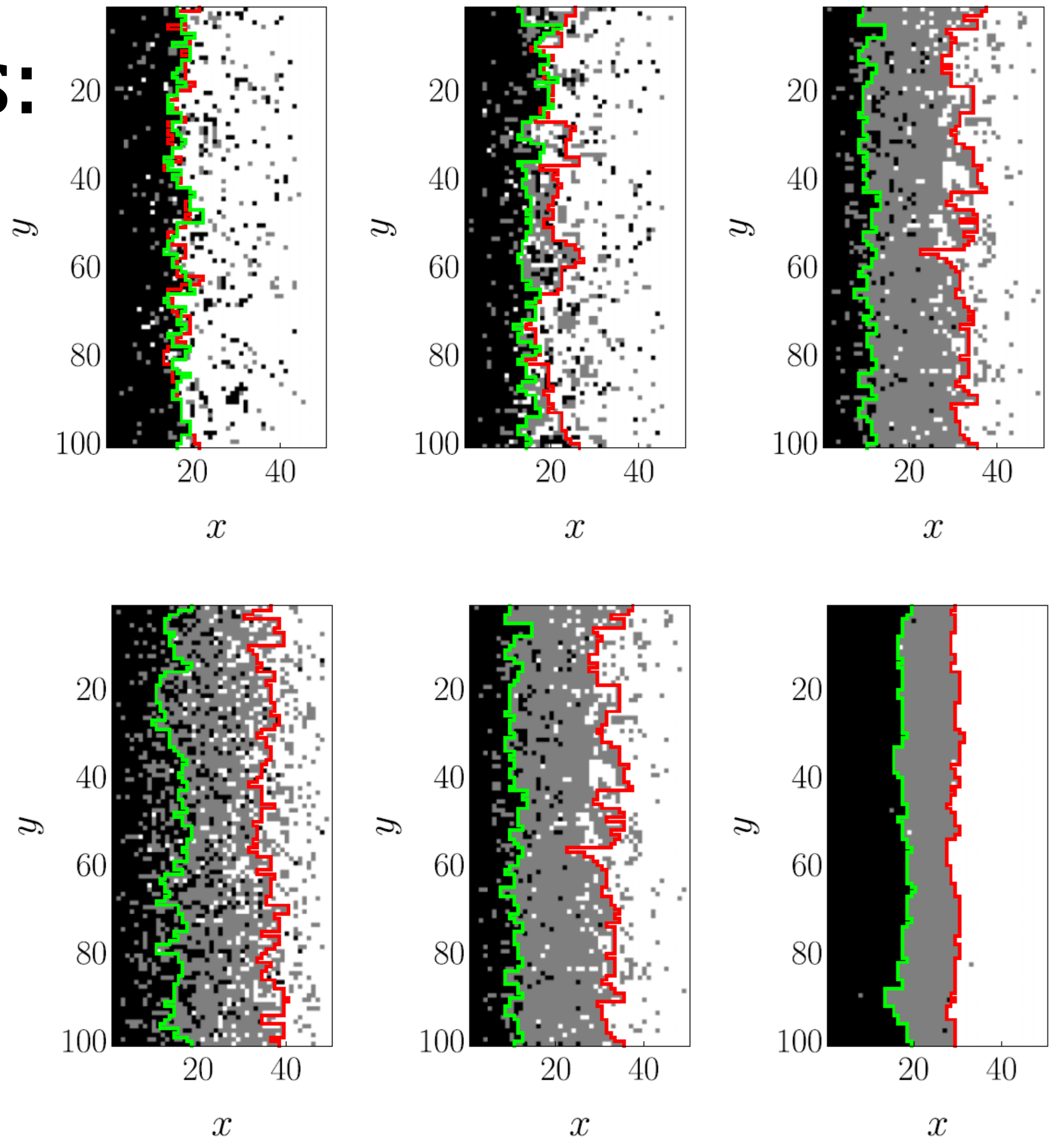
$A/k_B T$ $J/k_B T$



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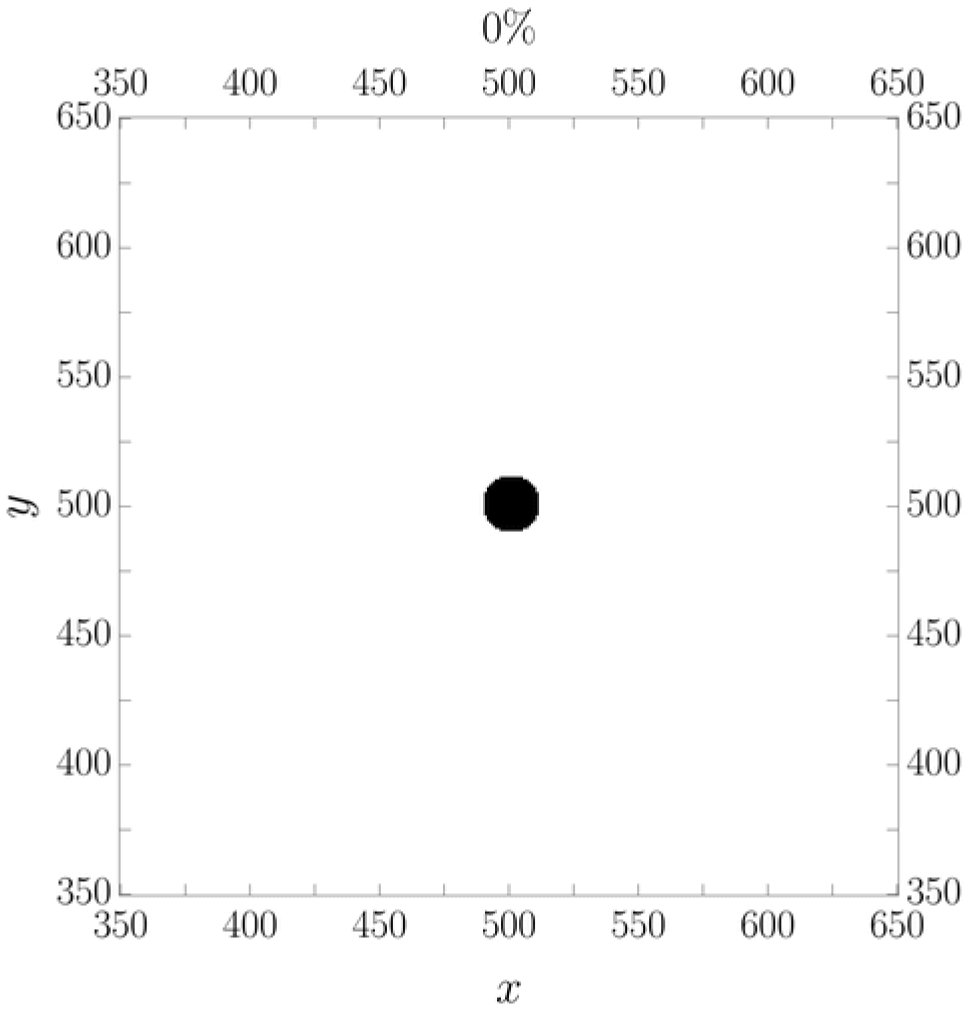
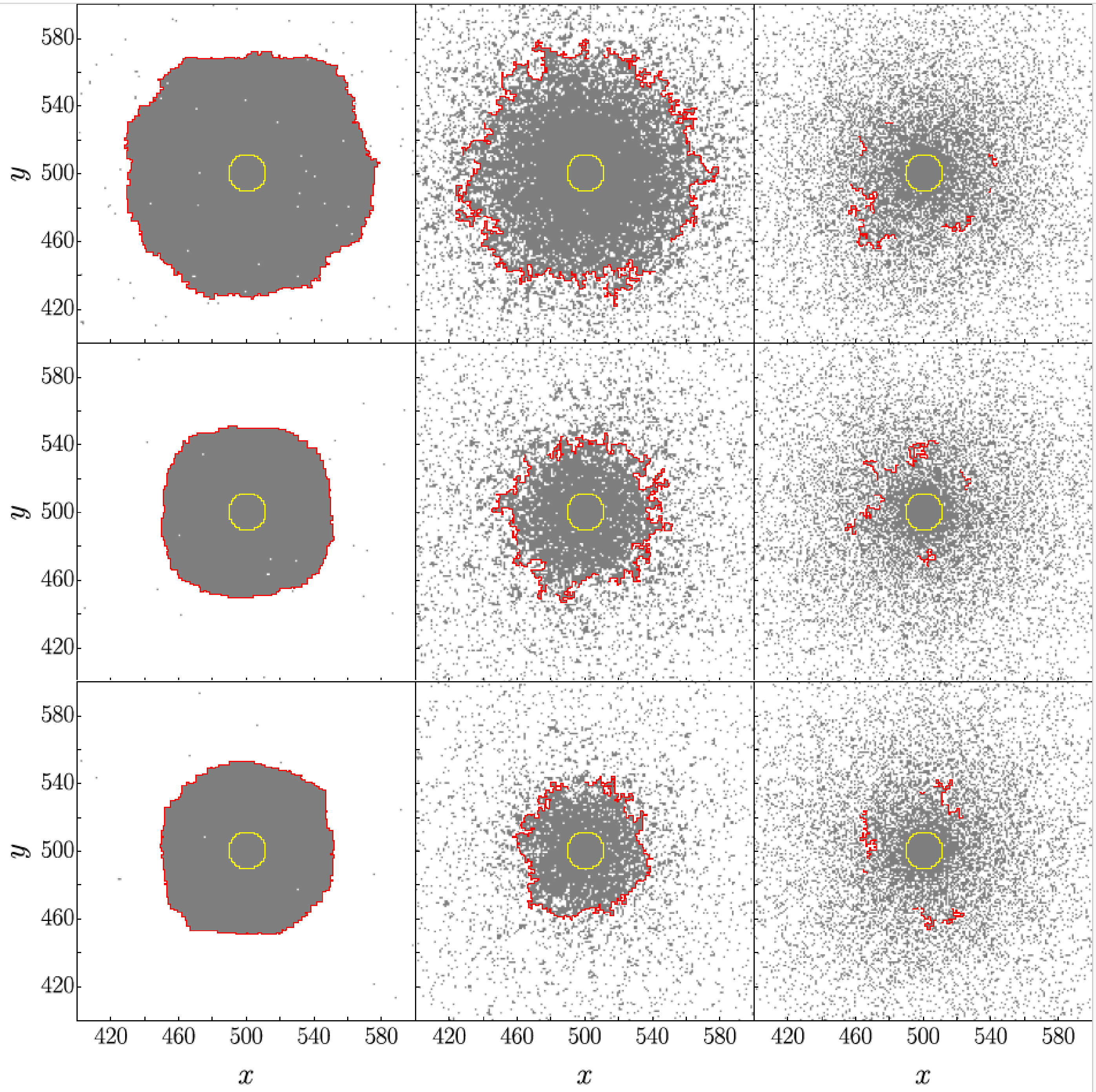
Evolution of the system.





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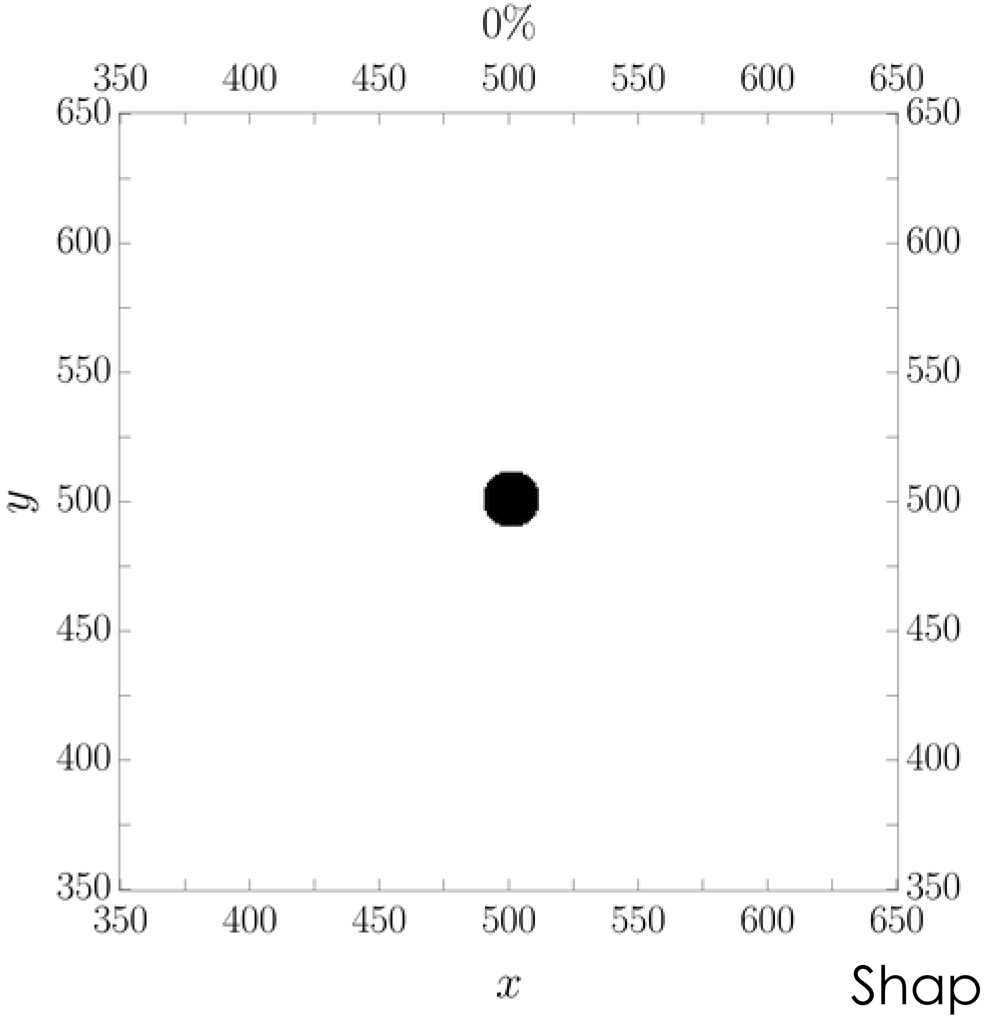

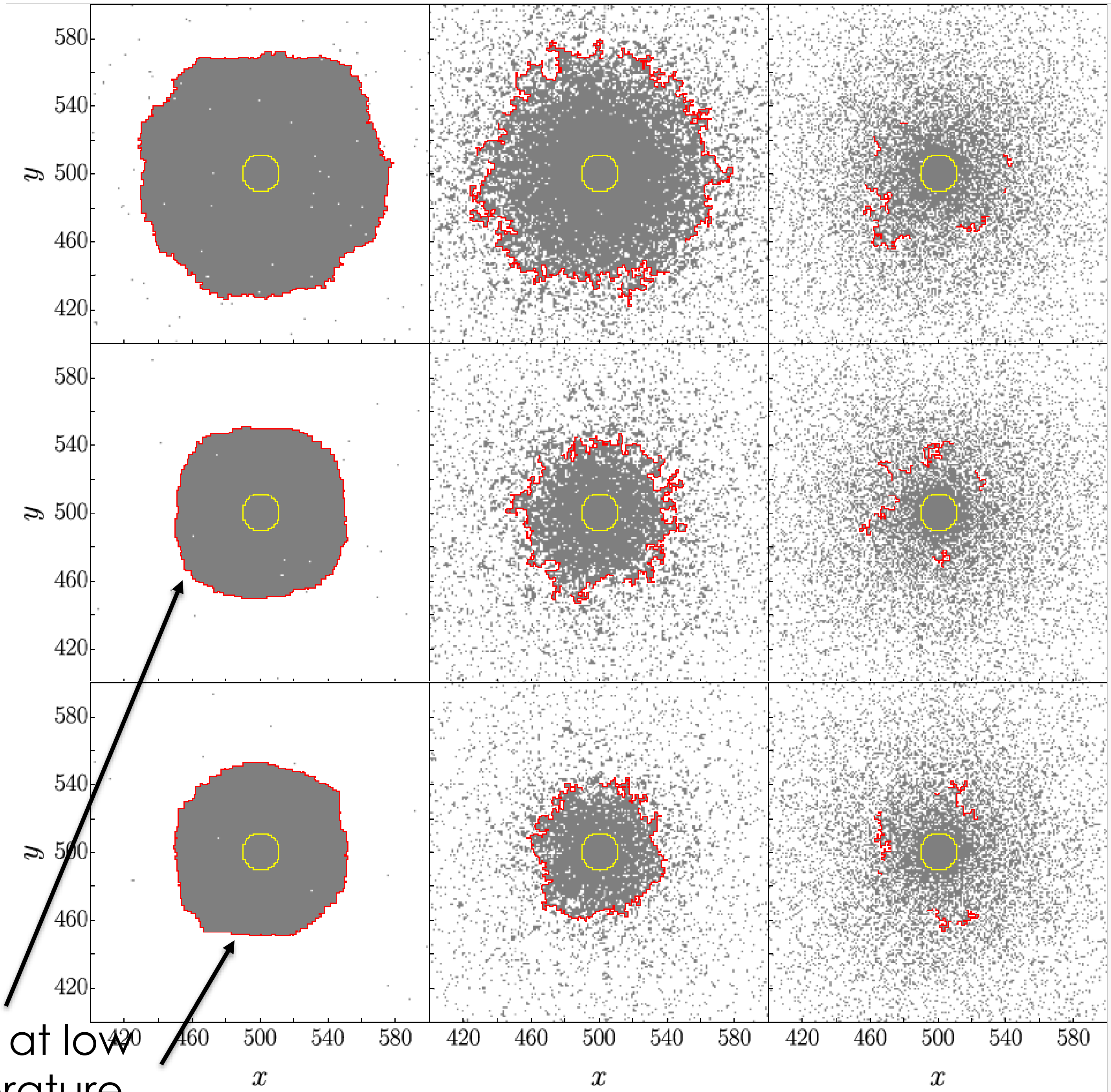

Shape at low temperature

Kinetic roughnening. Universality classes. KPZ growth equation

Family-Vicsek (FV) scaling law type

$$w(L_f, t) = t^\beta f\left(t/L_f^z\right)$$

$$\begin{cases} w \sim t^\beta & \text{for} \quad t \ll L_f^z \\ w_{\text{sat}} \sim L_f^\alpha & \text{for} \quad t \gg L_f^z \end{cases}$$

A.-L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, UK, 1995).

Halpin-Healy and Y.-C. Zhang, *Kinetic roughening phenomena, stochastic growth, directed polymers and all that. Aspects of multidisciplinary statistical mechanics*, Physics Reports **254**, 215(1995)

Kinetic roughnening. Universality classes. KPZ growth equation

Growth exponent

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Growth exponent (β)

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Growth exponent (β)

Dynamic exponent (z)

Roughness exponent (α)

$$\alpha = \beta z$$

Correlations propagate along the front $\xi(t) \sim t^{1/z}$ Saturation $\xi(t) \sim L_f$

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Kardar-Parisi-Zhang (KPZ) equation

$$\frac{\partial h}{\partial t}(\mathbf{x},t) = \nu \nabla^2 h(\mathbf{x},t) + \frac{\lambda}{2}\left(\nabla h\right)^2(\mathbf{x},t) + \eta(\mathbf{x},t) \quad \begin{cases} \alpha = 1/2 \\ \beta = 1/3 \\ z = 3/2 \end{cases}$$

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Front growth

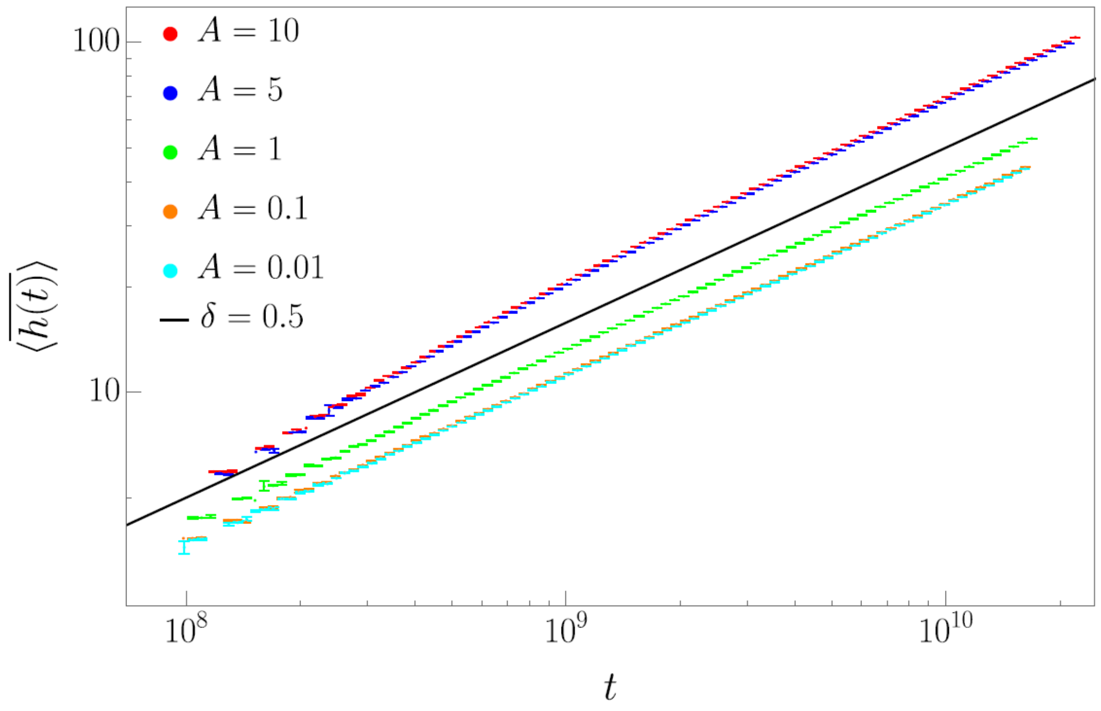
Band geometry:

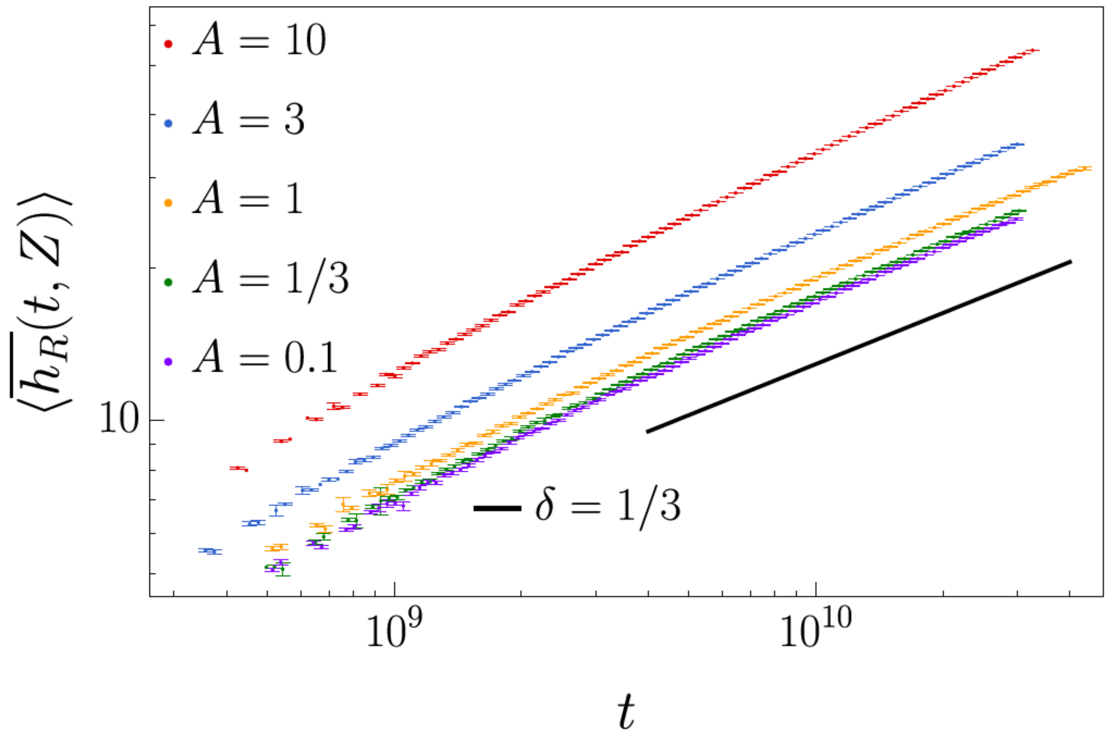
Always 1/2

$$R \sim t^{\delta}$$

Radial geometry:

Sub-diffusive growth





Front growth

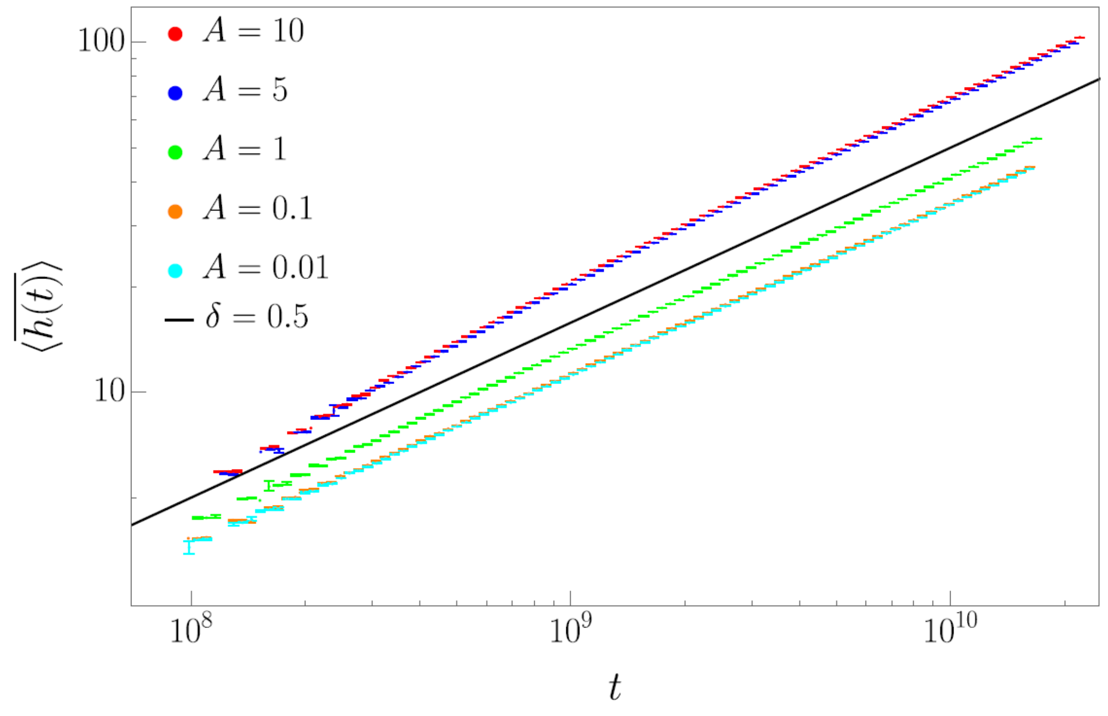
Band geometry:

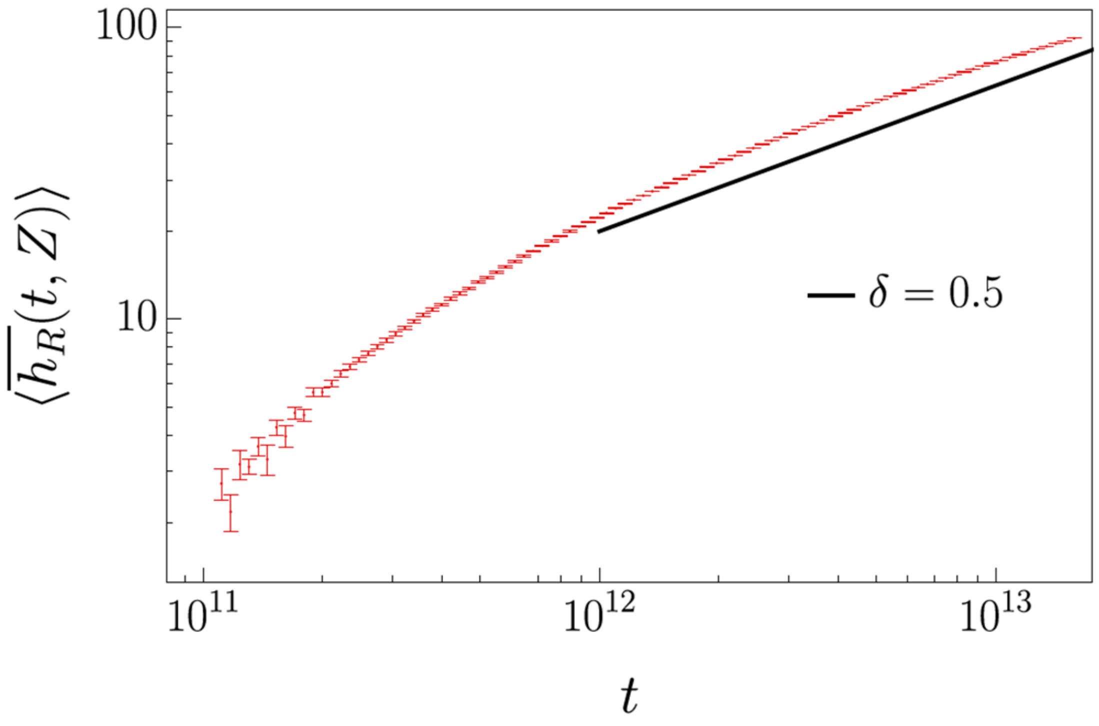
Always 1/2

$$R \sim t^{\delta}$$

Radial geometry:

Close to 1/2 in most realistic case for long times

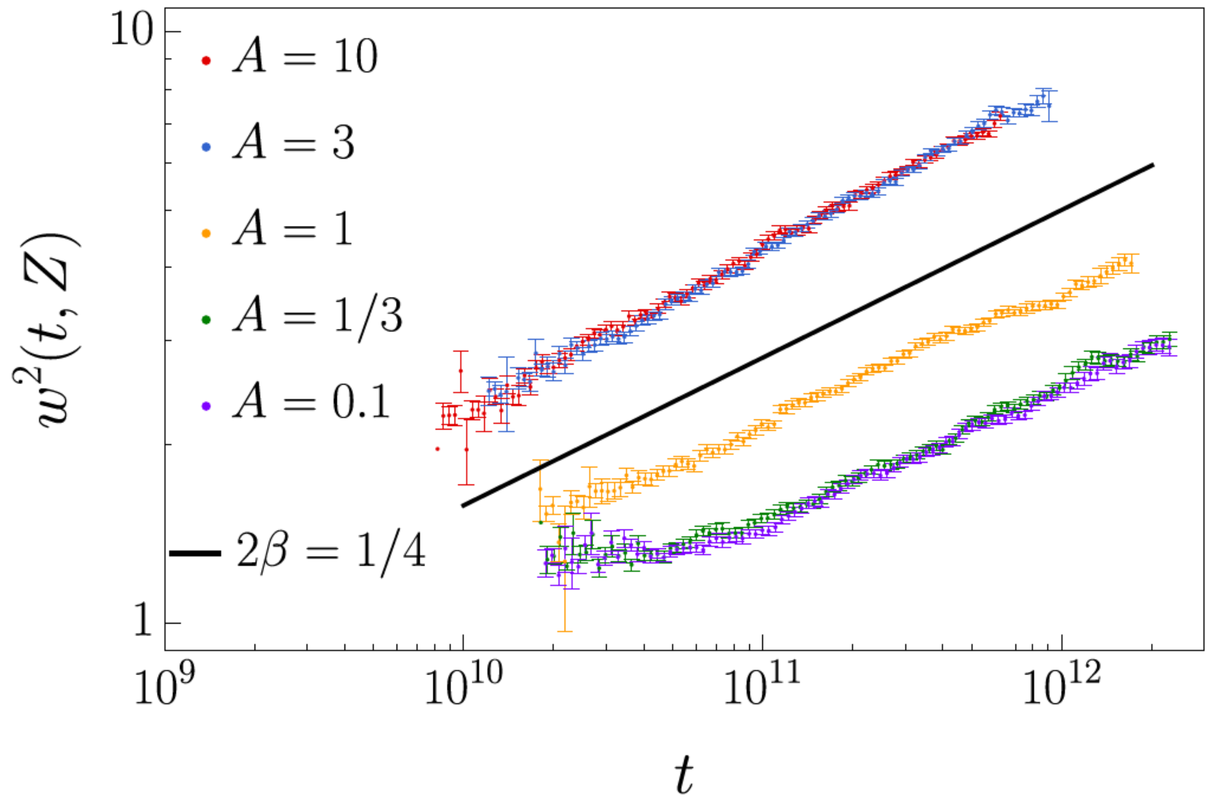




Width of the front: growth exponent

$$w^2(N_Z, t, Z) = \left\langle \overline{[h_i(t,Z) - \bar{h}(t,Z)]^2} \right\rangle$$

$$w \sim t^{\beta}$$ Growth exponent



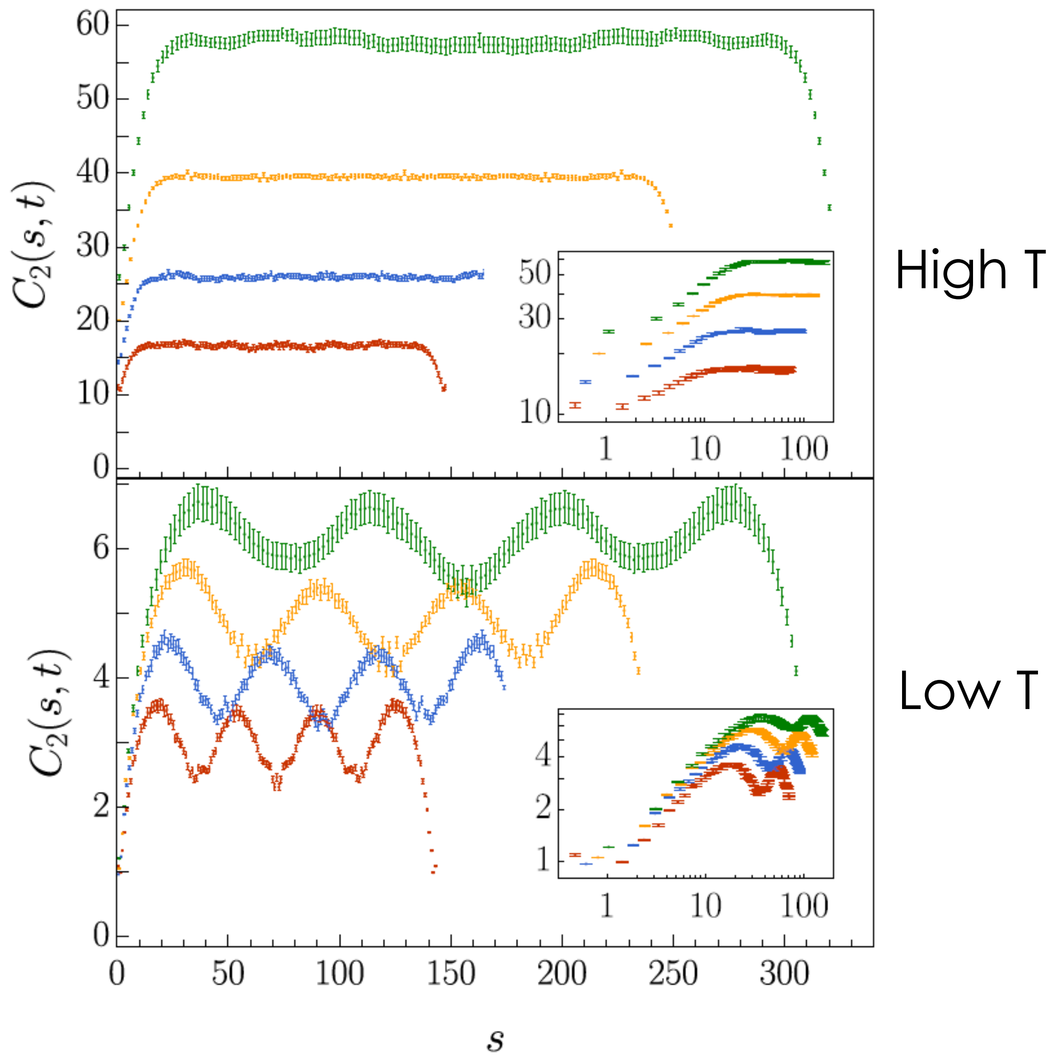
Height difference correlation function

$$C_2(s,t) = \frac{1}{N} \sum_{\bar{h}\Delta\theta_{ij} \in s} \langle [h_i(t) - h_j(t)]^2 \rangle$$

$$\Delta\theta_{ij} = (\theta_i - \theta_j) \bmod 2\pi$$

$$C_2(s,t) \sim s^{2\alpha}$$

$$C_2(\xi_a(t), t) = a\, C_2^{\text{plateau}}(t)$$



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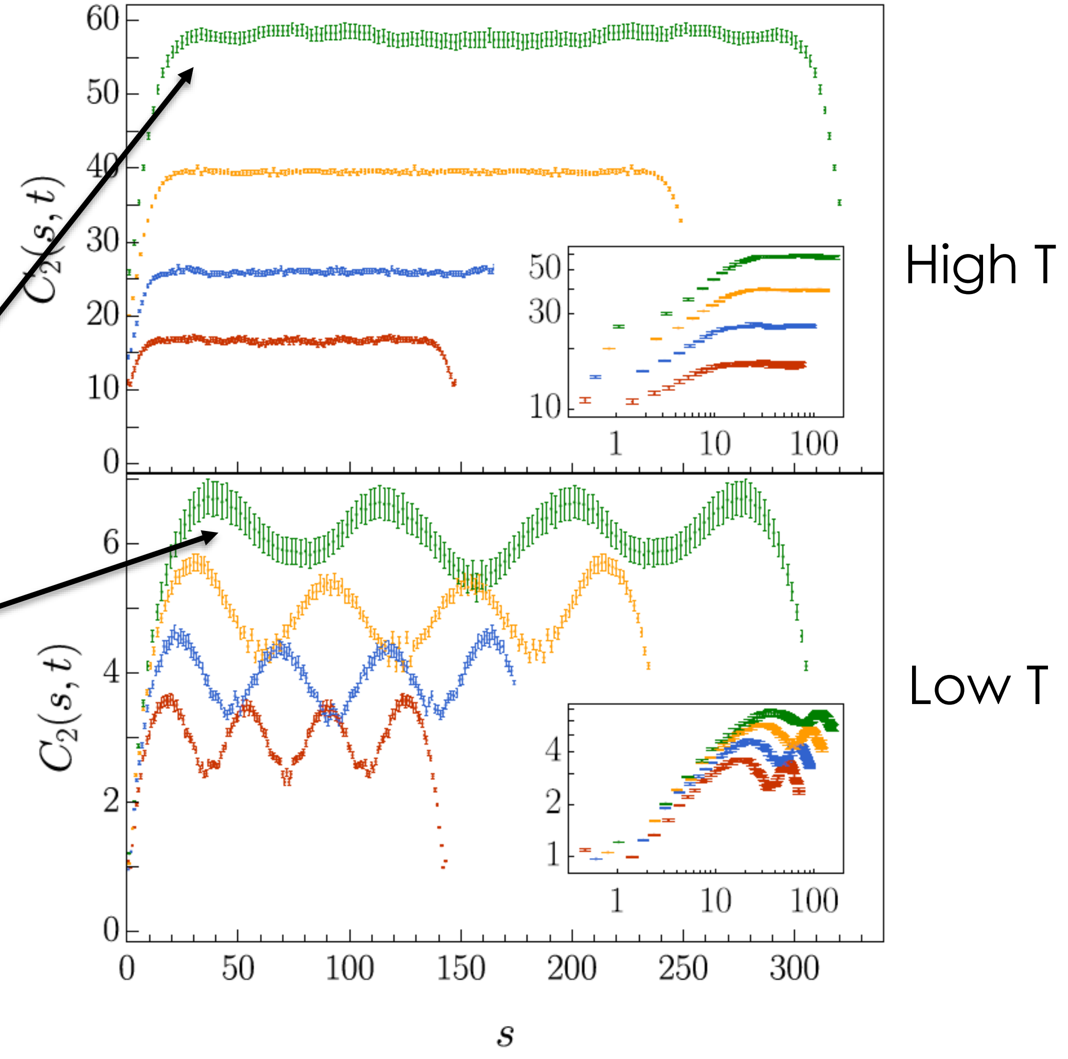

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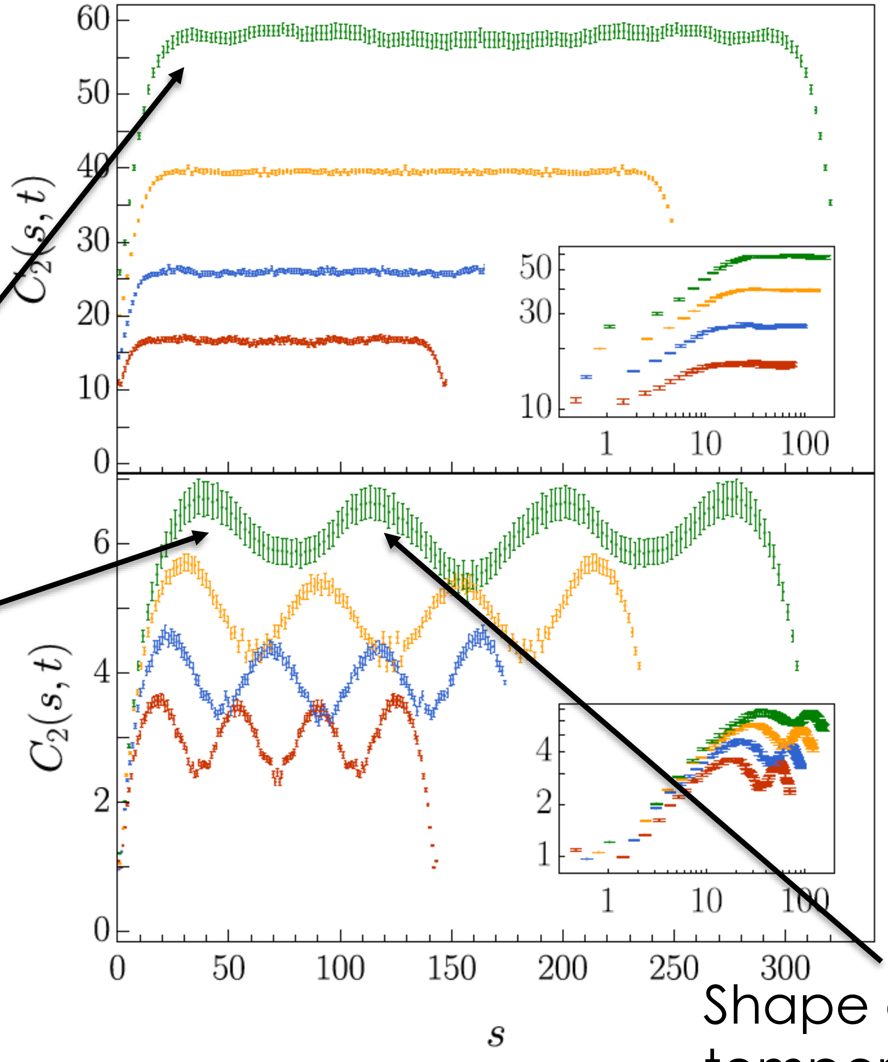
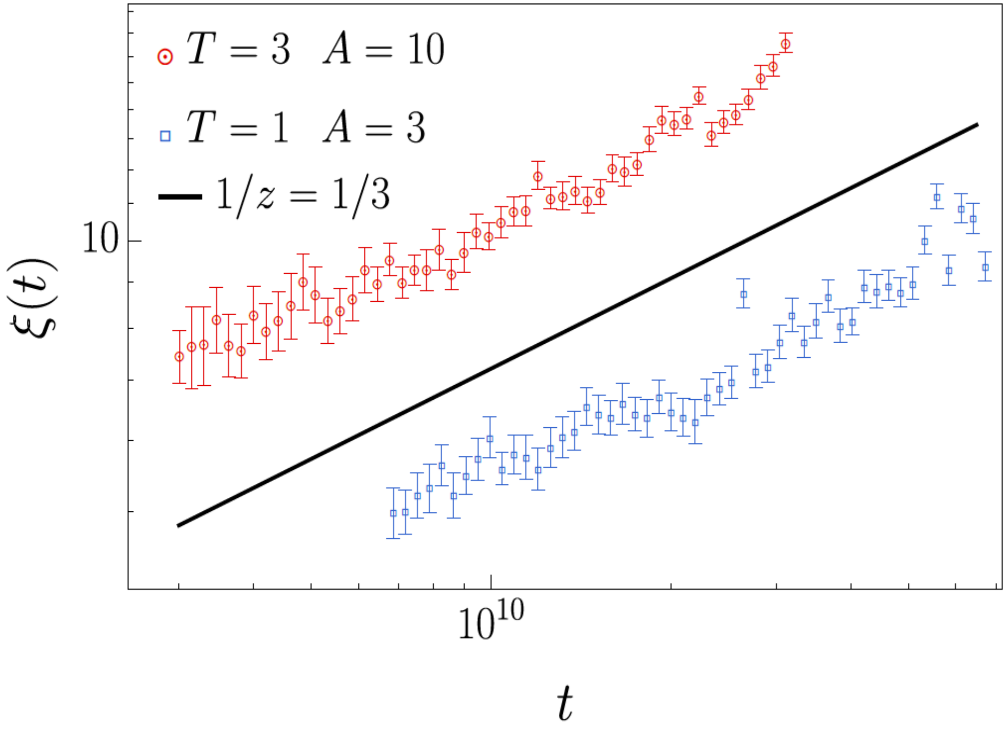


High T

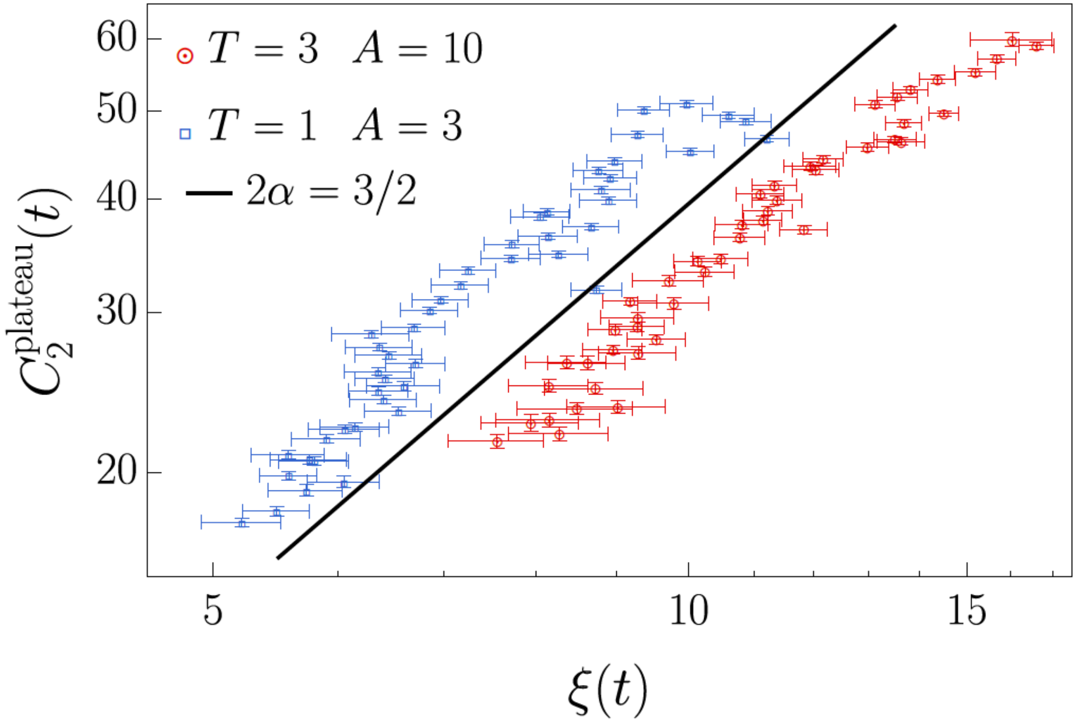
Low T

Shape at low temperature

Dynamic and roughness exponents



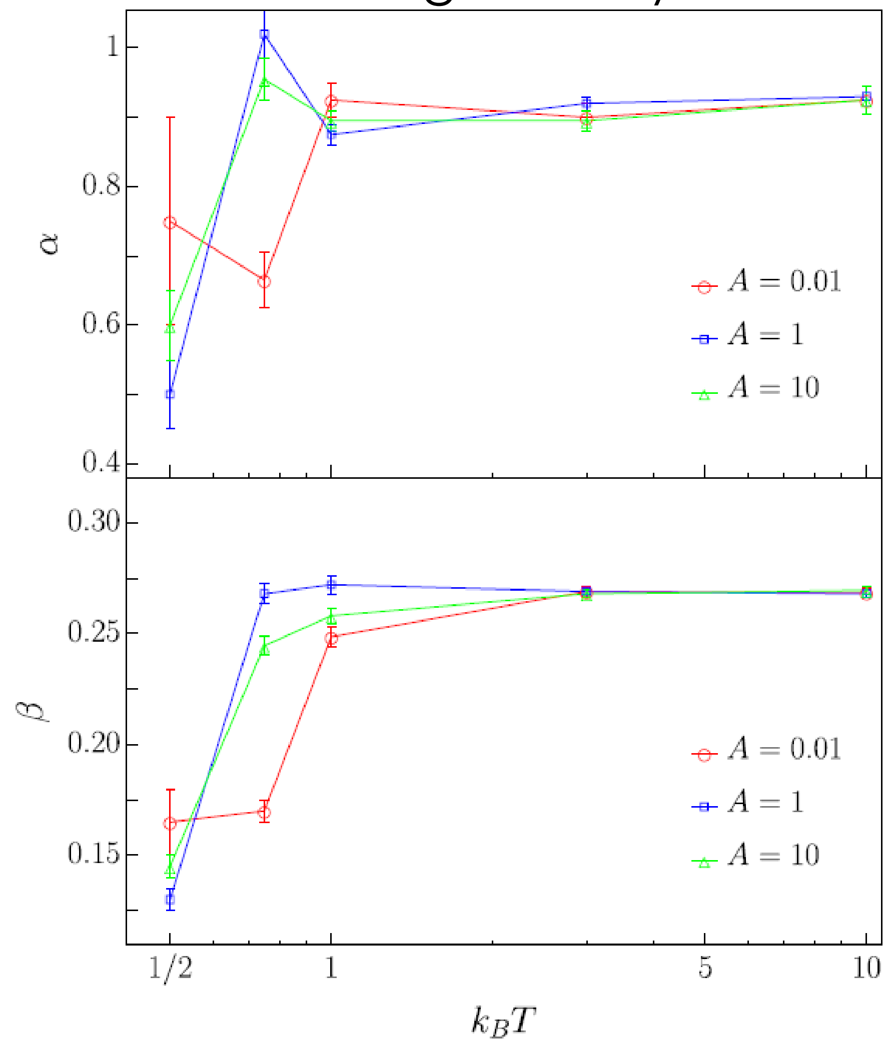
$$\xi(t) \sim t^{1/z}$$



$$C_2^{\text{plateau}}(t) \sim \xi(t)^{2\alpha}$$

Exponent comparasion $\alpha = \beta z$

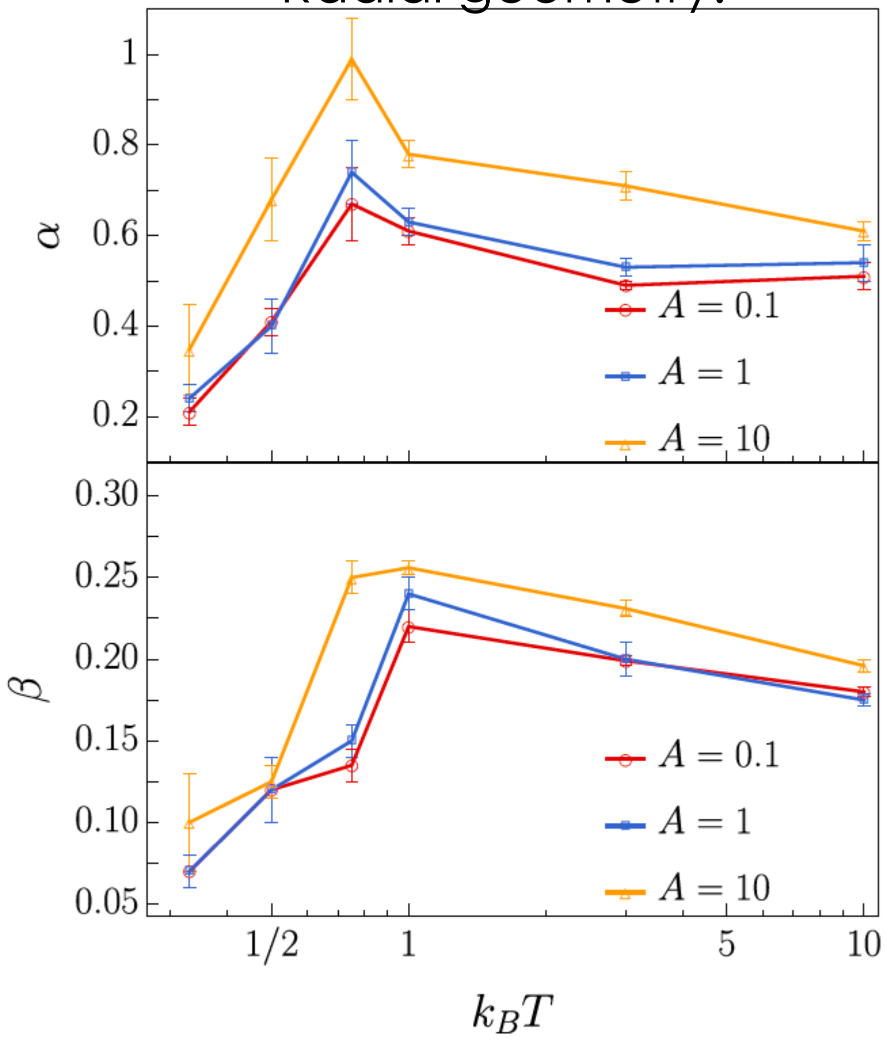
Band geometry:



Transition between a low T and a high T regimes

Not quite the same critical exponents

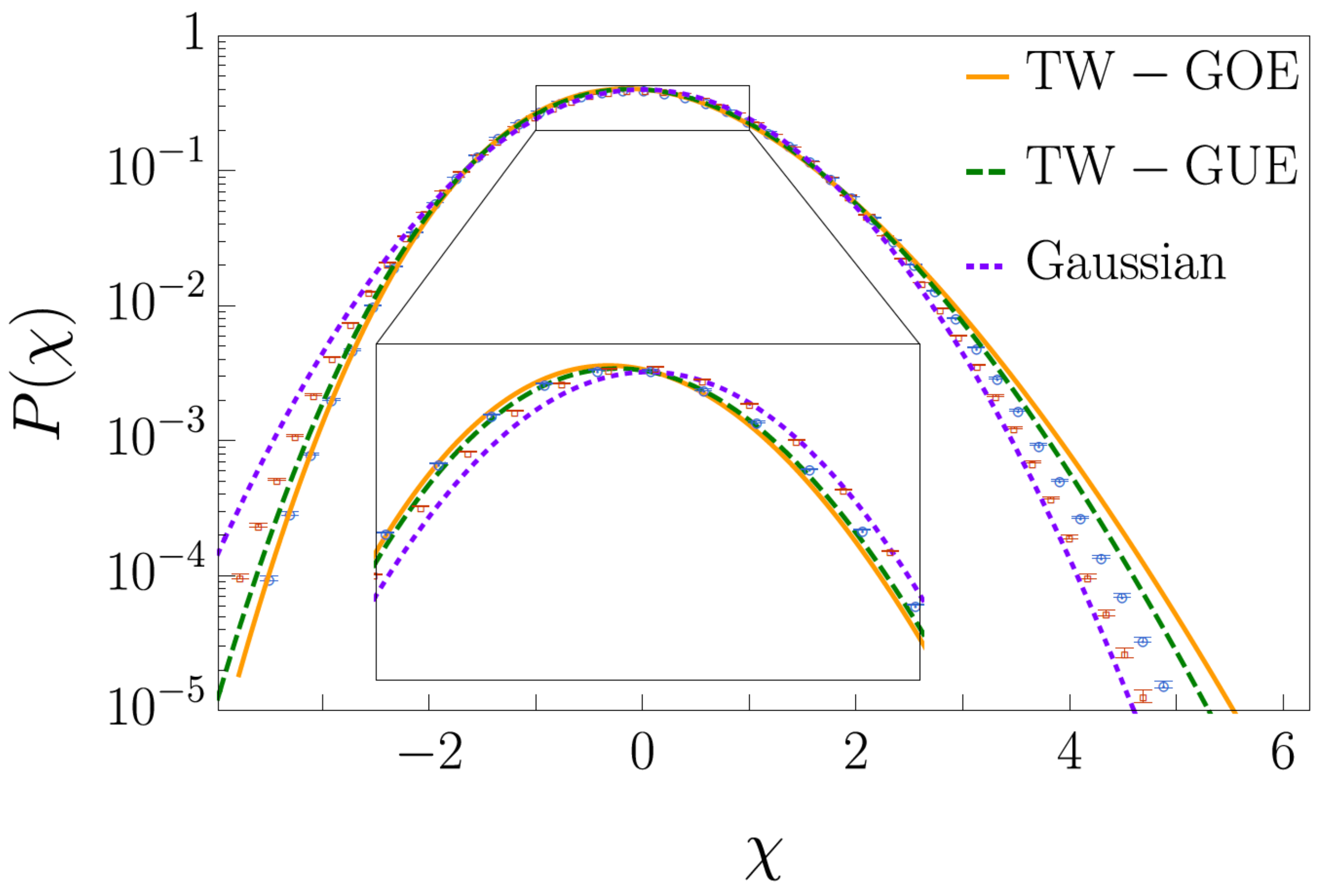
Radial geometry:



Histogram fluctuations

$$\chi_i(t) = \frac{h_i(t) - \bar{h}(t)}{t^{\beta}}$$

KPZ-like fluctuations
without KPZ
exponents!!!



Conclusions

J. M. Marcos, P. Rodríguez-López, J. J. Meléndez, R. Cuerno, and J. J. Ruiz-Lorenzo, Phys. Rev. E **105** (2022) 054801.
J. M. Marcos, J. J. Meléndez, R. Cuerno, and J. J. Ruiz-Lorenzo , arXiv:2411.07923

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Outlook

- Change cluster method?
- Change dynamics (energy barriers?)

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Any questions?

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MC simulations have been performed in the computing facilities of the Instituto de Computación Científica Avanzada de Extremadura (ICCAEx).

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MINISTERIO DE CIENCIA E INNOVACIÓN

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Financiado por la Unión Europea

Funded by the European Union

Supplementary material: Macroscopic spreading laws.

TABLE II. The scaling exponents n, with $R \propto t^n$, corresponding to different balances in droplet spreading in two and three dimensions. Driving forces are gravity (gr.) or surface tension (s.t.), dissipation occurs either at the contact line (c.l.), or in the bulk (vis.). Listed are also experimental papers frequently cited in support of a given scaling law.

Balance	n, theory	Experiment
3D, s.t.-c.l.	1/10	Cazabat and Cohen-Stuart, 1986
2D, s.t.-c.l.	1/7	McHale *et al.*, 1995
3D, gr.-c.l.	1/7	Ehrhard, 1993
2D, gr.-c.l.	1/4	None
3D, gr.-vis.	1/8	Huppert, 1982b
3D, gr.-vis. (pancake)	1/8	Cazabat and Cohen-Stuart, 1986
2D, gr.-vis.	1/5	None

D. Bonn, J. Eggers, J. Indekeu, J. Meunier, and E. Rolley, *Wetting and Spreading,* Rev. Mod. Phys. **81**, 739 (2009)

Supplementary material: kMC simulation details

- Acceptance rate for the μ → v transition. (Metropolis)

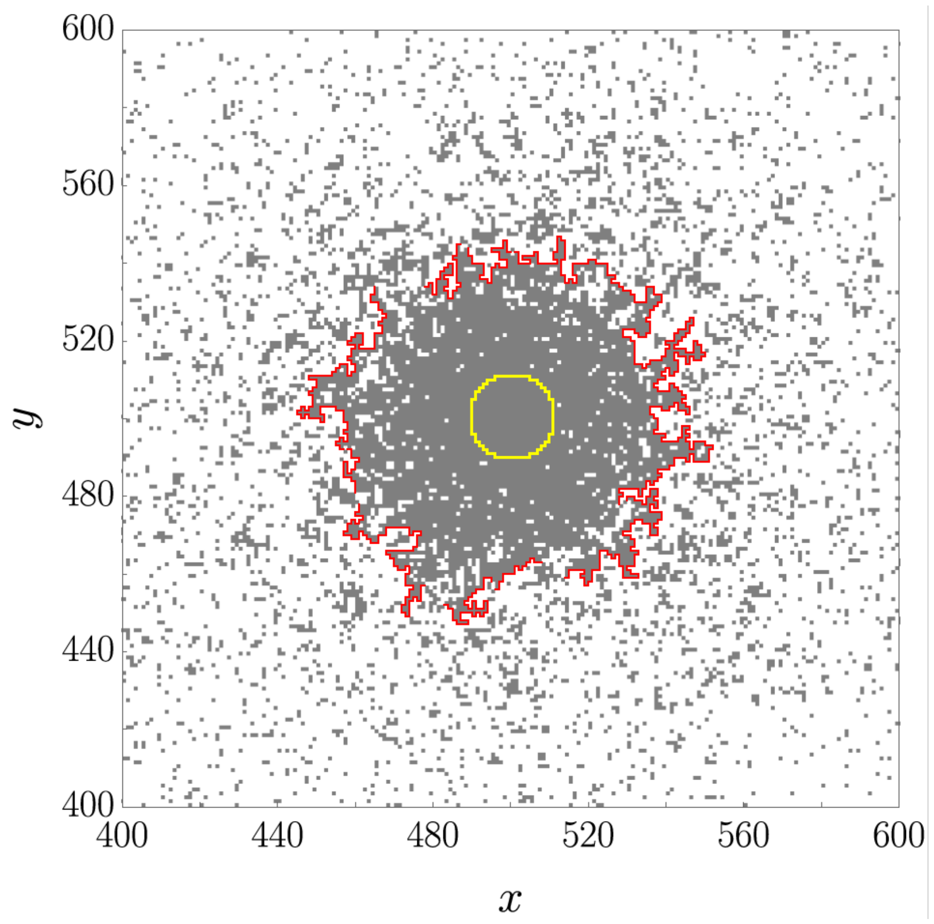
$$A(\mu \to \nu) = \begin{cases} e^{-\beta \Delta E}, & \Delta E > 0, \\ 1, & \Delta E \leqslant 0, \end{cases}$$

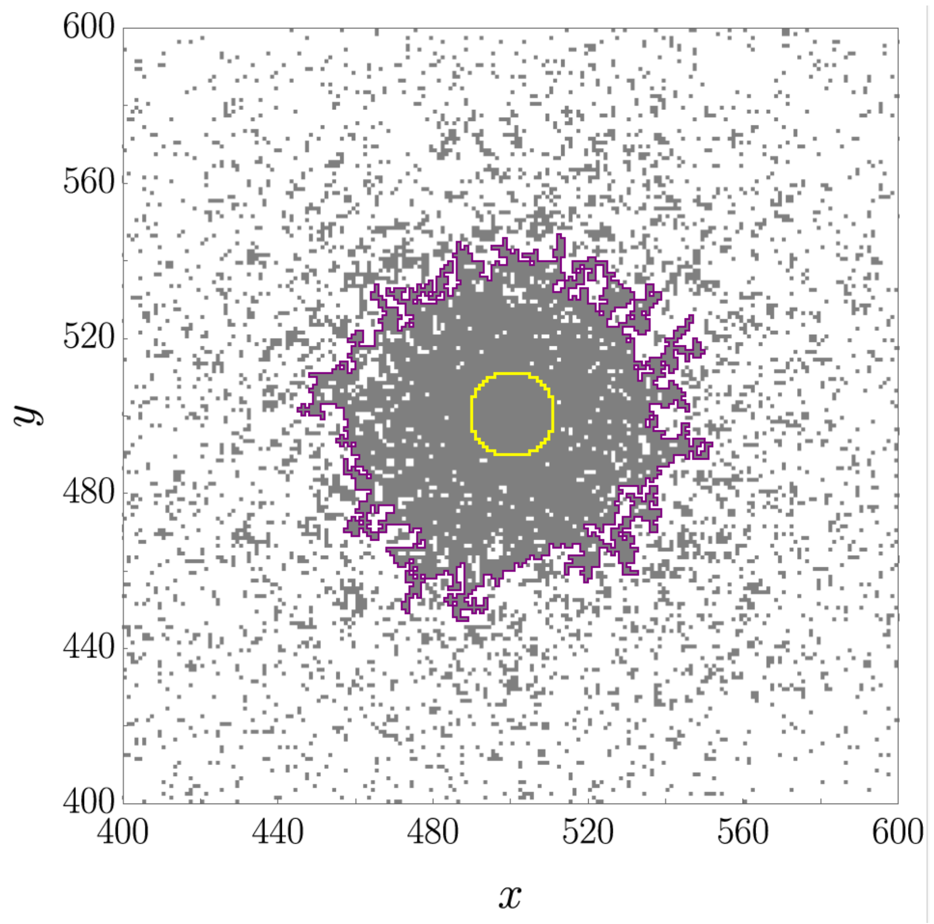
- Continuous time algorithm $\Delta t = \dfrac{-1}{\ln P(\mu \to \mu)}.$

$$P(\mu \to \mu) = \frac{1}{N_P}\left[\text{no. trivial exchanges} + \sum_{\nu \neq \mu}[1 - A(\mu \to \nu)]\right],$$

$$P(\mu \to \mu) = \frac{1}{N_P}\left[N_P - \sum_{\nu \neq \mu} A(\mu \to \nu)\right].$$

Supplementary material: alternative way of measuring the fronts. Closing the gaps





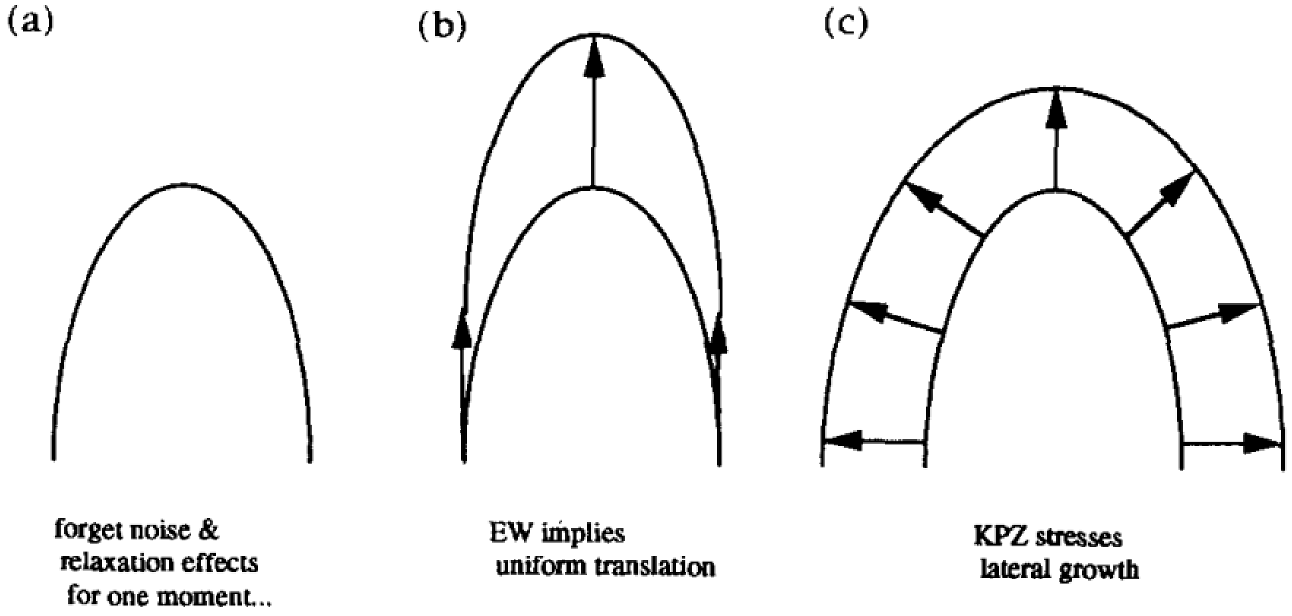
Supplementary material: KPZ and EW

Kardar-Parisi-Zhang (KPZ) $\frac{\partial h}{\partial t}(\mathbf{x},t) = \nu \nabla^2 h(\mathbf{x},t) + \frac{\lambda}{2}(\nabla h)^2(\mathbf{x},t) + \eta(\mathbf{x},t)$

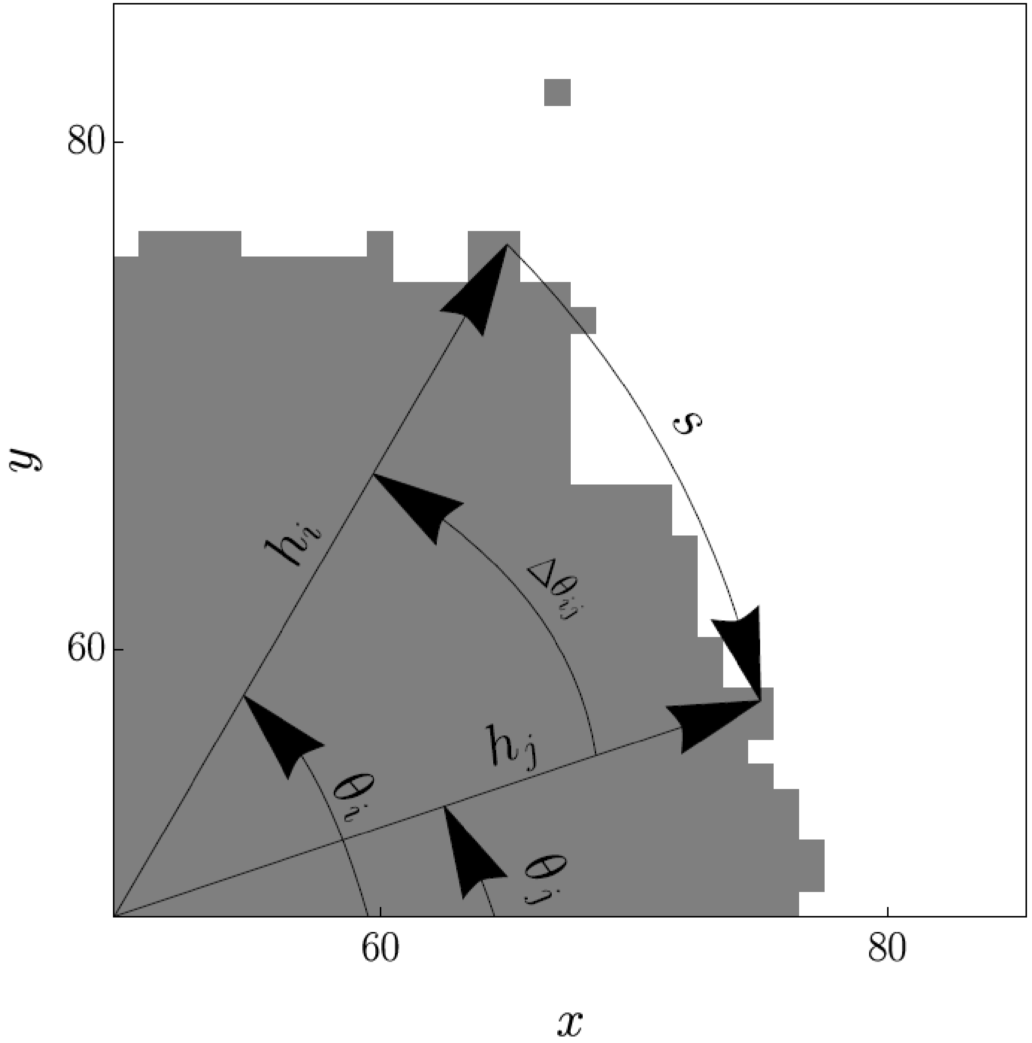
Edwards-Wilkinson (EW) $\lambda = 0$

$$\alpha = \frac{2-d}{2}, \quad \beta = \frac{2-d}{4}, \quad z = 2.$$

$$\alpha = 1/2, \quad \beta = 1/3, \quad z = 3/2.$$



Supplementary material: measuring correlations using the arc-length



Supplementary material: Emergence of anomalous scaling. Details

$$C_2(s,t) \sim s^{2\alpha_{\rm loc}}$$

$$C_2(s,t) = s^{2\alpha} g(s/\xi(t))$$

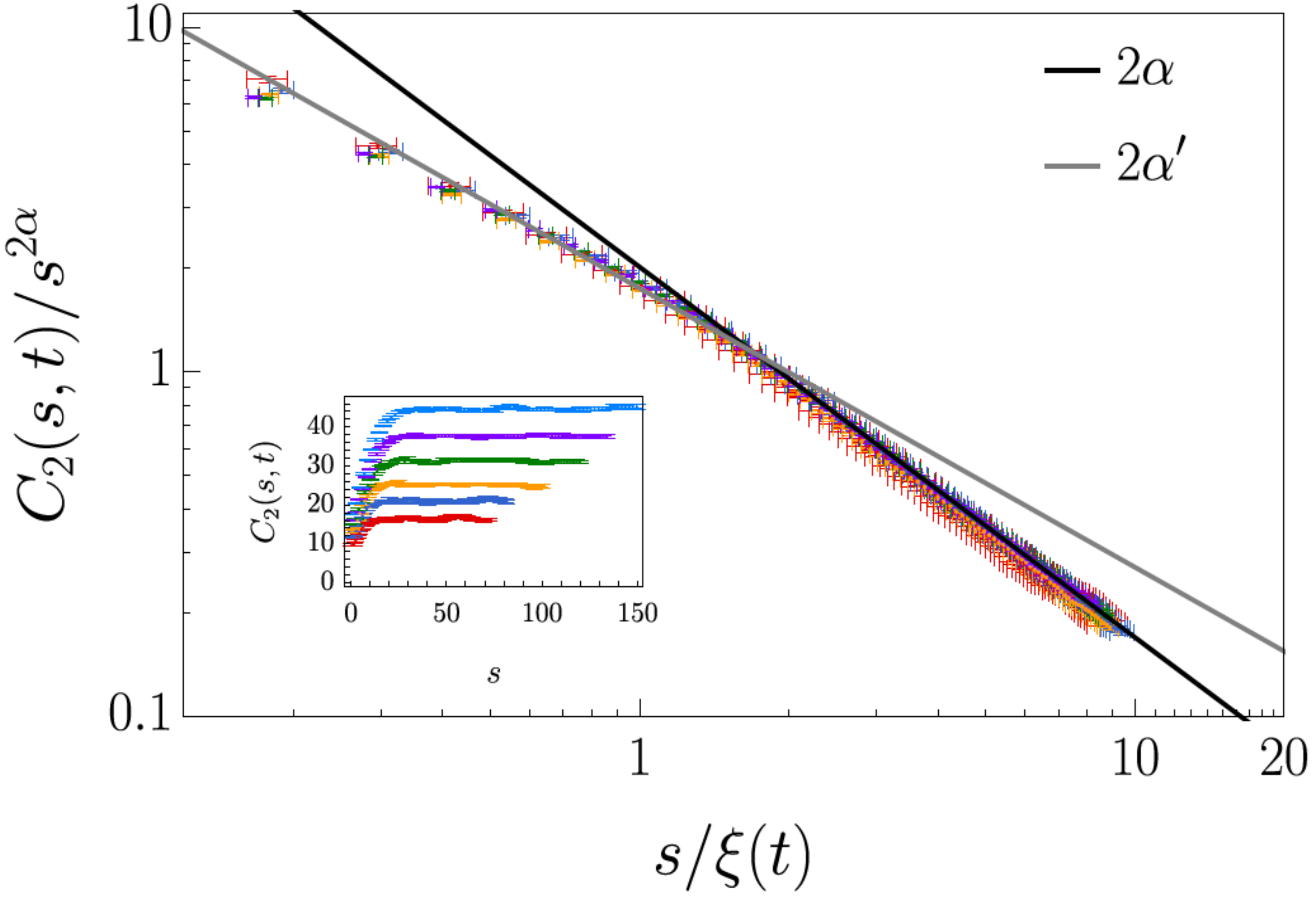
$$g(u) \sim u^{-2(\alpha-\alpha_{\rm loc})} \text{ for } u \ll 1$$

$$g(u) \sim u^{-2\alpha} \text{ for } u \gg 1$$

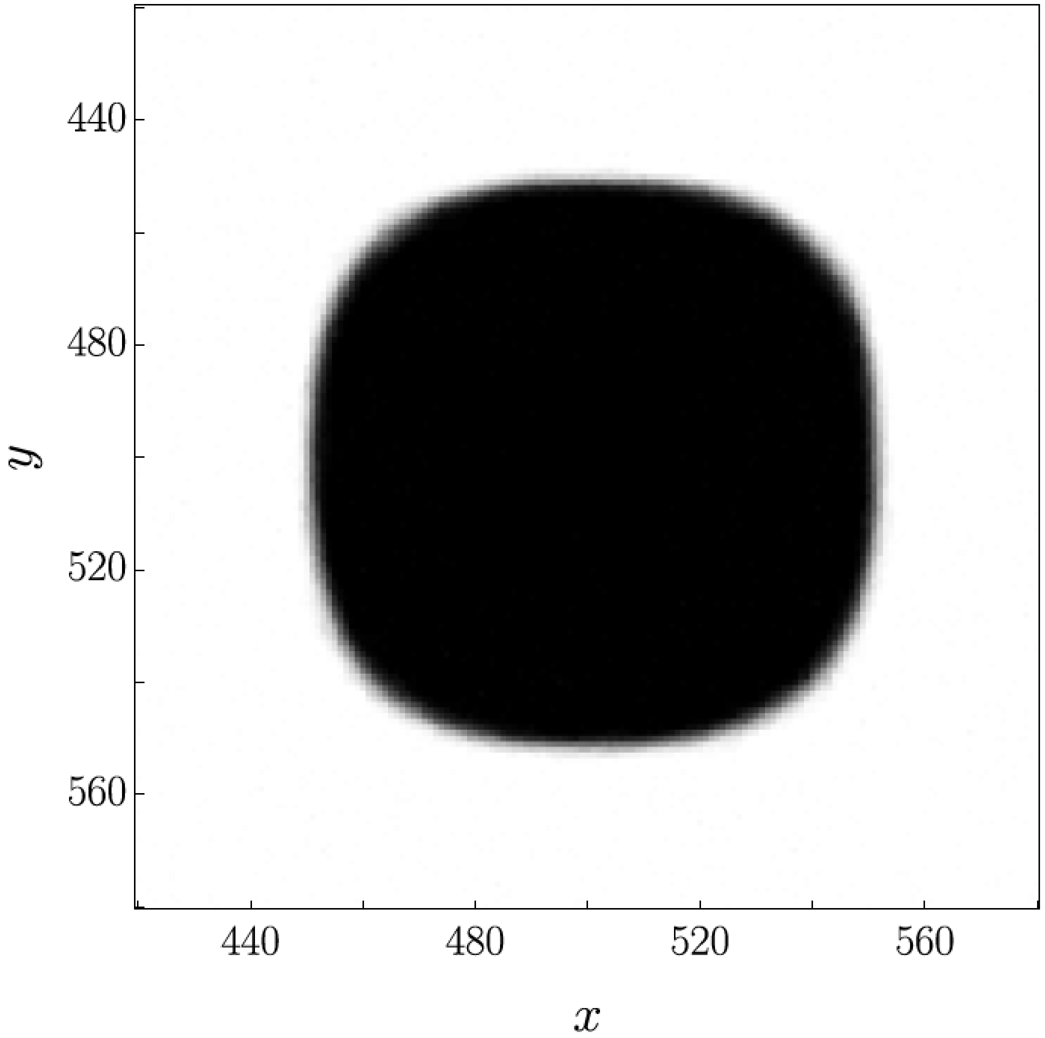
$$\alpha' \equiv \alpha - \alpha_{\rm loc}$$

$\alpha_{\rm loc} = \alpha$ FV scaling type

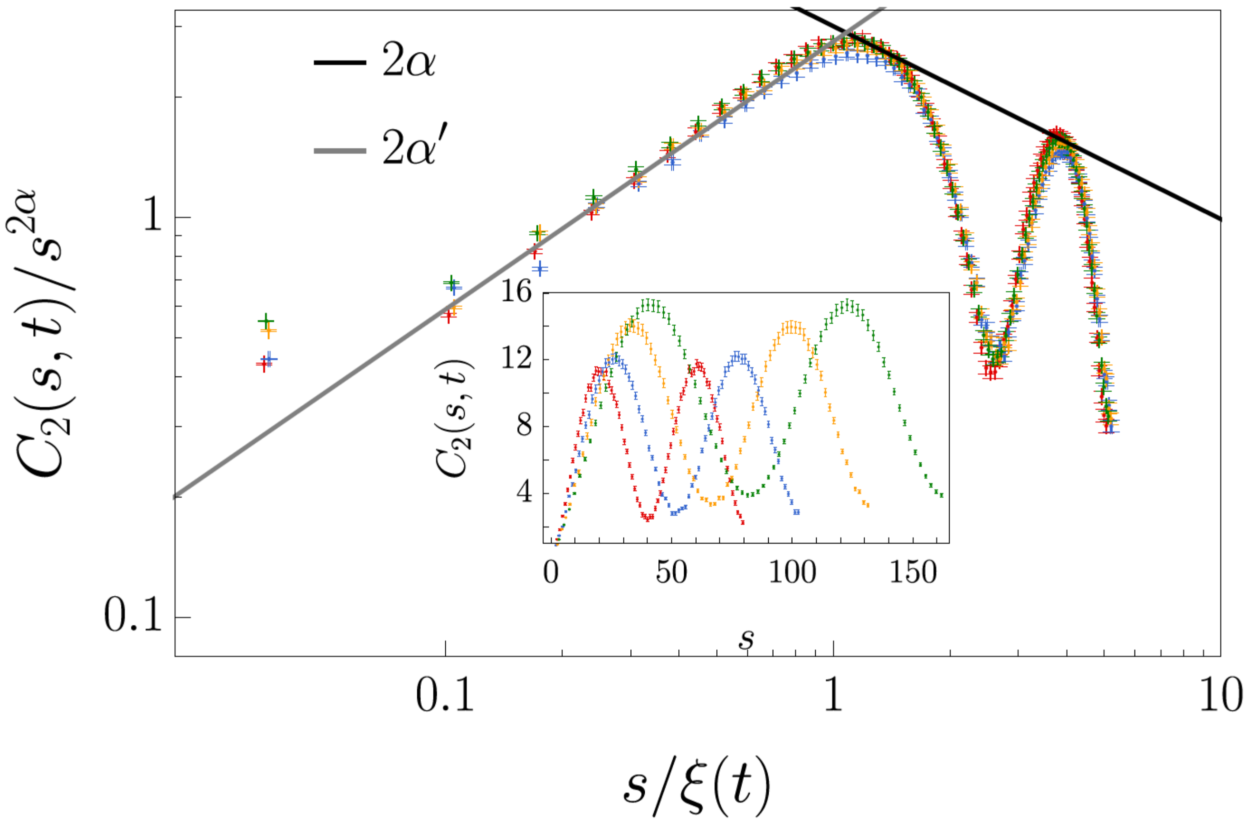
$\alpha_{\rm loc} \neq \alpha$ Anomalous scaling



Supplementary material: average of shapes at low temperature



Supplementary material: Anomolous scaling at low temperature. Data collapse



Supplementary material: fluctuations histograms at low temperature

$$\chi_{\Omega i}(t) = \frac{h_i(t) - \bar{h}_\Omega(t, Z)}{t^{\beta_\Omega}}$$

$$\bar{h}_\Omega(t, Z) = \frac{1}{N_Z(\Omega)} \sum_{i \in \Omega} h_i(t, Z)$$

$$w_\Omega^2(L_f, t, Z) = \left\langle \overline{[h_i(t, Z) - \langle \bar{h}_\Omega(t, Z) \rangle]^2} \right\rangle$$