Kinetic Theories in Multiscale Modeling of Polycrystals

Maria Emelianenko

Dept. of Mathematical Sciences
George Mason University

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Grand challenges in understanding polycrystalline microstructure

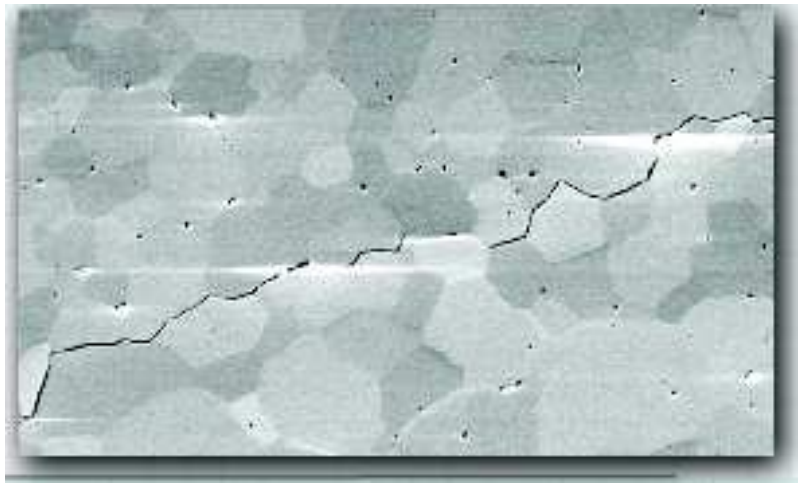
A central problem in materials science is the understanding and control of microstructure: ensemble of grains that comprise polycrystalline materials. Performance is influenced by the types of grain boundaries in the material and the way that they are connected.

Structure sensitive properties:

- Superconducting Critical Current Density
- Electromigration Damage Resistance
- Stress Corrosion Cracking
- Electrical activity
- Creep Behavior

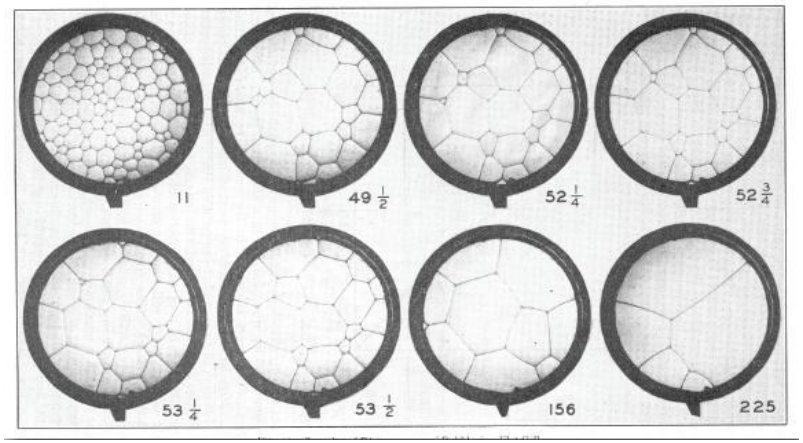
"Insensitive":

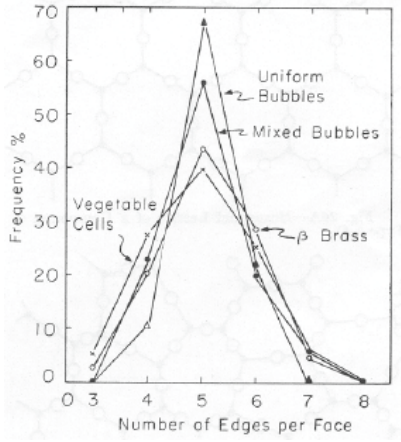
- Average elastic energy density



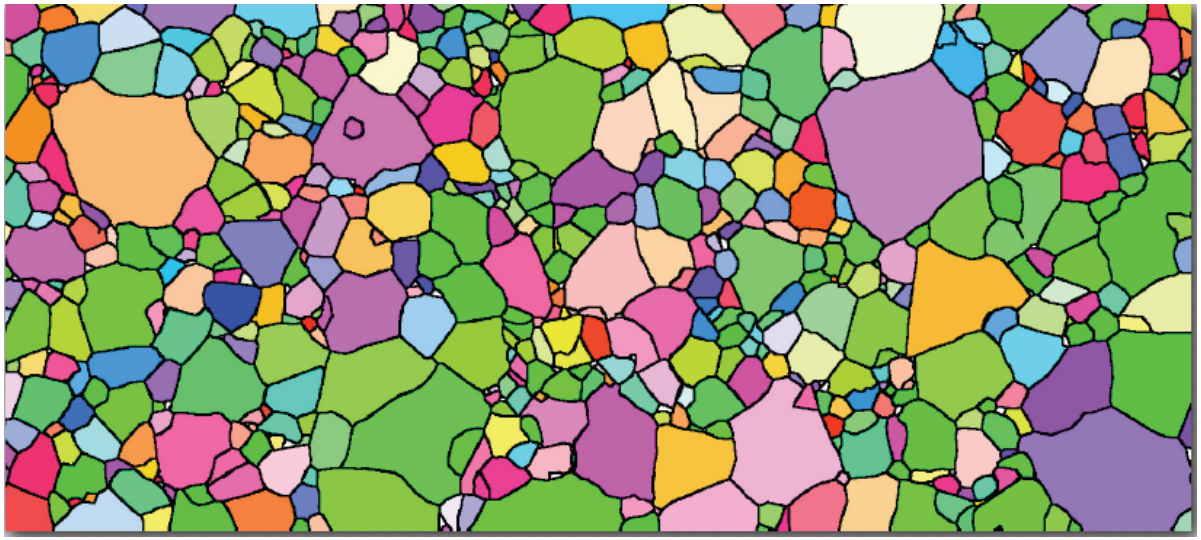
Fracture follows grain boundaries.

Questions: are microstructures all like soap froth? C.S. Smith (1951)



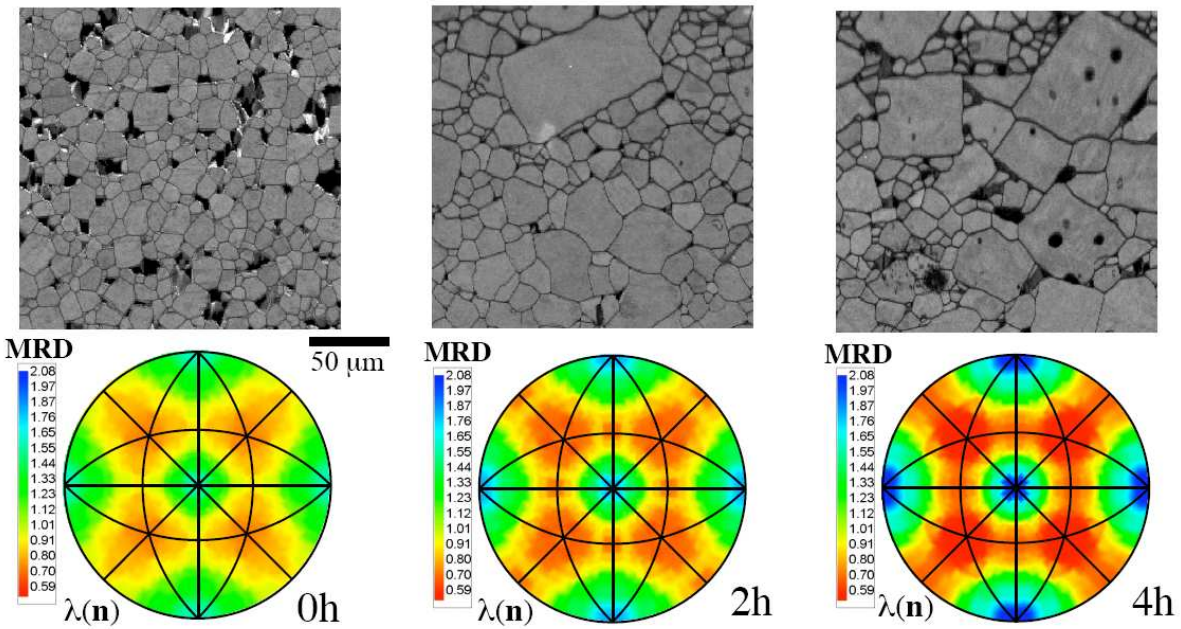


MgO looks very much unlike, but appearances can be deceiving.



Motivation

Experiment: evolution of grain boundary character distribution (GBCD)
GBCD = relative areas of grain boundaries sorted by misorientation angles and normal



Gorzkowski et al. Zeitschrift fur Metallkunde, 96 (2005) 207.

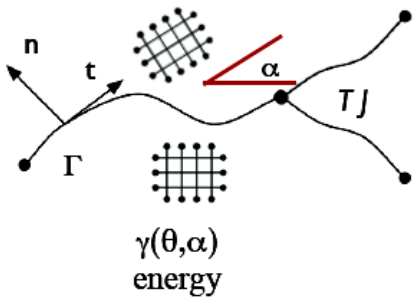
$$Pb(Mg_{1/3}Nb_{2/3})O_3 - 35\ PbTiO_3$$

Recent discovery

Grain boundary character (GBCD) is a scale invariant steady state characteristic of a material.

Foundations

- Curvature driven growth of a network of grain boundaries



E total energy
γ gb energy
n normal
t tangent
θ angle of normal
α misorientation
κ curvature of Γ

$$E = \int_\Gamma \gamma(\theta,\alpha)|t|ds \qquad \text{total energy}$$

$$T = \gamma_\theta n + \gamma t \qquad \text{line stress}$$

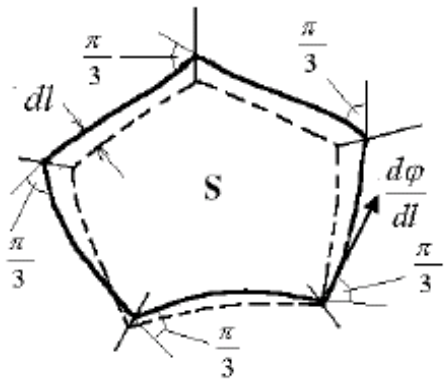
Mullins equation:

$$v_n = \mu(\frac{\partial^2\gamma}{\partial\theta^2} + \gamma)\kappa \text{ on } \Gamma^{(i)}$$

Herring condition for triple junctions:

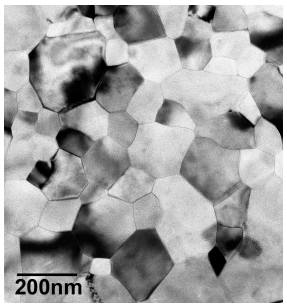
$$\sum_{TJ}(\frac{\partial\gamma}{\partial\theta}n + \gamma t) = 0, \text{ at TJ's}$$

- Interfacial energy depends on the misorientation angle and normal: $\gamma(\theta,\alpha)$
- Mullins-von Neumann law: $\frac{dA_n}{dt} = c(n-6)$ extended to $\mathbb{R}^n$ MacPherson & Srolovitz, Nature 2007
- Other simulation techniques: Potts model, Monte Carlo, mean field theory etc

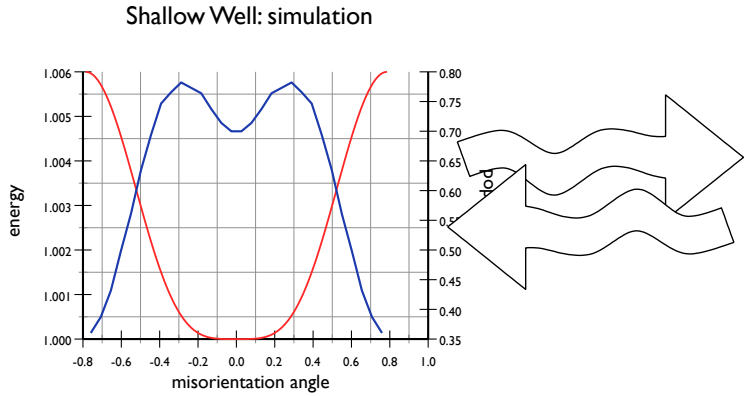


Bridging micro and macro scales

People have known for a long time that grain boundary populations vary with interfacial energy, as well as other features. We want to suggest that this relationship is quantitative and predictable.



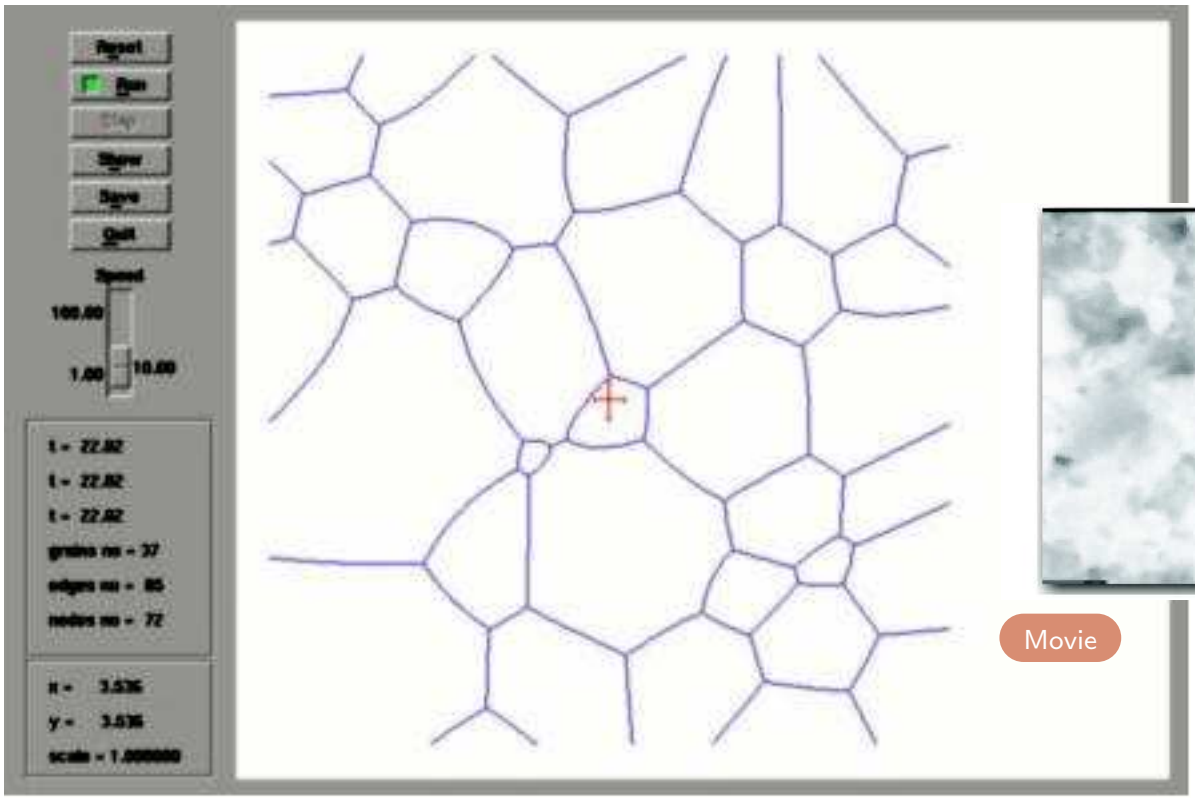
Individual grain dynamics



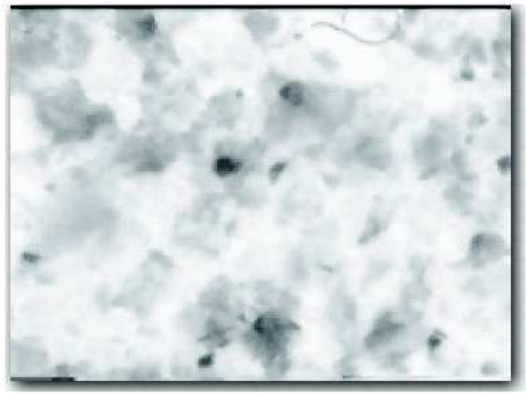
Cumulative probability distributions

Evolution equations for distribution functions

Coarsening: Numerical simulation and experiment



Movie



Movie

Some grains grow, some shrink. Is this the whole story?

Movies courtesy of D. Kinderlehrer, I. Livshits, S. Taasan, K. Barmak.

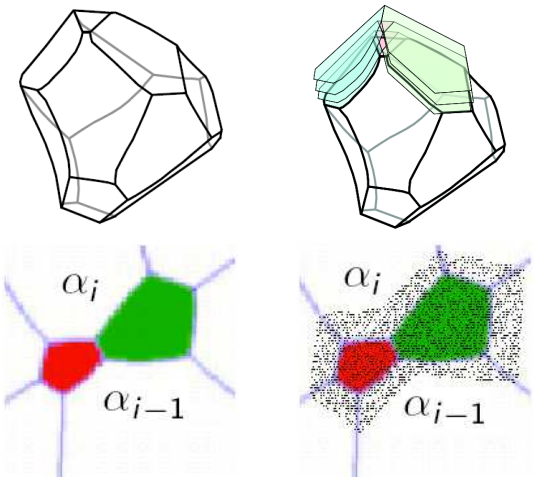
Motivation

Processes that alter GB distributions:

Continuous part

Will not be discussed in this presentation.

Incremental changes in the areas of faces: as grain boundaries migrate, areas of GB faces increase or decrease.



Doesn't affect misorientations, can be modeled using calculus on manifolds.

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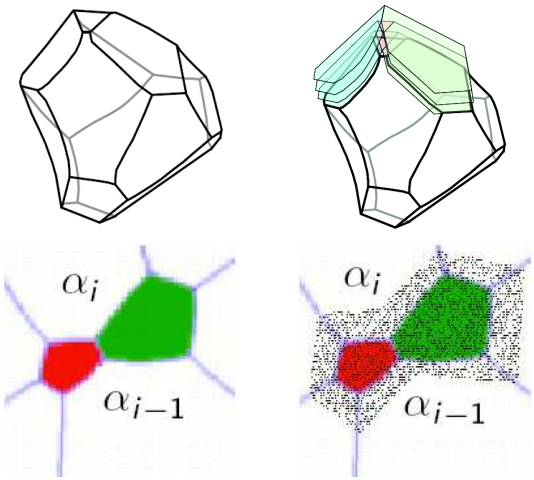


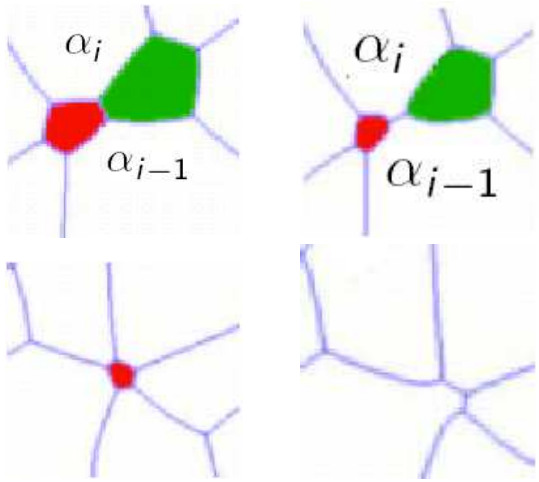
Doesn't affect misorientations, can be modeled using calculus on manifolds.

Discontinuous (jump) part

The focus of present work.

Critical events:

- collapse and creation of grain faces
- collapse of small grains



Affects the misorientations and interfacial energies. Stochastic tools are more effective.

Goals

Stable distribution has been identified (GBCD). Now need to identify ways to model it.

- **Grand challenge**: Develop predictive theory for the evolution of materials texture that will
  - identify stationary statistics
  - provide a dynamical model based on critical events
  - quantify rates at which critical events take place
  - help explain relationship between energy and GB distributions

- **What we talk about today**: Theoretical approaches to modeling evolution of critical events.
  - Modeling correlated populations via Boltzmann approach
  - Generalized continuous time random walk framework
  - Markov modulated Poisson processes
  - Numerical features, results, limitations

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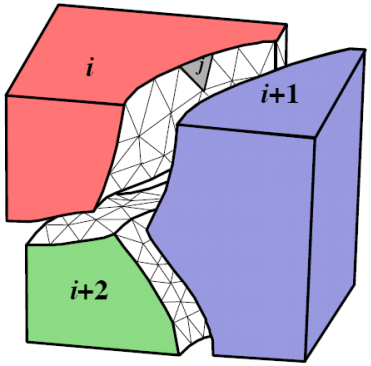
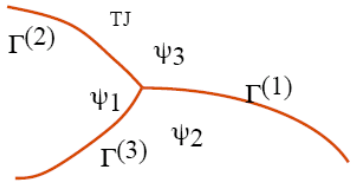
Simplified model

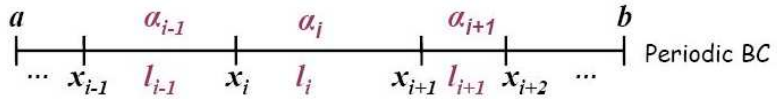
3D



2D



1D



α_i - misorientations

$\gamma(\alpha)$ - interfacial energies

x_j	–	"triple junctions"
(x_j, x_{j+1})	–	"grain boundaries"
$l_j = x_{j+1} - x_j$	–	"GB areas"
$\{\alpha_i\}_{i=1,\dots,n}$	–	"misorientations"

Given initial orientations α_i and total system energy in the form

$$E_n(t) = \sum f(\alpha_i)(x_{i+1}(t) - x_i(t))$$

define equations of motion through gradient flow dynamics

$$\dot{x}_i = f(\alpha_i) - f(\alpha_{i-1}), \;\; i = 0, \dots, n.$$
$$v_i = \dot{l}_i = \dot{x}_{i+1} - \dot{x}_i = f(\alpha_{i+1}) - 2f(\alpha_i) + f(\alpha_{i-1})$$

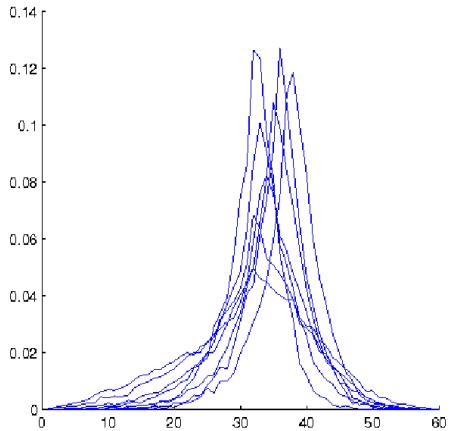
where $f(\alpha)$ plays the role of an interfacial energy.

Stable statistics

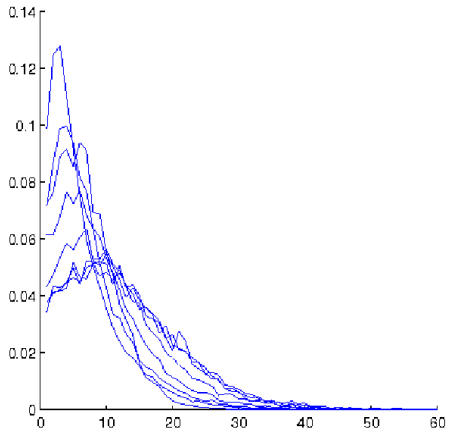
We consider the following statistics:

$$\rho_{or}(\alpha, t) = \sum_k \delta(\alpha - \alpha_k) \quad \text{pdf of orientations}$$
$$\rho_{len}(l, t) = \sum_i \delta(l - l_i) \quad \text{pdf of lengths}$$
$$\rho_w(\alpha, t) = \sum_k l_k \delta(\alpha - \alpha_k) \quad \text{weighted pdf of orientations (GBCD)}$$
$$\rho_{vel}(v, t) = \sum_j \delta(v - v_j) \quad \text{pdf of velocities}$$

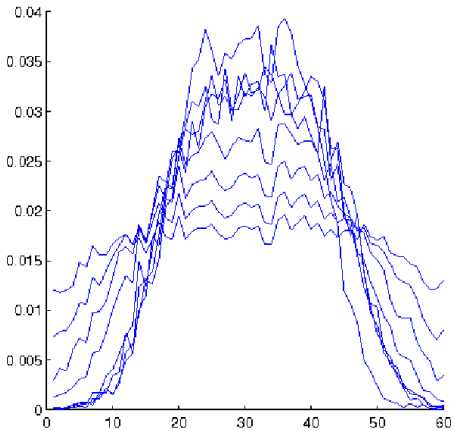
Distributions harvested from simulation stabilize early in the process. Gives evidence for a well-defined dynamic process.



grain boundary velocities



grain boundary lengths



orientations

Influence of interfacial energies

Conforming to experimental observations, distribution of orientation parameters α is inversely correlated with interfacial energies $f(\alpha)$.

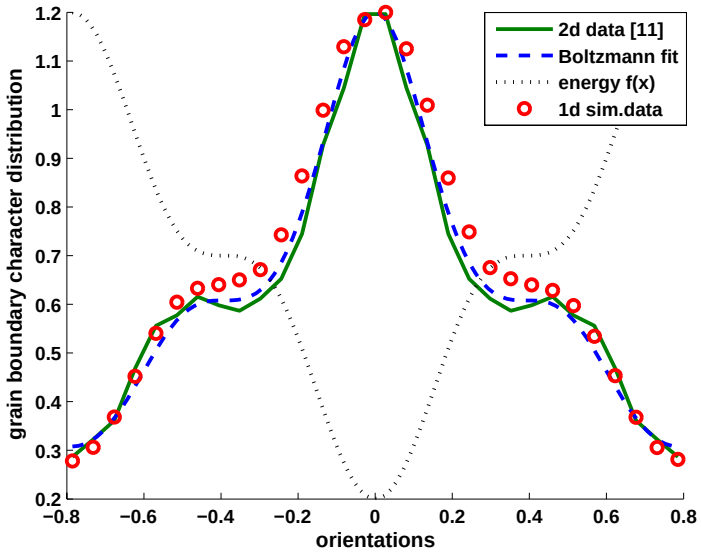
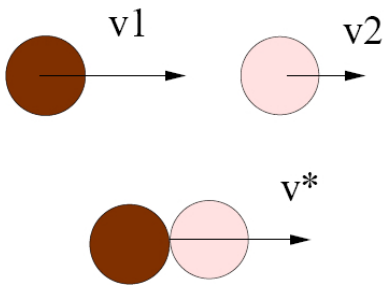


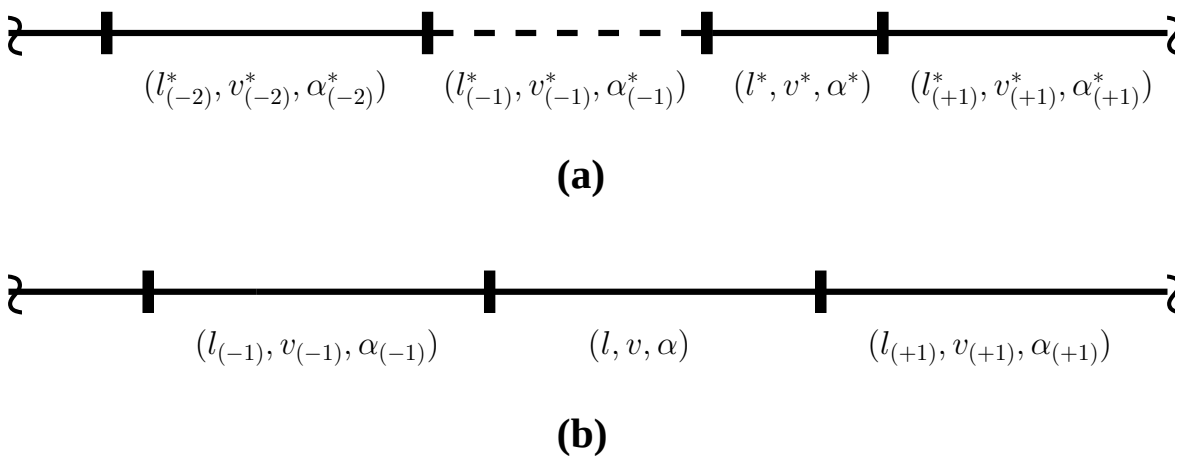
Figure: `GBCD distribution for 1d model, 2d large-scale simulation and its Boltzmann fit.`

Modeling critical events via inelastic Boltzmann equation

We can think of a critical event as a collision of inelastic particles: particle $\equiv$ GB.

$$(\alpha_2, v_2) + (\alpha_1, v_1) \Rightarrow (\alpha_1, v^*), \text{ where } v^* = v_1 + v_2 + f(\alpha_2) - f(\alpha_1),$$





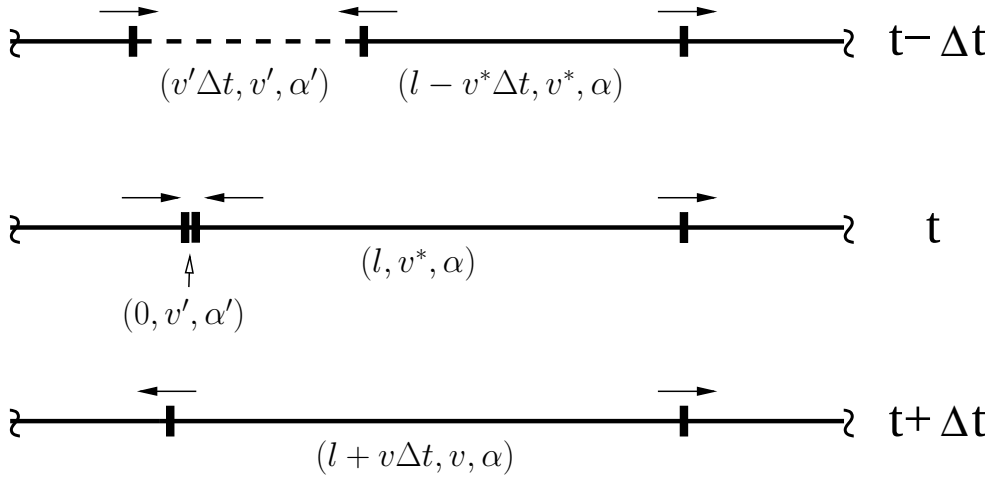
Continuity equation:

$$\frac{\partial \rho(l, v, \alpha, t)}{\partial t} + v \frac{\partial \rho(l, v, \alpha, t)}{\partial l} = \{gain\} - \{loss\}.$$

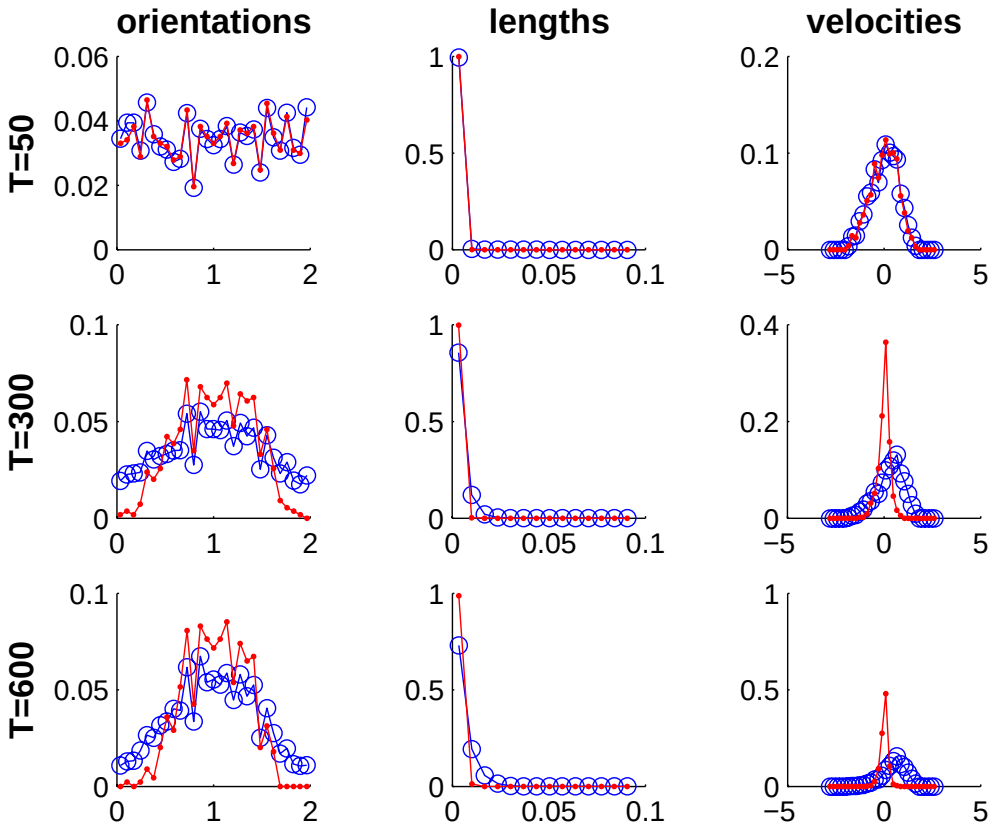
where the right hand side term can be written as an integral over all possible collisions:

$$-\frac{2}{N(t)} \int_{A_+} v' \rho(0, v', \alpha', t) \rho(l, v - v' + f(\alpha) - f(\alpha'), \alpha, t)\, d\alpha' dv'$$
$$+\frac{2}{N(t)} \int_{A_-} v' \rho(0, v', \alpha', t) \rho(l, v, \alpha, t)\, d\alpha' dv'.$$

subject to restricting conditions:
$A_- := \{f(\alpha) \le v' + 2f(\alpha')\} \cap \{f(\alpha') \le v + 2f(\alpha)\} \cap \{v' < 0\}$,
$A_+ := \{f(\alpha) \le v' + 2f(\alpha') \le v + 3f(\alpha)\} \cap \{v' < 0\}$.



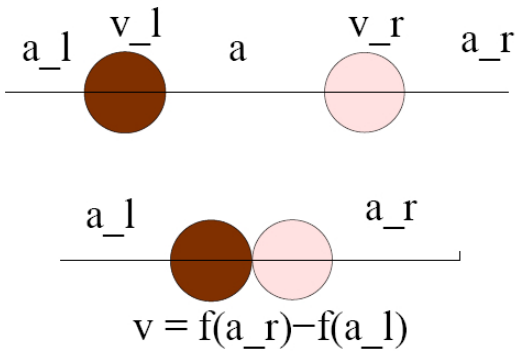
Simple model performance for quadratic potential



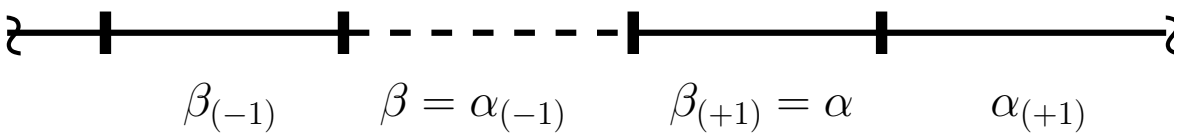
Lengths and orientations fit experimental results well, but velocities start to deviate at later stages. Possible correlations?

Extended space model

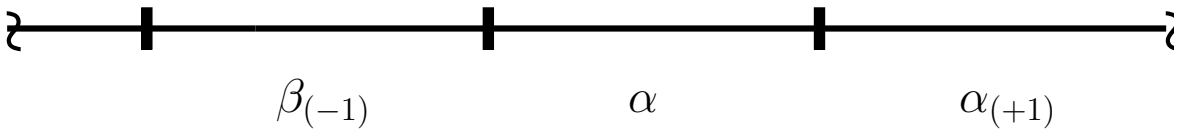
Is there a remedy for complexity and correlation issues?
Then let us expand the state space to include incidence relations: $\rho(l, \alpha_{(-1)}, \alpha, \alpha_{(+1)}, t)$.



Grain boundaries ≡ colliding particles.



(a)



(b)

By following these simple collision rules, we can write a Boltzmann-type collision equation for the density function:

$$\frac{\partial \rho\left(l, \alpha_{(-1)}, \alpha, \alpha_{(+1)}\right)}{\partial t} + \left(f\left(\alpha_{(-1)}\right) + f\left(\alpha_{(+1)}\right) - 2f(\alpha)\right) \frac{\partial \rho\left(l, \alpha_{(-1)}, \alpha, \alpha_{(+1)}\right)}{\partial l} = W,$$

where $W = W_+ - W_-$ with

$$W_+ := \frac{1}{N(t)} \int_{\mathcal{B}} \left(2f(s) - f(\alpha) - f(\alpha_{(-1)})\right) \rho\left(0, \alpha_{(-1)}, s, \alpha\right) \rho\left(l, s, \alpha, \alpha_{(+1)}\right) \, ds$$
$$+ \frac{1}{N(t)} \int_{\mathcal{B}} \left(2f(s) - f(\alpha) - f(\alpha_{(+1)})\right) \rho\left(0, \alpha, s, \alpha_{(+1)}\right) \rho\left(l, \alpha_{(-1)}, \alpha, s\right) \, ds.$$

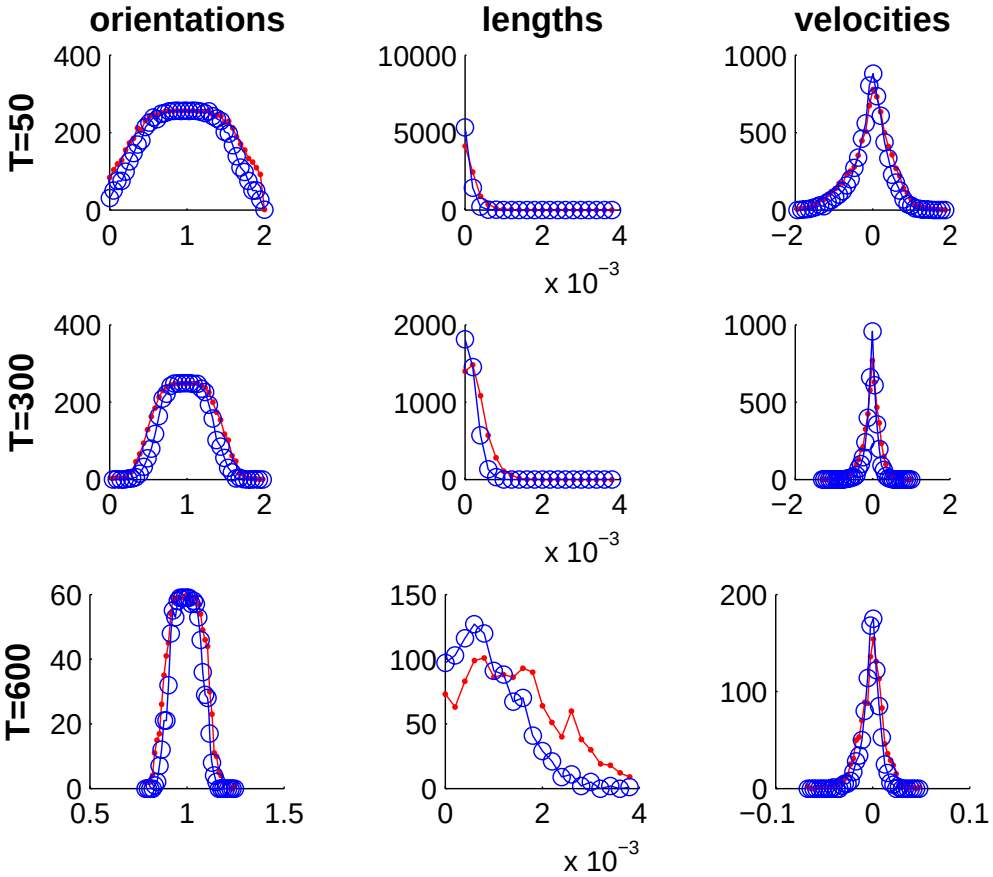
Similarly

$$W_- := \frac{1}{N(t)} \int_{\mathcal{B}} \left(f(\alpha) + f(s) - 2f(\alpha_{(-1)})\right) \rho\left(0, s, \alpha_{(-1)}, \alpha\right) \rho\left(l, \alpha_{(-1)}, \alpha, \alpha_{(+1)}\right) \, ds$$

$$+ \frac{1}{N(t)} \int_{\mathcal{B}} \left(f(\alpha) + f(s) - 2f(\alpha_{(+1)})\right) \rho\left(0, \alpha, \alpha_{(+1)}, s\right) \rho\left(l, \alpha_{(-1)}, \alpha, \alpha_{(+1)}\right) \, ds.$$

where $\mathcal{B} := \mathbf{R}_+ \times \mathbf{R}^3$.

Extended model performance for quadratic potential



Lengths, orientations and velocities are in agreement with experimental results.

Quick summary

Recap: We have discussed two models capable of describing the evolution of $\rho(\alpha, l, t)$.

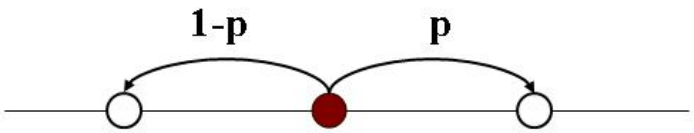
1. Advantages:
   - both models have clear physical interpretation;
   - extended Boltzmann model **completely describes system evolution**
2. Drawbacks:
   - simple model does not take into account correlations
   - extended approach is informationally complex.

Continuous time random walk theory can possibly eliminate some of these difficulties, while also giving more insight into characteristics of underlying **stochastic process**.

Simple random walk model and its master equations

The walker takes a step in a random direction with fixed probabilities p_i, s.t. $\sum p_i = 1$.

Symmetric walk in 1d: $p_i = 1/2$



$$P_i(t + \Delta t) = \frac{1}{2} P_{i-1}(t) + \frac{1}{2} P_{i+1}(t)$$

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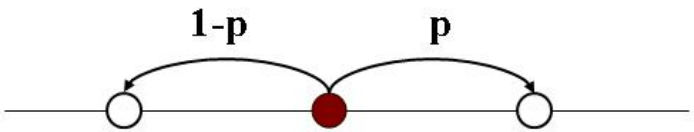
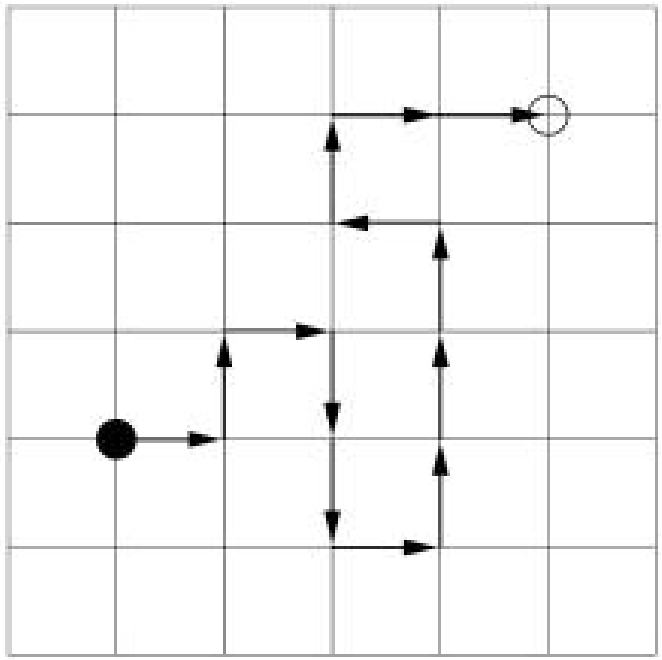


$$P_i(t + \Delta t) = \frac{1}{2} P_{i-1}(t) + \frac{1}{2} P_{i+1}(t)$$

Symmetric walk in 2d: $p_i = 1/4$



$$P_{i,j}(t + \Delta t) = \frac{1}{4}(P_{i,j-1}(t) + P_{i,j+1}(t) + P_{i-1,j}(t) + P_{i+1,j}(t)$$

Master equation and PDE

In the symmetric 1d random walk case, the master equation is given by:

$$P_i(t+\Delta t) = \frac{1}{2}P_{i-1}(t) + \frac{1}{2}P_{i+1}(t)$$

In the continuum limit, $\Delta t \to 0$, $\Delta x \to 0$:

$$P_i(t+\Delta t) = P_i(t) + \Delta t \frac{\partial P_i}{\partial t} + O(\Delta t^2)$$
$$P_{i\pm1}(t) = P_i(t) \pm \Delta x \frac{\partial P_i}{\partial x} + \Delta x^2 \frac{\partial^2 P_i}{\partial x^2} + O(\Delta x^2)$$

leads to the diffusion equation

$$\frac{\partial P_i}{\partial t} = K \frac{\partial^2 P_i}{\partial x^2}$$

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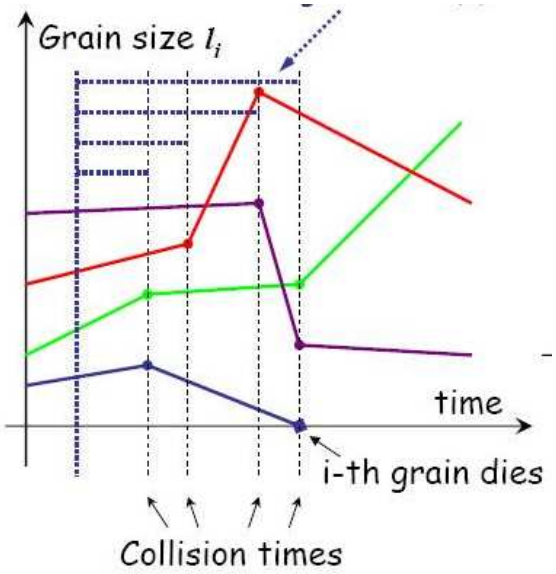
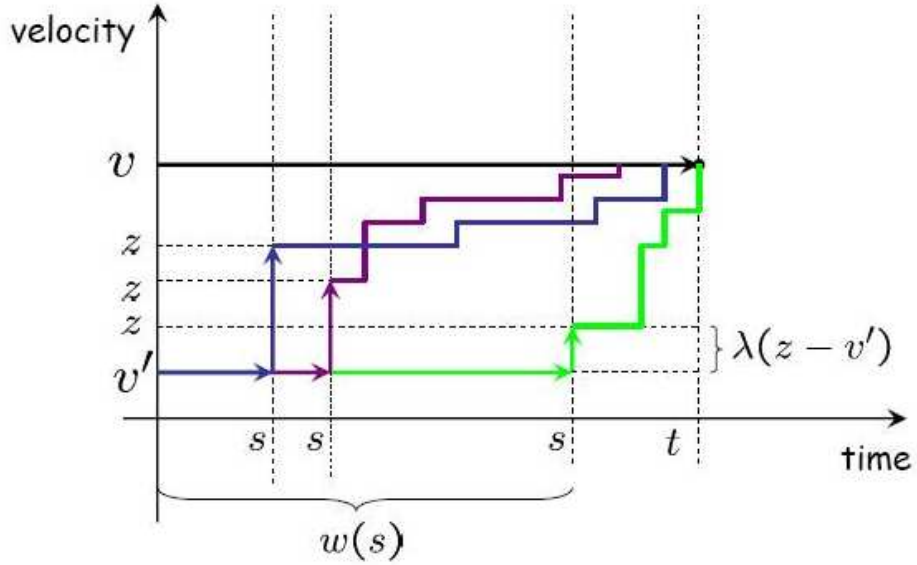
Many generalizations are possible.
Continuous time random walk: lattice is replaced by continuous state space.
Jump sizes and waiting times are drawn from probability distributions.

Continuous time random walk of the lengths and velocities

Grain areas



Grain velocities



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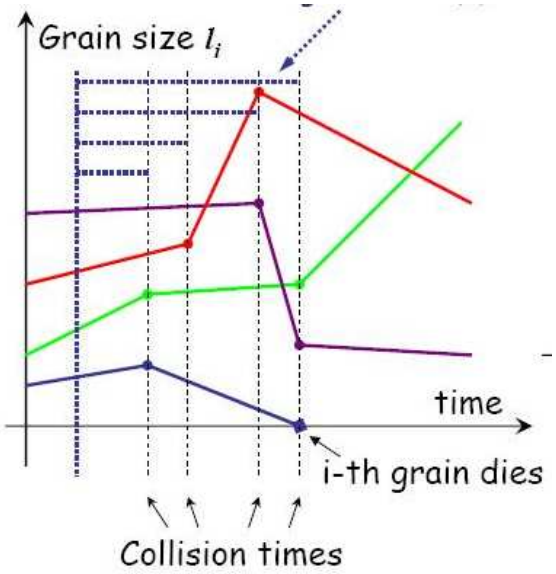

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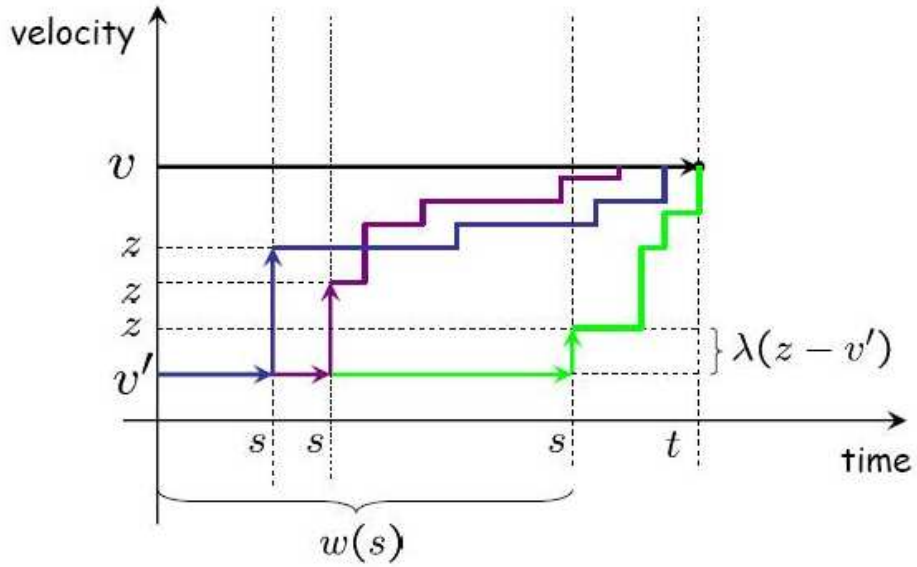
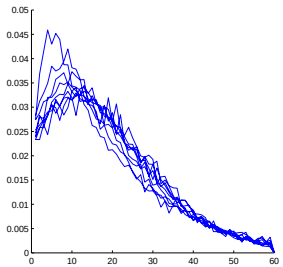
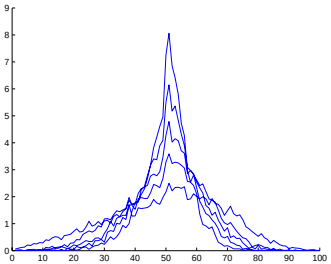


Simulation results: steady-state statistics of l_i and v_i (exists for all types of f)

relative areas

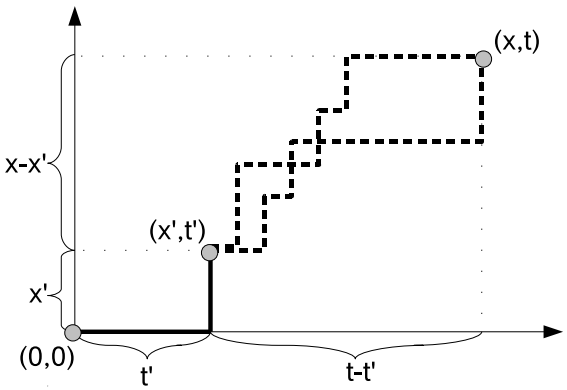


relative velocities



Continuous Time Random Walk framework

The walker moves according to two parameters:
(1) Waiting times with pdf $w(t)$ (2) Jump sizes with pdf $\mu(x)$.



Chapman-Kolmogorov type master equation:

$$p(s,t) = p_0(s)\psi(t) + \int\int p(s-s',t-t')\mu(s')w(t')dt'ds'$$

We derive an equivalent form in terms of the memory kernel $\hat{\Phi}(u) = \dfrac{1-\hat{w}(u)}{u\hat{w}(u)}$:

$$\int \Phi(t-t')\frac{\partial}{\partial t}p(x,t')dt' = \int [p(x-x',t) - p(x,t)]\mu(x')dx'$$

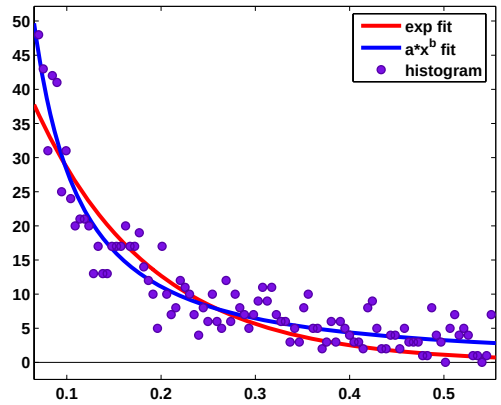
- $\Phi(t) = c\delta(t)$ (equivalent to $w(t) \sim \exp(\lambda t)$: Markov process, no memory
- $\Phi(t) \neq c\delta(t)$: non-Markov process with memory kernel $\Phi(t)$.
  For the choice of $\hat{\Phi}(u) = \frac{1}{\lambda}u^{\beta-1}$

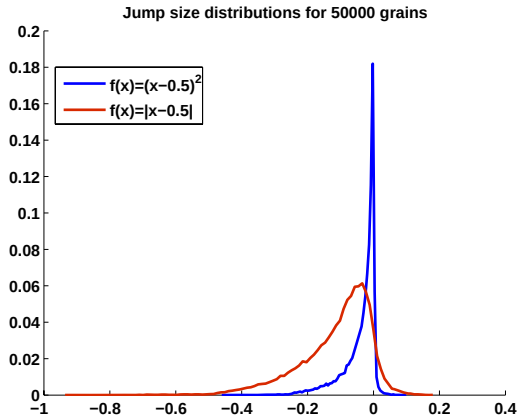
$$\frac{\partial^\beta}{\partial t^\beta}p(x,t) = \lambda \int [p(x-x',t) - p(x,t)]\mu(x')dx'$$

Non-Markov nature of dynamics

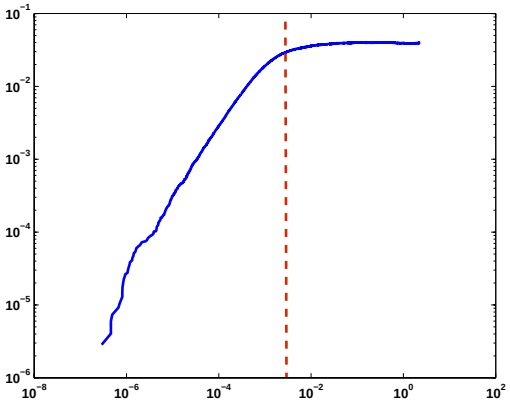
Do we have a Markov process in our model problem? No!

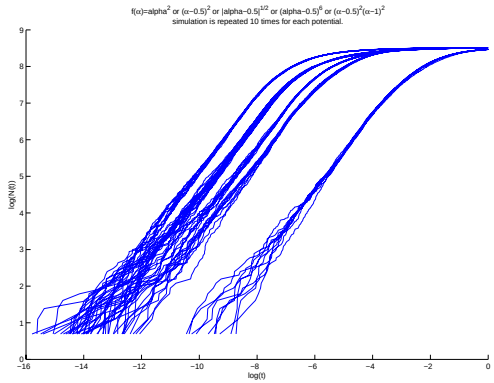
- Waiting times have fat tails (left), plus jump sizes are non-Gaussian (right)





- MSD $\langle v^2 \rangle$ (left) and arrival rate (right) both show nonlinear trend in the long term limit, independent of the form of the potential. Log-log slope changes from $\beta = 1$ to $\beta \approx 0.7$.





Closed form solution

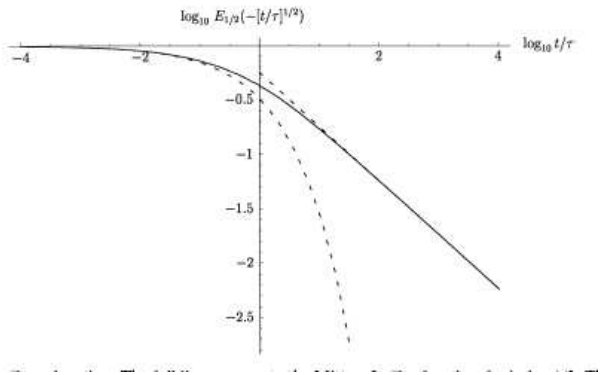
- For stable jump sizes distribution $\mu(s) = \alpha s^{-\alpha-1}, \quad 0 < \alpha < 1, \quad s > 0$ and waiting times $w(t) \sim t^{-1-\beta}$, the master equation coincides with the fractional kinetic equation:

$$\frac{\partial^\beta p(x,t)}{\partial t^\beta} = K_\gamma \frac{\partial^\alpha}{\partial x^\alpha} p(x,t),$$

- It admits a closed form solution in the form

$$p(x,t) = \frac{1}{t^{\beta/\alpha}} W_{\alpha,\beta}\left(\frac{x}{t^{\beta/\alpha}}\right),$$

$W_{\alpha,\beta}$ is the inverse Laplace transform of the Mittag-Leffler function $E_{\alpha,\beta}(z) = \sum_{j=0}^{\infty} \frac{z^j}{\Gamma(\alpha j + \beta)}$



- Fractional derivative in Caputo form

$$\frac{d^\beta}{dt^\beta} f(t) = \frac{1}{\Gamma(1-\beta)} \int_0^t \frac{f'(\tau)}{(t-\tau)^\beta} d\tau,$$

for which

$$\mathcal{L}\left[\frac{d^\beta}{dt^\beta} f(t)\right] = u^\beta \hat{f}(u) - u^{\beta-1} f(0).$$

Numerical results

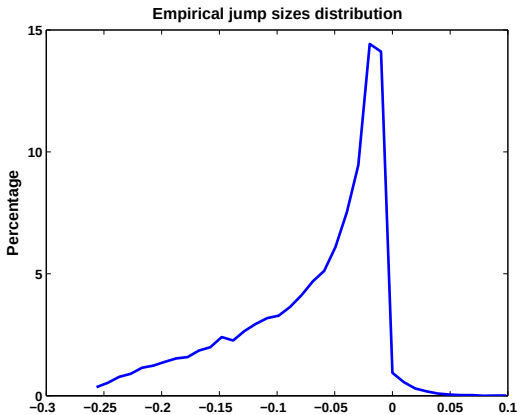
We look at our simplified model in the coupled 2-dimensional case space of velocities and lengths:

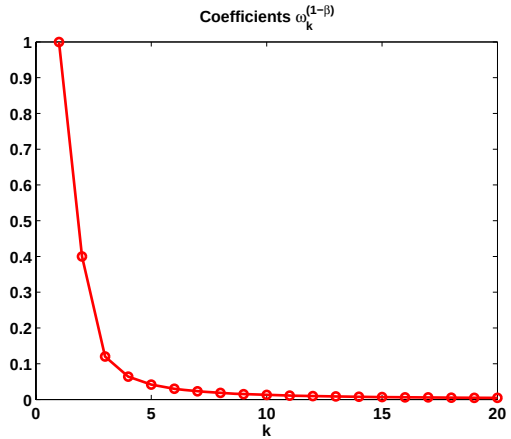
$$\frac{\partial^\beta}{\partial t^\beta}\rho(l,v,t) = -v\frac{\partial \rho(l,v,t)}{\partial l} + \lambda \int_{-\infty}^{\infty} [\rho(l,v-s,t) - \rho(l,v,t)]\mu(s)ds.$$

Using Grunwald-Letnikov definition of fractional derivative:

$$\frac{\partial^\beta f(t)}{\partial t^\beta} = \lim_{h\to 0} \frac{1}{h^\beta} \sum_{k=0}^{[t/h]} \omega_k^\beta f(t-kh), \quad \text{where} \quad \omega_k^{(\beta)} = (-1)^k \begin{pmatrix} \beta \\ k \end{pmatrix}.$$

We determine $\mu(s)$ from the empirical jump probability density and run the model with parameter $\lambda = 1$.





Comparison of simulation with fractional PDE.

- Taking $\beta = 0.7$, numerical solution of the fractional equation can be obtained by using the explicit FTCS difference scheme

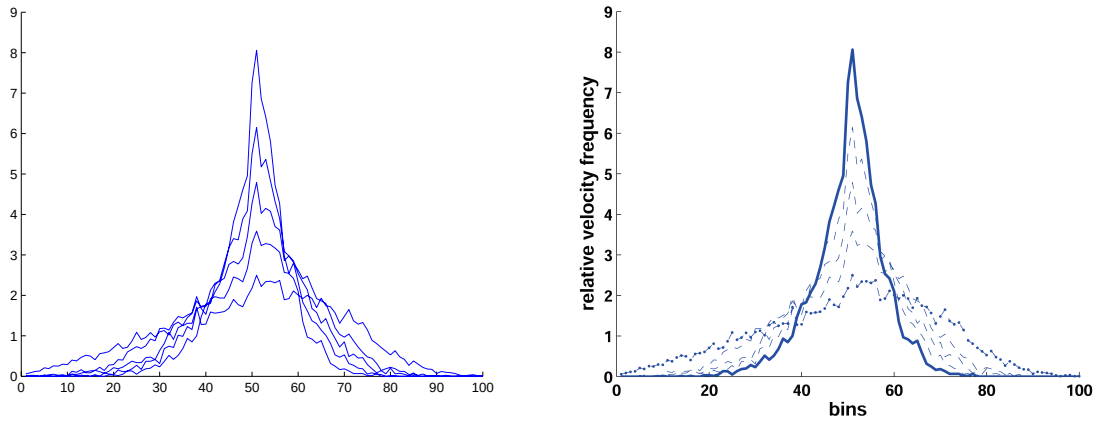
$$p_{i,j}^{(m+1)} = p_{i,j}^{(m)} + S_\beta \sum_{k=0}^{m} \omega_k^{(1-\beta)} I_{i,j}^{(m-k)},$$

where $S_\beta = \frac{\Delta^\beta}{(\Delta x)^2}$, $\Delta t \leq \frac{1}{4^{3/2-\beta}} (\Delta x)^{\frac{2}{\beta}}$ and

$$I_{i,j}^{(m-k)} = v_j (p_{i-1,j}^{(m-k)} - p_{i+1,j}^{(m-k)}) \Delta x + \sum_{s_l : j+s_l \in [1,Nj]} \mu_l [p_{i,j+s_l}^{(m-k)} - p_{i,j}^{(m-k)}].$$

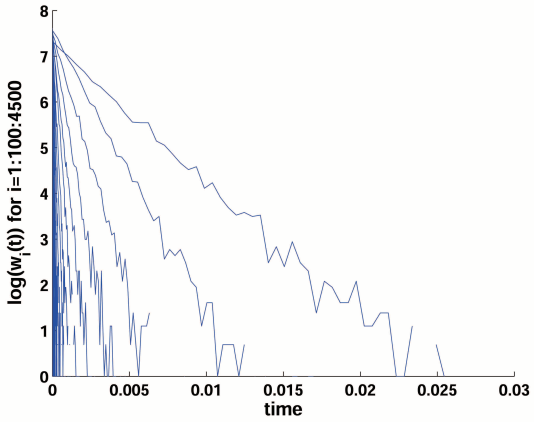
which has been shown to be numerically stable.

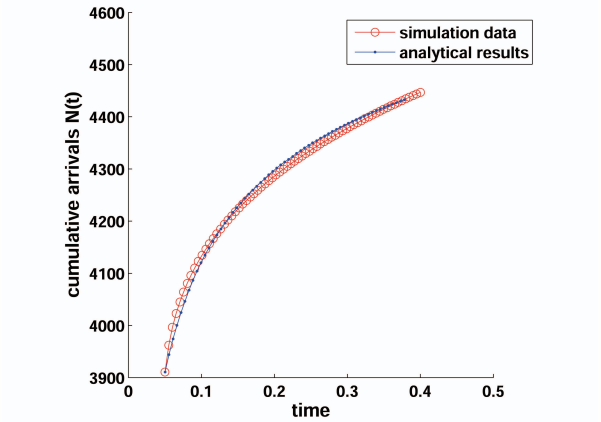
- FPDE (right) are in agreement with simulation (left).

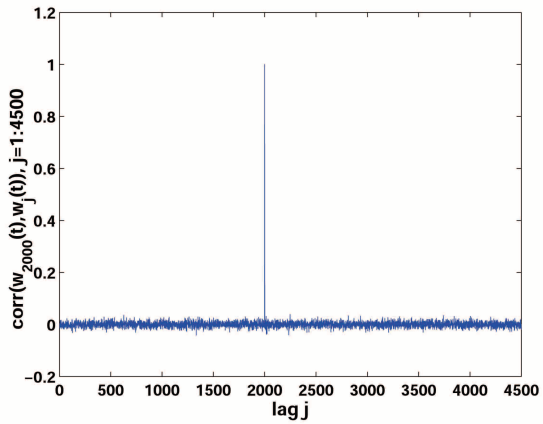


Slowing down effect

Departure from fractional random walk theory: non-identical waiting times distributions







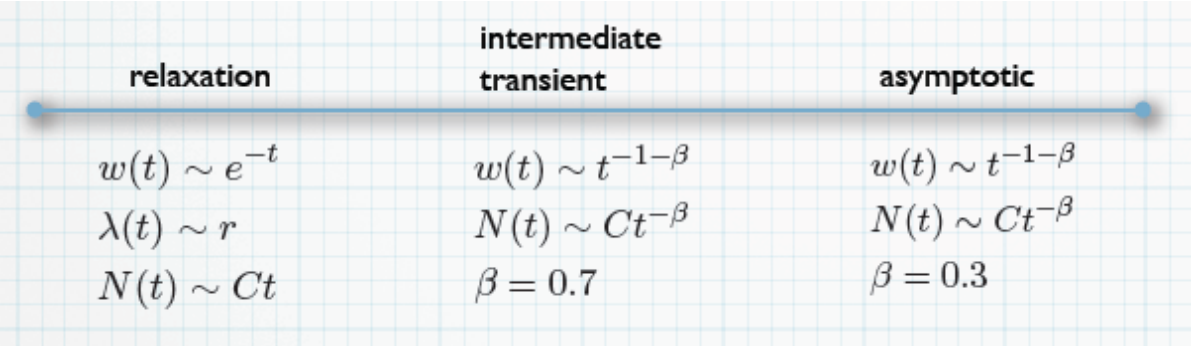
Recall: for a random walk theory, one has to assume that all waiting times are i.i.d. variables. In this case, variables are independent, but not identically distributed. Analogy to non-homogeneous Poisson process with decaying frequency.

Connections to renewal processes theory

$N(t)$ - cumulative arrivals, $\lambda(t)$ - arrival rate, $w(t)$ - waiting times pdf

$$\lambda(t) = w(t) + \int_0^t \lambda(s)w(t-s)ds, N(t) = \int_0^t \lambda(s)ds$$

Fractional regime only describes intermediate steps of the process.



Is there a formalism capable of describing the complete evolution process?
Can we find an evolution equation for pdfs related to velocities and GBCD in this context?

Markov-modulated Poisson processes

Relation to hidden Markov chains:

- $w_i \sim \exp(r_i)$, with mean parameter r_i that increases with i due to the slowing effect of coarsening dynamics
- Markov modulated Poisson process(MMPP) - Poisson process varying its arrival rate according to an m-state irreducible continuous time Markov chain.
- The observable process can be written as $X_t = X_0 + \int_0^t AX_u du + M_t$ with a martingale part M_t and with rate of arrivals depending upon the state of an indirectly observed Markov chain with rate matrix A having entries r_i.

MMPP is an attractive model for short- and middle-range autocorrelations because it can be easily parameterized to produce dependence in the series and remains analytically tractable.

MMPP parameter estimation

$X(t)$ - underlying continuous-time homogeneous Markov chain with state space $1, \ldots, r$.
r - order of the MMPP.
$Q = q_{ij}$ - generator of the Markov chain.
$N(t)$ observed process - variable-rate Poisson process with rate λ_i when $X(t) = i$.
T_i - time of the i-th Poisson event. Inter-event times $Y_k = T_k - T_{k-1}$.
$X(T_k)$ - embedded discrete-time Markov chain obtained from sampling the continuous-time chain at the Poisson event times.
$(X(T_k), Y_k)$ - Markov renewal process with transition probability matrix $F(y) = \{F_{ij(y)}\}$, where $F_{ij}(y) = P(Y_k \leq y, X(t_{k-1} + y) = j | X(t_{k-1} = i)$. The transition density matrix, obtained from differentiation of F_{ij} w.r.t. y, is given by

$$f(y) = exp((Q - \Lambda)y)\Lambda, \Lambda = diag\,\lambda_1, \ldots, \lambda_r.$$

Parameters of the stationary MMPP: (Q, Λ) can be estimated by EM algorithm.
Preliminary results: $r = 10$ fits well with 1-d simulation interarrival times for $N = 10^3$ grain boundaries.

Work in progress

Further progress can only be achieved through close collaboration with materials scientists.

- Building MMPP formalism for GBCD evolution
- Understanding the slowing down effect and its relationship to the fractional dynamics
- Providing physical validation
- Studying numerical and analytical issues
- Developing the complete predictive theory for the evolution of materials statistical (texture, GBCD etc) and structural properties from microstructure

  In particular, it will

  - identify stationary statistics
  - provide a dynamical model based on critical events
  - quantify rates at which critical events take place
  - provide the set of simple rules to be used for grain boundary engineering

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THANKS!