Single-File Diffusion far from thermal equilibrium

Gunter M. Schütz

IFF, Forschungszentrum Jülich, 52425 Jülich, Germany

1. Statistical mechanics far from equilibrium
2. Stochastic processes and quantum field theory
3. Boundary-induced phase transitions
4. Boundary-driven systems
5. Conclusions and acknowledgments

1. Statistical Mechanics Far From Equilibrium

,,(Die Physik) verdankt ihren phänomenalen Erfolg ihrem Mut und ihrer Bereitschaft, Dinge auf abstraktester Ebene zu formulieren, vereinfachte, paradigmatische Anordnungen auszuwählen oder zu konstruieren, und ganze unterschiedliche Phänomene durch einfache Prinzipien zu verstehen.“

C. von der Malsburg, Phys. Blätt. 57 (2001), 3

Physics owes its tremendous success to its courage and its readiness to handle matters on a most abstract level, to choose or construct simplified paradigmatic settings, and to understand entirely different phenomena in simple generic terms.

Statistical Mechanics: simple paradigmatic settings ➔ simple models

❖ Address fundamental problems of many-body theory

emergence of new properties on coarse grained level of description
Universality provides for the study of highly stylized models (ideal gas, Ising model, ...)

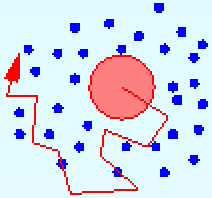
❖ Applications to specific systems

quantitative: critical exponents … (exploiting scale invariance and universality)
semiquantitative: smal number of fitting parameter for dominant mechanisms (and “easy” to simulate)
qualitative: existence of or mechanisms for specific phenomena (shocks, hysteresis, nucleation, …)

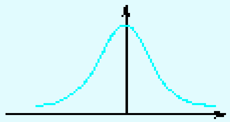
Self-organisation on all scales

microscopic → mesoscopic → macroscopic

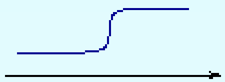
microscopic	mesoscopic	macroscopic
Particles Forces	Brownian motion effective forces	Density, transport coefficients diffusion, shocks, ...



$(\vec{x}_i, \vec{p}_i)$ $(\vec{x}_i, \vec{p}_i)$



$P(\vec{x}_i)$



$\rho(\vec{x})$

Newtonian equations	master equation	nonlinear PDE
(deterministic)	(stochastic)	(deterministic)

Mesoscopic level: we use lattice gas models with diffusive motion and particle interactions

Macroscopic level: we wish to derive hydrodynamic equations (Burgers, shallow-water, Fisher, …) and generic properties of transport coefficients

Fundamental problems:
- novel phenomena: existence and dynamical origin of phase transitions
- hydrodynamic limit
- large deviation theory for nonequilibrium steady states (fluctuation theorems, equations of state)

Applications of quasi one-dimensional diffusive particle systems (single-file diffusion):

- physics: diffusion in zeolithes, colloidal systems in confined geometry, entangled polymers ...
- biology: molecular motors, protein synthesis, ant trails ...
- other: automobile traffic, option pricing, ...

Methods:

❖ Exact results: Integrability, probabiilistic concepts, PDE theory

❖ Scaling theory

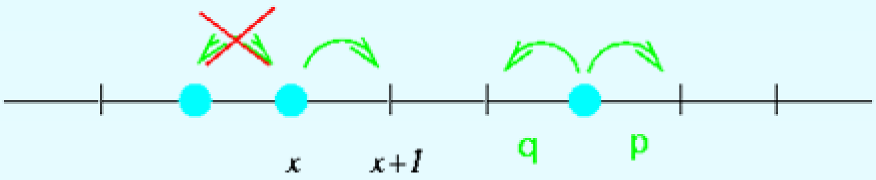
❖ Numerics: Monte-Carlo simulation, DMRG, PDE

❖ ~~Mean field~~ (coincidental if it works, no physics)

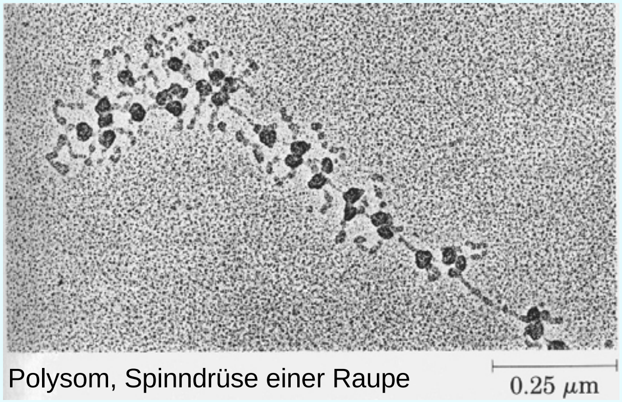
2. Stochastic Processes and Quantum Field Theory

Asymmetric Simple Exclusion Process (ASEP)

- short range interaction (excluded volume)
- diffusive motion (random walk with exponential waiting time)
- drift (external potential or self-propelled)
- open boundaries ➔ permanent nonequlibrium state



MacDonald, Gibbs, Pipkin (1968); Spitzer (1970)



Polysom, Spinndrüse einer Raupe

Proteinsynthese: Übersetzung der mRNA durch mehrere Ribosomen. Die Polypeptidketten werden in Translationsrichtung länger.

density $\rho_x(t) = \langle n_x(t) \rangle$ current $j_x = p \langle n_x(1-n_{x+1}) \rangle - q \langle (1-n_x) n_{x+1} \rangle$

continuity equation $\rho_x(t) = j_{x-1}(t) - j_x(t)$

Master equation:

$$\frac{d}{dt} P(\eta,t) = \sum_{\eta'\neq\eta} [\, w(\eta',\eta\} P(\eta',t) - w(\eta,\eta') P(\eta,t)] \qquad \eta \; 2 \; \{0,1\}^{- L}$$

Quantum hamiltonian representation:

$\eta \rightarrow | \eta > \; 2 \; C^{- L}$ $\qquad P(\eta,t) = <\eta \,|\, P(t) >$

Schrödinger equation
in imaginary time

$$\frac{d}{dt} | P(t) > = - H \,|\, P(t) >$$

$H = - \sum_k [\, \sigma^x_k \sigma^x_{k+1} + \sigma^y_k \, \sigma^y_{k+1} + \Delta \, \sigma^z_k \sigma^z_{k+1}]$ - n.h. boundary terms (Heisenberg ferromagnet; spinless fermions)

Bethe ansatz → dynamical exponent, current fluctuations [Gwa,Spohn (1992), Derrida, Lebowitz, Speer (1999)

quantum group → dynamics and microscopic structure of shocks [Belitsky, G.M.S. (2002)]

matrix product ansatz → Bethe ansatz, open boundaries

Poster Tabatabaei

Dynamic matrix product ansatz (DMPA)

Derrida, Evaans,, Pasquieer, Saleur (1993); Stinchcombe, G.M.S. (1995); Alcaraz, Lazo (2004)

periodic n-component system: $| P(t) > = \mathrm{Tr} \begin{pmatrix} E \\ A\_1 \\ A\_2 \\ \ldots \\ A\_n \end{pmatrix}^{-L}$

$<00101110\ldots> = \mathrm{Tr}\,(EEAEAAAE\ldots)$

Fourier transformation of $E^{k-1} A_i E^{-k}$ $\qquad D(p) = \begin{pmatrix} D_1(p) \\ D_2(p) \\ \ldots \\ D_n(p) \end{pmatrix}$

$$D(p) - D(q) = \Sigma(p,q)\, D(q) - D(p)$$

$$S_{12}\, S_{13}\, S_{23} = S_{23}\, S_{13}\, S_{12}$$

$\Sigma(p,q)\,\Sigma(q,p) = 1$ (unitarity) $\qquad S = \Sigma P$ (S-matrix of integrable Zamolodchikov field theory (1979)) periodic system: DMPA → functional equation → Bethe wave function → relaxation spectrum

stationary state (periodic und open): recursion relations → stationary nonequilibrium distribution

TASEP: density profile from empty-interval probabilities $Y^k_L = \langle \ldots 0000 \rangle$, $X^k_L(p) = \langle \ldots 1 \ldots 0000 \rangle$

$Y^k_L = Y^{k-1}_L + \alpha\beta\, Y^k_{L-1}$ $\quad 1 < k < L+1$

$Y^1_L = \beta\, Y^1_{L-1}$

$Y^{L+1}_L = Y^L_L + \alpha\, Y^L_{L-1}$

$X^k_L(p) = X^{k-1}_L(p) + \alpha\beta\, X^k_{L-1}(p)$ $\quad p+1 < k < L+1$

$X^{1+p}_L(p) = \alpha\beta\, Y^{1+p}_{L-1}$

$X^{L+1}_L(p) = X^L_L(p) + \alpha\, X^L_{L-1}(p)$

$Y^1_1 = \beta$, $Y^2_1 = \alpha + \beta$

Guess solution from explicit computation for small system size:

$$j = Z_{L-1} / Z_L$$

$$\rho_k = \frac{1}{Z_L} \sum_{p=1}^{k} \Phi_p(\alpha)\, \Phi_{L-p}(\beta)$$

$$\Phi_N(x) = 2 \binom{2N}{N} \; {}_2F_1\,(1,N+1/2,1/2;(1-2x)^2) + (1/2-x)/[x(1-x)\}^N$$

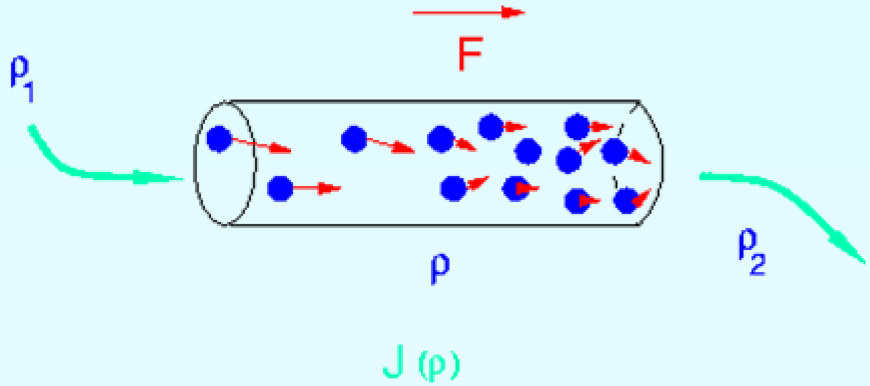
G.M.S., Domany (1993)
Derrida, Evans, Hakim, Pasquier (1993)

3. Boundary induced phase transitions

Driven diffusive systems (DDS)

- bulk: (i) biased random motion, (ii) short range interaction, (iii) particle conservation

- boundaries: coupling to external reservoirs with fixed densities



stationary particle current after relaxation

⟶ permanent nonequilibrium steady state

selection of NESS?

dynamical mechanism?

} no answer from known principles

phase diagram of ASEP with open boundaries, reservoir densities $\rho_{+/-}$

➔ current $j = (p-q)\, \rho\, (1-\rho)$ + f.s. corrections (approx. uncorrelated bulk state)

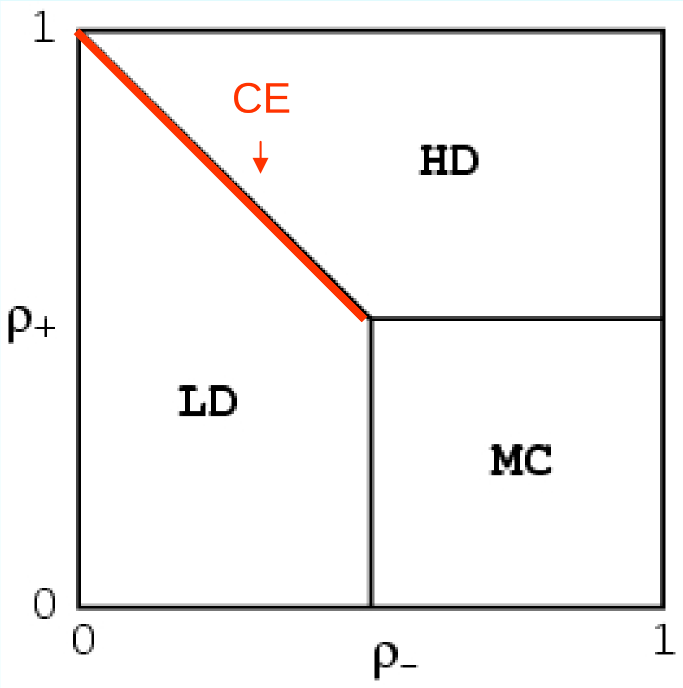
➔ three phases with different bulk densities ρ

LD: low density, $\rho = \rho_- < ½$,

HD: high density, $\rho = \rho_+ > ½$,

MC: maximal current $\rho = ½$, self-organized (second-order transition)

CE: coexistence line, linear density profile (first-order transition)



macroscopic theory (hydrodynamic limit a->0):

$$\partial_t \rho + \partial_x j(\rho) = 0$$

Rezakhanlou (1991)

shock discontinuities, nonuniqueness of solutions of initial- and boundary-value problem resp.
→ entropy solutions or: regularization with phenomenological infinitesimal viscosity

regularization: excess current (Krug 1991), lattice constant (Popkov, G.M.S. 2003)

↓

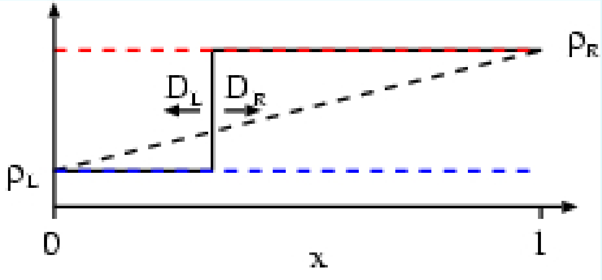
Generic extremal principle for stationary current

$$j = \begin{cases} \max\limits_{\rho_- > \rho > \rho_+} \{ j(\rho) \} & \rho_- > \rho_+ \\ \min\limits_{\rho_+ > \rho > \rho_-} \{ j(\rho) \} & \rho_+ > \rho_- \end{cases}$$

Popkov, G.M.S. (1999)
rigorous proof: Bahadoran (2005)

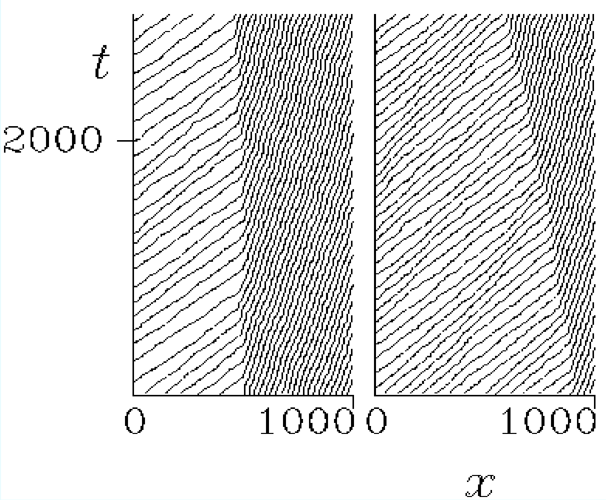
microscopic derivation:

shock diffusion (first order transition)



biased Brownian motion of
microscopically sharp shock

shock velocity:



$$v_s = \frac{j_L - j_R}{\rho_L - \rho_R}$$

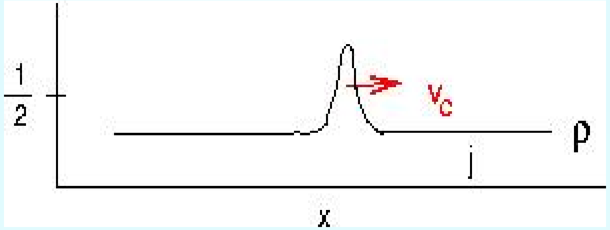
(mass conservation)

flow of microscopically localized perturbations

biased (superdiffusive) motion
with collective velocity

$$v_c = j'(\rho)$$

(Nonequilibrium-FDT, mass conservation)



- microscopic shock stability $v_c^L > v_s > v_c^R$

- saturation (second order transition) at $v^L_\wedge = 0$

➔ explicit construction of steady state selection ➔ extremal principle

No model-dependent assumption → generic behaviour

hydrodynamic limit: collective velocity -> speed of characteristics
shock stability -> Lax-criterion for shocks

DDS with slow reaction kinetics

ASEP + bulk Langmuir kinetics A ⇔ 0 [Willmann, GMS, Challet (2002); Parmeggiani, Franosch, Frey (2003); Klumpp, Lipowsky (2004)]

- Localization of shock (“freezing”) despite extra noise (“heating”)
- effective potential for shock
- hydrodynamic description (contiuity equation with source term)

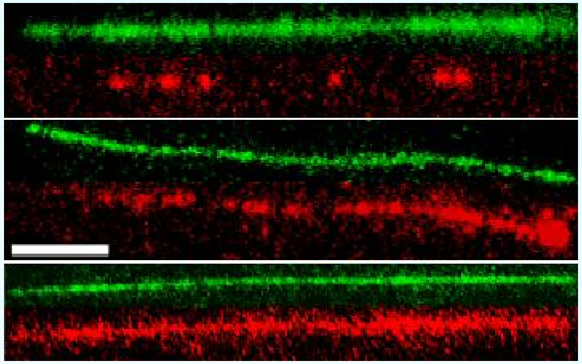
ASEP + activated bulk Langmuir kinetics A0A ⇔ AAA [Rakos, Paessens, GMS (2003)]

- new phase of broken ergodicity (despite 1-d nonconservative short range interactions)

Experiment: transport of molecular motors along microtubuli

[Nishinari, Okada, Schadschneider, Chowdhury (2005)]

(1) low density regime of kinesin
(2) Formation of comet-like accumulation of kinesin
at the end of microtubule:
(3) high density regime



Two-component DDS: two-species (A,B) exclusion process, two-lane models, bricklayer models, ...

New physics:

- Spontaneous symmetry breaking [Evans, Foster, Godreche, Mukamel (1995)]

  origin: amplification of fluctuations [Willmann, GMS, Grosskinsky (2005)]
  boundary blockage [Godreche et al. (1995); Willmann et al (2005)]

- Phase separation:
  1) Strong [Lahiri and Ramaswamy (1997), Arndt, Heinzel, Rittenberg (1998); Evans, Kafri, Koduvely, Mukamel (1998)]
  2) Soft (condensation transition) [Kafri, Levine, Mukamel, GMS, Willmann (2002)]

  origin: nucleation [Mahnke, Kaupusz, Harris (2005)], ← Poster Harris
  coarsening [Evans et al. (1998); Grosskinsky, GMS, Spohn (2003); Godreche (2003)]

- Infinite boundary reflection of shocks [Popkov (2002)]

  origin: unclear

Challenge: No coherent picture, not even hydrodynamic description (except strong phase separation)

4. Boundary-driven systems

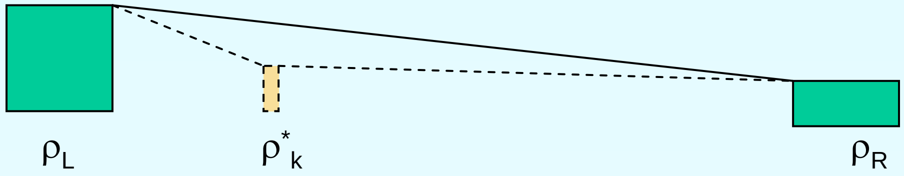
Absence of bulk force:

- macroscopic gradient current $j = - D(\rho)\, \rho'/L$
- no shocks
- example: symmetric exclusion process (p=q=D): $j_k = D\,(\rho_{k-1} - \rho_k)$

Large deviation function for NESS (SEP): Derrida, Lebowitz, Speer (2002)

$P[\rho_x = c(x)] \sim \exp(-L\, g[c(x)])$

- exact calculation using MPA
- Additivity principle → differential equation für g
- g nonlocal ⇔ long-range correlations [Spohn (1983)]



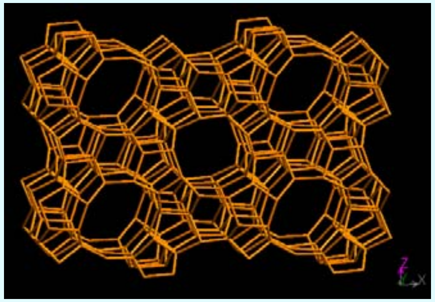
General macroscopic theory Bertini, De Sole, Gabrielli, Jona-Lasinio, Landim (2002)

- Additivity → g
- arbitrary dimension

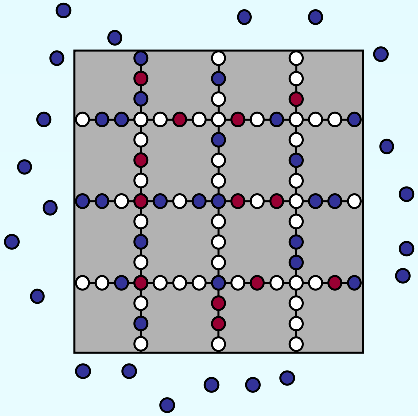
Molekulare Verkehrskontrolle in katalytischen Reaktionen:

Zeolithe: Ionentauscher, Adsorbenten, Katalyse

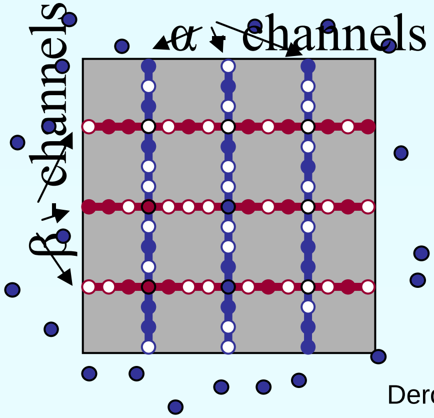
Korngröße 60-70 μm,
Porendurchmesser Benzolmolekül
Single-File-Diffusion
N=O(10^4) Kanäle pro Korn



Katalyse: Gegenseitige Behinderung von Reaktand und Produkt durch Single-File-Effekt



Molekulare Verkehrskontrolle: Verschiedene Diffusionspfade für Reaktand und Produkt



Deroune, Gabelica (1980)

→ stark diffusionslimitierte eff. Reaktivität

→ erhöhte eff. Reaktivität? (MTC-Effekt)

Kanalselektivität: Ja (MD-Simulationen) [Clarrk, Ye, Snurr (2000)]

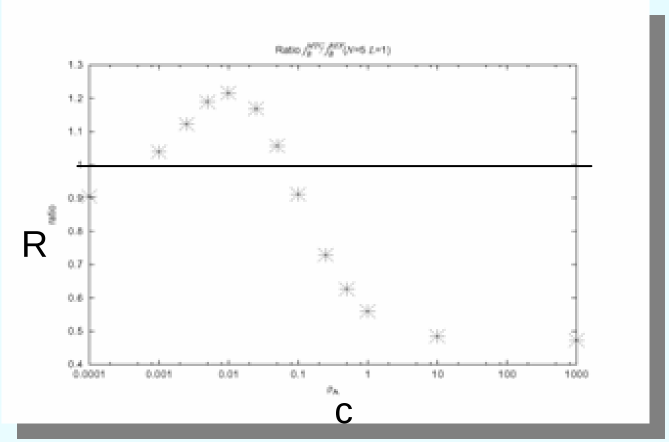
NBK-Modell: quadratische Anordnung von SEP mit katalytischen Kreuzungspunkten mit Rate c
[Neugebauer, Bräuer, Kärger (2000)]

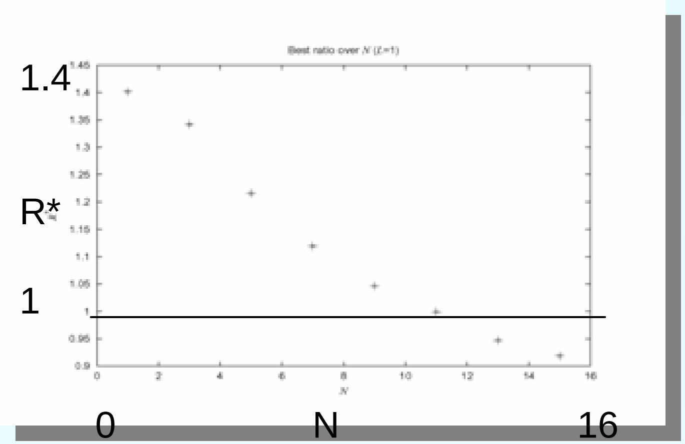
Resultate:

- MTC-Effekt für Kanallänge L > L* (Single-File-Diffusion: D ~ 1/L), aber j_{out} ~ $1/L^2$
- Sättigung (randinduzierter Phasenübergang) für c>>1 (Ansatz: effektive Dichten an Kreuzungspunkten)
- kleines L: R ~ 1/N (Sättigung), aber MTC-Effekt bei j_{out} ~ 1/L

Poster Brzank

- MTC-Effekt mit neuer Kanaltopologie





5. Conclusions

Results:

- Description of stochastic many-body dynamics in field-theoretic terms ➔ exact results
- microscopic derivation of macroscopic notions in DDS (collective velocity, shock stability, shock localization)
- shocks as collective one-particle excitations (bound states)
- phase diagram of open one-component DDS: shock motion, effective potential, saturation
- experimental evidence for boundary-induced phase transition in molecular motors
- large deviation theory for boundary-driven systems
- Two-component systems: amplification of fluctuations, blocking, coarsening
- molecular traffic control in structured binary pore systems

Open problems:

- detailed experimental verification of phase diagram of open DDS
- phase diagram and hydrodynamic description of two-component systems
- large deviation theory for bulk driven and n-component systems

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