## Paradoxical Noise-Effects far from Thermal Equilibrium

**Peter Reimann** Universität Bielefeld

- Ratchet Effects
- Negative Mobility in a Microfluidic Device
- Sorting Chiral Particles

## **Ratchet and Pawl**

#### [Smoluchowski 1912, Feynman 1963]



# **Ratchet and Pawl**

#### [Smoluchowski 1912, Feynman 1963]





$$m \ddot{x} = -\gamma \dot{x} - V'(x) + \xi(t)$$
  
$$m \rightarrow 0: \quad \gamma \dot{x} = -V'(x) + \xi(t)$$



It follows:

 $egin{array}{ll} \xi(t) \ {
m Gauss} \ , \ \langle \xi(t) 
angle = 0 \ , \ \langle \xi(t) \, \xi(s) 
angle = 2 \gamma k T \, \delta(t-s) \end{array}$ 



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#### **Theory of Fokker-Planck processes:**

$$\langle \dot{x} 
angle := \lim_{t \to \infty} rac{x(t)}{t} = \mathbf{0}$$
 (2nd law)



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#### **Theory of Fokker-Planck processes:**

$$\langle \dot{x} 
angle := \lim_{t o \infty} rac{x(t)}{t} = 0$$
 (2nd law)

Generalization:

$$\gamma \dot{x} = -V'(x) + F + \xi(t)$$
  
 $F < 0 \Rightarrow \langle \dot{x} \rangle < 0 \text{ for any } T > 0$ 



# **Temperature Ratchet**

$$\gamma \dot{x} = -V'(x) + \xi(t) + F$$
  
 $\langle \xi(t) \xi(s) \rangle = 2\gamma k T(t) \delta(t-s)$ 



dimensionless units:

 $kT_{max} = 3, kT_{min} = 0.5,$  $\tau = 5, \gamma = 1$ 



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### **Explanation**

 $(F = 0, kT_{min} \ll \Delta V, kT_{max} \gg \Delta V, \tau \text{ large})$ 



### **Explanation**

 $(F = 0, kT_{min} \ll \Delta V, kT_{max} \gg \Delta V, \tau \text{ large})$ 



- Particle pump
- Mechanism robust (provided  $\tau$  large)
- No contradiction to 2nd law
- x(t) and T(t) "loosely coupled"

### **Transport Direction**

 $\gamma\,\dot{x} = -V'(x) + \xi(t) \;\;, \;\;\; \langle \xi(t)\,\xi(s)
angle = 2\gamma kT(t)\,\delta(t-s)$ 

Consider  $\langle \dot{x} \rangle$  as a function of an <u>arbitrary</u> parameter  $\mu$  ( $\gamma$ ,  $\tau$ ,  $T_{min}$ ,...) and choose  $\mu_0$  <u>arbitrarily</u>.

There exists a V(x) with a current inversion at  $\mu_0$ 

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**Example:**  $\mu = \gamma$ ,  $\mu_0 = 0.7$ ,  $kT_{max} = 0.18$ ,  $kT_{min} = 0.02$ ,  $\tau = 0.02$ 



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#### as a quantum effect:

Keay et al., PRL 75, 4102 (1995)

#### as a classical effect:

Eichhorn, Reimann, Hänggi, PRL 88, 190601 (2002)



## **Theoretical Prediction**

[Eichhorn & Reimann, 2005]

 $(2 \,\mu\text{m} \text{ particle diameter}, U_0 = 30 \text{ V}, \tau = 25 \text{ s})$ 



### **Experiment versus Theory**



### **Physical Mechanism**



## **Physical Mechanism**



 $U_{DC} < 0$ ,  $U_{DC} + U_0 > 0$ 



 $U_{DC} < 0$ ,  $U_{DC} - U_0 < 0$ 

### **Theoretical Prediction**

 $(U_0 = 6 \text{ V}, \tau = 70 \text{ s})$ 

 $1.9\,\mu m$  particles: blue

2.8  $\mu$ m particles: red



## **Experiment versus Theory**

[Eichhorn, Regtmeier, Anselmetti, Reimann, Soft Matter  $\mathbf{6}$ , 1858 (2010)]

**1.9**  $\mu$ **m particles: blue** 

 $2.8\,\mu m$  particles: red



# A tunable microfluidic ratchet for particle sorting

[Bogunovich, Eichhorn, Regtmeier, Anselmetti, Reimann, submitted]



 $U(t) = U_{DC} + U_{AC} \sin(\omega t) \Rightarrow \text{velocity in } x\text{-direction }?$ 

### A tunable microfluidic ratchet for particle sorting

[Bogunovich, Eichhorn, Regtmeier, Anselmetti, Reimann, submitted]



### A tunable microfluidic ratchet for particle sorting

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Electrophoresis:  $\vec{F}(\vec{r}) = q \vec{E}(\vec{r})$ 

 $\vec{T}(\vec{z})$ 

Dielectrophoresis:  $F(\vec{r}) = \vec{\nabla} [\vec{p} \cdot \vec{E}(\vec{r})]$  $\vec{p} = \alpha \vec{E}(\vec{r})$ 

 $\vec{\nabla}$   $\vec{\Gamma}^2(\vec{z})$ 

## **Sorting Chiral Particles**

[Speer, Eichhorn, Reimann, PRL 105, 090602 (2010)]



### **Sorting Chiral Particles**

[Speer, Eichhorn, Reimann, PRL 105, 090602 (2010)]

static force  $\vec{A} = A \vec{e}_{\alpha}$  with  $\alpha = 45^{\circ}$ 

resulting average velocity  $\vec{v} = v \, \vec{e}_\vartheta$  :



### **Sorting Chiral Particles**

[Speer, Eichhorn, Reimann, PRL 105, 090602 (2010)]

time-periodic  $\vec{A}(t) = A(t) \vec{e}_{\alpha}$ ,  $\alpha = 45^{\circ}$ , A(t) alternating between 6 and -4



 $\vec{r}(t)$  for  $t \in [0, 100]$ , kT = 0.02, A(t) = 6 for 2 time units, A(t) = -4 for 4 time units

### **Other chiral objects**



## **First experimental steps**

[Experiment: Wegener, Regtmeier, Anselmetti. Theory: Fliedner, Reimann]













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Theory: A. Engel, P. R. Experiment: H.-W. Müller, A. Jung PRL **91**, 060602 (2003)

 $H_y(t) \propto \sin(\omega t) + \sin(2\omega t + \delta)$ 





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In the plane:

#### Supersymmetry

[P. R., PRL 86, 4992 (2001)]

#### **Granular Ratchet**

[v.d. Meer, P. R., v.d. Weele, Lohse, PRL 92, 184301 (2004); J. Stat. Mech. P07021 (2007)]



#### **Granular Ratchet**

[v.d. Meer, P. R., v.d. Weele, Lohse, PRL 92, 184301 (2004); J. Stat. Mech. P07021 (2007)]



#### **Negative Absolute Resistance in a Josephson Junction**

J. Nagel, D. Speer, T. Gaber, A. Sterck, R. Eichhorn, P. Reimann,

K. Ilin, M. Siegel, D. Koelle, R. Kleiner, Phys. Rev. Lett. 100, 217001 (2008)



#### **Directing Brownian Motion on a Periodic Surface**

D. Speer, R. Eichhorn, P. Reimann, Phys. Rev. Lett. 102, 124101 (2009)



#### **Molecular Motors**

[Huxley 1957, Vale and Oosawa 1990, ... ]



- x(t) : mech./geom. configuration (cyclic) or position
- V(x) periodic & asymmetric
- length scale  $\sim$  10nm  $\Rightarrow$  thermal noise relevant
- Chemical reaction cycle (ATP-hydrolysis)  $\Rightarrow$  heat production  $\Rightarrow$  local temperature changes T(t)

#### **Molecular Motors**

[Huxley 1957, Vale and Oosawa 1990, ...]





### **Physical Mechanism**



 $U_{DC} < 0, \quad U_{DC} + U_0 > 0$ 



 $U_{DC} < 0 \,, \quad U_{DC} - U_0 < 0$ 

Indispensable:

- "particle traps"
- fluctuations (diffusion)

#### **Physical Mechanism**



 $U_{DC} < 0$ ,  $U_{DC} - U_0 < 0$ 

Theoretical concept: Phys. Rev. Lett. 88, 190601 (2002) Experiment versus theory: Nature 436, 928 (2005)

#### **Single Particle Trajectories**

 $(U_0 = 6 \text{ V}, \tau = 70 \text{ s}, U_{DC} = 2 \text{ V})$ 



#### **Optimized Microstructure**

[Regtmeier, Grauwin, Eichhorn, Reimann, Anselmetti, Ros, J. Sep. Sci. 30, 1461 (2007)]



#### **Optimized Microstructure**

[Regtmeier, Grauwin, Eichhorn, Reimann, Anselmetti, Ros, J. Sep. Sci. 30, 1461 (2007)]



$$\vec{F}(\vec{r}) = q_{\text{eff}} \vec{E}(\vec{r}) , \quad \vec{E}(\vec{r}) = -\nabla \phi(\vec{r}) = ?$$

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fluid (buffer): conductor,  $\vec{j}(\vec{r}) \parallel \vec{E}(\vec{r})$ , e.g.  $\vec{j} = \sigma \vec{E}$ solid (PDMS): insulator,  $\vec{j}(\vec{r}) = \vec{0}$ 

$$\dot{
ho} + 
abla ec{j} = 0$$
 (charge conservation) ,  $\dot{
ho} = 0$  (steady state)  
 $\Rightarrow \quad 
abla ec{j}(ec{r}) = 0 \quad \Rightarrow \quad ec{j}_{\perp}(ec{r}) = ec{0}$  for  $ec{r}$  at fluid-solid border

$$\vec{F}(\vec{r}) = q_{\text{eff}} \vec{E}(\vec{r}) , \quad \vec{E}(\vec{r}) = -\nabla \phi(\vec{r}) = ?$$

fluid (buffer): conductor,  $\vec{j}(\vec{r}) \parallel \vec{E}(\vec{r})$ , e.g.  $\vec{j} = \sigma \vec{E}$ solid (PDMS): insulator,  $\vec{j}(\vec{r}) = \vec{0}$ 

$$\dot{\rho} + \nabla \vec{j} = 0$$
 (charge conservation) ,  $\dot{\rho} = 0$  (steady state)

$$\Rightarrow \quad 
abla ec j(ec r) = 0 \quad \Rightarrow \quad ec j_{\perp}(ec r) = ec 0 \quad ext{for} \quad ec r \quad ext{at fluid-solid border}$$

$$\Rightarrow \qquad \vec{E}_{\perp}(\vec{r}) = \vec{n}(\vec{r}) \cdot \nabla \phi(\vec{r}) = \vec{0} \text{ at border } (\text{Neumann b.c.})$$

#### $\Rightarrow$ no particle trap at border possible

Earnshaw's Theorem: no trap inside fluid ( $\nabla \vec{E} = \rho/\epsilon = 0$ )

**Electrical Field** 

 $\Delta \phi(\vec{r}) = 0$  with mixed boundary conditions  $\Rightarrow \vec{E}(\vec{r}) = - \nabla \phi(\vec{r})$ 



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z-direction trivial

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z-direction trivial

Central "unit cell" periodically continued

## **Electroosmosis**



$$ec{v}(ec{r}) = \lambda \, ec{E}(ec{r}) \qquad \qquad \lambda = -rac{\epsilon \, \zeta}{
u}$$



$$\vec{v}(\vec{r}) = \lambda \vec{E}(\vec{r}), \ \lambda = -\frac{\epsilon \zeta}{\nu}$$

#### Electrophoresis



(no form factor, Smoluchowski 1903)





$$v(r) = \lambda E(r), \ \lambda = -\frac{c\varsigma}{\nu}$$

 $\Rightarrow$  net particle velocity

$$ec{v}_0(ec{r}) = ( ilde{\lambda} + \lambda) \, ec{E}(ec{r}) = rac{\epsilon \, ( ilde{\zeta} - \zeta)}{
u} \, ec{E}(ec{r})$$

## Electroosmosis $\vec{E}$ $\vec{E}$





 $\vec{v}(\vec{r}) = \lambda \vec{E}(\vec{r}), \ \lambda = -\frac{\epsilon \zeta}{\kappa}$ 

 $\Rightarrow$  net particle velocity

$$ec{v}_0(ec{r}) = ( ilde{\lambda} + \lambda) \, ec{E}(ec{r}) = rac{\epsilon \, ( ilde{\zeta} - \zeta)}{
u} \, ec{E}(ec{r})$$

 $\Rightarrow$  general dynamics

$$m\ddot{\vec{r}}(t) = \vec{F}(\vec{r}(t)) - \eta \left[\dot{\vec{r}}(t) - \vec{v}_0(\vec{r}(t))\right]$$

m particle mass: negligible (overdamped)

 $ec{v}$ 

- $\eta~$  viscous friction coefficient
- $\vec{F}(\vec{r})$  non-electric forces: hard walls  $-\nabla V(\vec{r})$ , thermal noise  $\vec{\xi}(t)$





$$\Rightarrow$$
 net particle velocity

$$ec{v}_0(ec{r}) = ( ilde{\lambda} + \lambda) \, ec{E}(ec{r}) = rac{\epsilon \, ( ilde{\zeta} - \zeta)}{
u} \, ec{E}(ec{r})$$

 $\Rightarrow$  general dynamics

$$m\,\ddot{ec r}(t)=ec F(ec r(t))-\eta\left[\dot{ec r}(t)-ec v_0(ec r(t))
ight]$$

$$\Rightarrow \quad \eta \, \dot{ec r}(t) = - 
abla V(ec r(t)) \, + \, ec ec t(t) \, + \, q_{ ext{eff}} \, ec E(ec r(t)) \qquad q_{ ext{eff}} := rac{\eta \, \epsilon \, (ec \zeta - \zeta)}{
u}$$

#### **Quantitative Theory**

# $egin{aligned} &\eta\,\dot{ec r}(t) = abla V(ec r(t))\,+\,ec \xi(t)\,+\,q_{ ext{eff}}\,ec E(ec r(t))\,\,,\quad q_{ ext{eff}} := \eta\,\epsilon\,(ec \zeta-\zeta)/ u \ &\langle\xi_i(t)\,\xi_j(s) angle = 2\eta kT\,\delta(t\!-\!s)\,\delta_{ij}\,\,,\quad T=293\,ec k \end{aligned}$

#### **Quantitative Theory**



 $ec{E}(ec{r}) \; \mapsto \; ec{E}(ec{r},t) := ec{E}_{*}(ec{r}) \, [U_{AC}(t) + U_{DC}]/U_{*}$  (quasi-static)



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#### $q_{ m eff}/U_{*}$ and $\eta$ unknown

Theoretical determination practically impossible

Experimental determination by measuring free mobility and diffusion  $(D = kT/\eta)$ 

## **Kipp-Ratsche: Symmetrien**

$$\gamma \dot{x} = \xi(t) - V'(x) + F(t) V(x+L) = V(x), F(t+\tau) = F(t), \int_{0}^{\tau} F(t) dt = 0$$

#### **Kipp-Ratsche: Symmetrien**



#### **Quantum-Ratchet**



[Figure: M. Brooks, New Scientist 2222, 28 (2000)]

### **Quantum-Ratchet**



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high  $T \Rightarrow \langle \dot{x} 
angle \! > \! 0$ 

### **Quantum-Ratchet**



[Figure: M. Brooks, New Scientist 2222, 28 (2000)]

high  $T \Rightarrow \langle \dot{x} 
angle \! > \! 0$  low  $T \Rightarrow \langle \dot{x} 
angle \! < \! 0$
## **Quantum-Ratchet**



Theory: Reimann et al., Phys. Rev. Lett. **79**, 10 (1997) Experiment: Linke et al., Science **286**, 2314 (1999)

## 1. Beispiel: Quanten-Ratsche



Theorie: Reimann et al., Phys. Rev. Lett. **79**, 10 (1997) Experiment: Linke et al., Science **286**, 2314 (1999)

### Einzelmolekül-Kraftspektroskopie

Experimente: R. Ros, D. Anselmetti (Bielefeld); R. Merkel (Jülich) Theorie: M. Raible, P. Reimann



z.B. expG-Protein und expE-DNA

 $f\simeq\kappa\,s$  ,  $\kappa\simeq$  3 pN/nm

Zentrale Grösse: Verteilung der Abreisskräfte

## Verteilung der Abreisskräfte

(*expG*-Protein und expE-DNA)



Verteilung der Abreisskräfte ist abhängig von Ziehgeschwindigkeit v: Interpretation ?

#### Standard-Theorie von Evans & Ritchie

[Biophys. J. 72, 1541 (1997); ca. 400 mal zitiert]

 $\dot{p}_v(f(t)) = -r(f(t))\,p_v(f(t))$ 

 $p_v(f)$  Überlebensw'keit, r(f) Dissoziationsrate, f(t) Kraft

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#### Theoretische Voraussage:

[Raible, Evstigneev, Reimann]

 $\Rightarrow -v \ln p_v(f)$  unabhängig von v[Phys. Rev. E 68, 045103(R) (2003)]

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#### Vergleich mit Messdaten:

[von Ros & Anselmetti sowie Merkel]

- ⇒ Standard-Theorie falsch !
- [J. Biotechnology 112, 13 (2004)]



#### **Neue Theorie**

[Raible & Reimann, submitted]

Heterogenität der Dissoziationsrate

 $r(f)\simeq r_o \; e^{lpha f} \;, \;\;\; lpha$  Zufallsvariable

 $\langle lpha 
angle \simeq 0.13 \ {
m pN}^{-1}, \ \sigma \simeq 0.07 \ {
m pN}^{-1}, \ r_o \simeq 0.0034 \ {
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#### Verteilung der Abreisskräfte

Experimentelle Messdaten: schwarze Histogramme Standard-Theorie [Evans & Ritchie]: blaue Kurven Neue Theorie [Raible & Reimann]: rote Kurven



# **Atomic Friction**



# **Atomic Friction**



## **Atomic Friction**



Quantity of main interest:

$$ar{F} := \lim_{t o \infty} rac{1}{t} \int \limits_0^t dt' \, F(t')$$

#### **Velocity Dependence of Atomic Friction**

Experiment: Riedo et al., PRL 91, 084502 (2003)

(mica surface, L = 0.52 nm, T = 293 K,  $F_N = 12$  nN)



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(mica surface, L = 0.52 nm, T = 293 K,  $F_N = 12$  nN)



**Theory:** P. R. & Evstigneev, PRL 93, 230802 (2004)

$$v(f) \simeq rac{L\,k\,T\,[1-e^{-Lf/kT}]}{\gamma \int_0^L dx \int_x^{x+L} dy \,e^{[U(x)-U(y)+(x-y)f]/kT}} , \quad \bar{F}(f) \simeq f - \gamma v(f)$$

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• stick-slip-amplitude  $\hat{=}$  dissipation  $\Rightarrow \bar{F}(v)$  decreasing

• slips: thermally activated transitions

 $\Rightarrow \bar{F}(v)$  increasing



- stick-slip-amplitude  $\hat{=}$  dissipation  $\Rightarrow \bar{F}(v) \underline{\text{decreasing}}$
- slips: thermally activated transitions  $\Rightarrow \bar{F}(v)$  increasing

together  $\overline{F}(v)$  non-monotonic





 $\dot{p}_v(F(t)) = -r(F(t)) p_v(F(t))$ 

F(t) instantaneous force, r(F) "slip rate",  $p_v(F)$  "stick probability"



 $\dot{p}_v(F(t)) = -r(F(t)) \, p_v(F(t))$ 

F(t) instantaneous force, r(F) "slip rate",  $p_v(F)$  "stick probability"

 $\Rightarrow$   $-v\ln p_v(F)$  independent of v

[Evstigneev, Schirmeisen, Jansen, Fuchs, P. R., PRL 97, 240601 (2006)]



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together  $\overline{F}(v)$  **non-monotonic** 



[P.R. & Evstigneev, New J. Phys. 7, 25 (2005)]

## **Feature Articles**



• D. Bradley, **IOP press release**, Feb. 2005 Scientists close in on "superbrakes" for cars

• L. Hutson, Material World (London), March issue 2005 Breaking news

• P. Grumberg, **Science & Vie (Paris)**, March issue 2005 Un effet inattendu du frottment pourrait améliorer le freinage