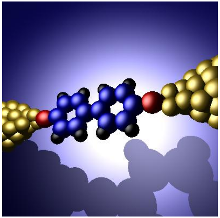
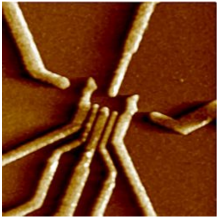
Driven transport through nanoscale conductors

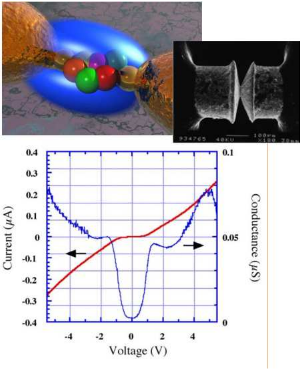
Sigmund Kohler

Universität Augsburg

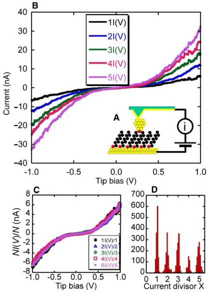




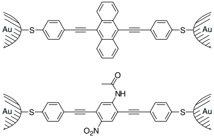
single-molecule conduction



M. A. Reed *et al.*, Science **278**, 252 (1997)



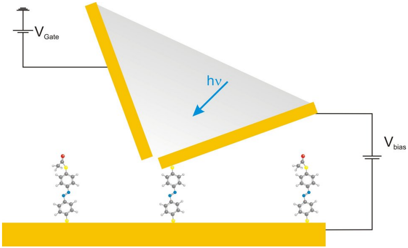
X. D. Cui *et al.*, Science **294**, 571 (2001)



J. Reichert *et al.*, Phys. Rev. Lett. **88**, 176804 (2002)

excitations of molecules

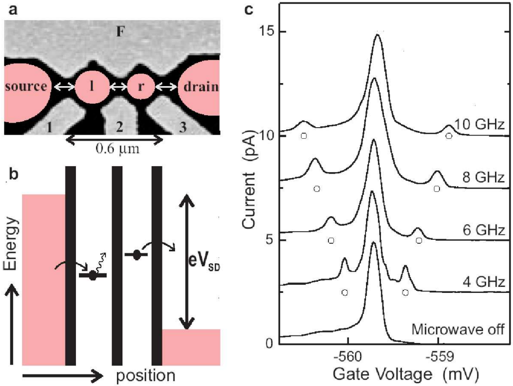
- direct exposure to light
  problem: affects contacts
- solution: coated SNOM tip
  (*scanning near-field optical microscope*)



© J. Reichert (TU Munich)

- focussing on molecule
- TERS (*tip-enhanced Raman spectroscopy*):
  local field enhancement by several orders of magnitude

“artificial molecules”: coupled quantum dots

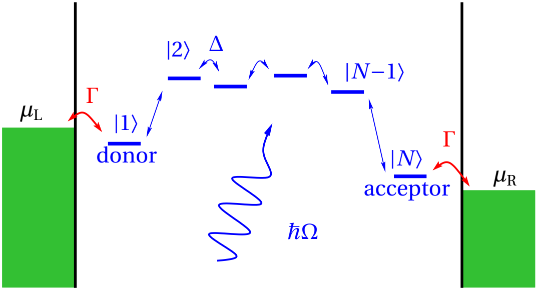


T. H. Oosterkamp *et al.*, Nature **395**, 873 (1998)

interesting questions

- conduction under laser excitation
- ratchet and pump effects
- current control by laser field
- noise properties
- Coulomb blockade
- ...

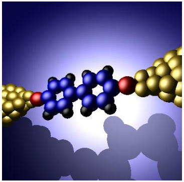
tight-binding model



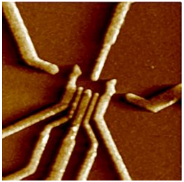
- hopping matrix elements Δ
  [molecule: Hückel model of a "molecular bridge"]
- metallic contacts: ideal Fermi gases with chem. potential μ
- effective coupling to metal contacts: Γ
- laser field: $H_{mol} \longrightarrow H_{mol}(t)$, dipole coupling

Driven transport through nanoscale conductors

- **Floquet transport theory**



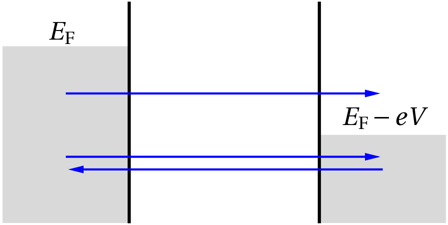
- **DC current from AC driving**
  - ratchet effect
  - nonadiabatic electron pump
  - pumping heat



- **Coulomb blockade effects**

static case: scattering formula

- Landauer (1957): „conductance is transmission“



- current $$I = \frac{e}{2\pi\hbar} \int dE\, T(E,V)\left[f(E+eV) - f(E)\right]$$
- transmission of an electron with energy E

$$T(E,V) = \Gamma_L \Gamma_R |\langle 1|G(E,V)|N\rangle|^2$$

Fischer, Lee, PRB’81; Meir, Wingreen, PRL’92

static case: current noise

- zero-frequency noise / noise power:

  static component of the current-current correlation function

$$\bar{S} = S(\omega = 0) = \int_{-\infty}^{+\infty} \mathrm{d}\tau \, \langle \Delta I(t) \, \Delta I(t+\tau) \rangle$$

$$\bar{S} = \frac{e^2}{2\pi\hbar} \int \mathrm{d}E \; T(E) \Big\{ [1 - T(E)] \, [f(E+eV) - f(E)]^2 + f(E+eV)[1 - f(E+eV)] + f(E)[1 - f(E)] \Big\}$$

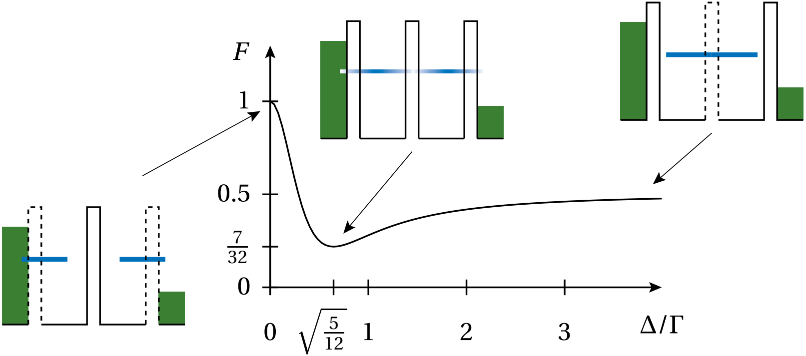
  - shot noise (remains for $k_B T = 0$)
  - equilibrium noise (remains for $eV = 0$)

Büttiker, PRB'92

depends only on transmission probability $T(E)$

current noise: Fano factor

- relative noise strength: Fano factor $F = S/eI$
- example: transport double quantum dot



single barrier / point contact: $F \approx 1$ (Poisson process)

double barrier: $F \approx \frac{1}{2}$

current noise: Fano Factor

- Landauer: „noise is the signal“

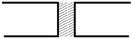
ohmic resistor

$U = RI$

thermal noise

$\longrightarrow$ temperature dependent

tunnel contact



$U = R_{\mathrm{T}} I$

shot noise $S = qI$

$\longrightarrow$ $F = q/e$ (size of charge carrier)

- Cooper pair tunneling: $F = 2$
- fractional quantum Hall effect: $F = \frac{1}{3}$

AC driven transport: Tien-Gordon theory

voltage ⟶

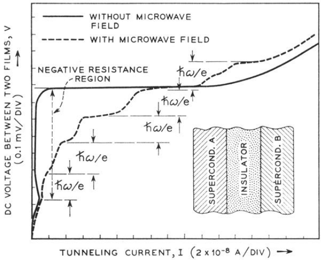


FIG. 1. Bias voltage vs tunneling current of a superconducting Al-Al_2O_3-In diode as measured by Dayem and Martin with and without the microwave field. $\hbar\omega/e = 0.16$ mV.

current ⟶

Josephson contact in microwaves

- current-voltage characteristics exhibits steps of size $\Delta V = \hbar\Omega/e$

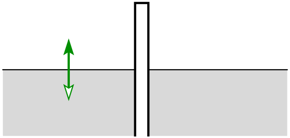
  Dayem & Martin, PRL 1962
- model:
  - tunnel barrier
  - oscillating chemical potential in one superconductor

  Tien & Gordon, Phys. Rev. 1963

Tien-Gordon theory

- microwaves induce ac bias voltage:

$$V_0 \longrightarrow V_0 + V_{\rm ac}\cos(\Omega t)$$



- time-dependent energy shift by $eV_{\rm ac}\cos(\Omega t)$

$$\exp\left(-\frac{\rm i}{\hbar}Et\right) \longrightarrow \exp\left(-\frac{\rm i}{\hbar}Et - {\rm i}\frac{eV_{\rm ac}}{\hbar\Omega}\sin(\Omega t)\right)$$

$$= \sum_{k=-\infty}^{\infty} J_k\left(\frac{eV_{\rm ac}}{\hbar\Omega}\right)\exp\left(-\frac{\rm i}{\hbar}(E + k\hbar\Omega)t\right)$$

  - sidebands occupied with probability $J_k^2(\ldots)$
  - energy $k\hbar\Omega$ corresponds to additonal DC bias voltage $k\hbar\Omega/e$

$$I(V_0, V_{\rm ac}) = \sum_{k=-\infty}^{\infty} J_k^2\left(\frac{eV_{\rm ac}}{\hbar\Omega}\right) I_0(V_0 + k\hbar\Omega/e)$$

DC conductivity determines the current !
…and also the shot noise

Tucker, Feldmann, Rev. Mod. Phys. 1985

- derivation rather heuristic
- ? rigorous derivation
- ? when is Tien-Gordon theory applicable

driven systems

- problem: $U(t,t') = \overleftarrow{T} \exp\left(-\frac{\mathrm{i}}{\hbar}\int_{t'}^{t} \mathrm{d}t''\, H(t'')\right)$

  $\overleftarrow{T}$: time-ordering operator
- periodic time-dependence:
  „Bloch theory in time“ (Floquet 1883)
- periodic time-dependence:
  Floquet theorem: time-periodic Schrödinger equation has complete solution of the form

$$|\psi_\alpha(t)\rangle = \mathrm{e}^{-\mathrm{i}\epsilon_\alpha t/\hbar}|\phi_\alpha(t)\rangle, \text{ where } |\phi_\alpha(t)\rangle = |\phi_\alpha(t+\mathcal{T})\rangle$$

- quasienergies ϵ_α, Brillouin zone structure
  Floquet states $|\phi_\alpha(t)\rangle = \sum_k \mathrm{e}^{-\mathrm{i}k\Omega t}|\phi_{\alpha,k}\rangle$

non-linear response

transport and driving: computation of the Green function and current formula for time-dependent situation

- Floquet equation
  with self-energy $\Sigma = |1\rangle \frac{\Gamma_L}{2} \langle 1| + |N\rangle \frac{\Gamma_R}{2} \langle N|$

$$\left(H(t) - \mathrm{i}\Sigma - \mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\right)|\varphi_\alpha(t)\rangle = (\epsilon_\alpha - \mathrm{i}\hbar\gamma_\alpha)|\varphi_\alpha(t)\rangle$$

- propagator in the presence of the contacts

$$G(t, t-\tau) = \sum_{k=-\infty}^{\infty} \mathrm{e}^{\mathrm{i}k\Omega t} \int \mathrm{d}\epsilon\, \mathrm{e}^{-\mathrm{i}\epsilon\tau} \underbrace{\sum_{\alpha,k'} \frac{|\varphi_{\alpha,k+k'}\rangle\langle\varphi_{\alpha,k'}|}{\epsilon - (\epsilon_\alpha + k'\Omega - \mathrm{i}\hbar\gamma_\alpha)}}_{G^{(k)}(\epsilon)}$$

propagation under absorption/emission of $|k|$ photons

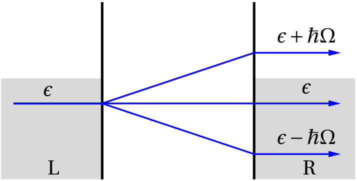
Floquet transport theory: current

- dc current [note: no blocking factors $(1 - f_\ell)$]

$$I = \overline{e\langle \dot{N}_\mathrm{L}\rangle} = \ldots = \frac{e}{h} \sum_{k=-\infty}^{\infty} \int d\epsilon \left\{ T_\mathrm{LR}^{(k)}(\epsilon) f_\mathrm{R}(\epsilon) - T_\mathrm{RL}^{(k)}(\epsilon) f_\mathrm{L}(\epsilon) \right\}$$

- transmission under absorption of k photons

$$T_\mathrm{LR}^{(k)}(\epsilon) = \Gamma_\mathrm{L}\Gamma_\mathrm{R} |\langle 1|G^{(k)}(\epsilon)|N\rangle|^2 \neq T_\mathrm{RL}^{(\pm k)}(\epsilon \pm k\hbar\Omega)$$



Camalet, Lehmann, SK, Hänggi, PRL **90**, 210602 (2003) time-averaged zero-frequency noise

$$\bar{S} = \frac{1}{\mathcal{T}} \int_0^{\mathcal{T}} \mathrm{d}t \int_{-\infty}^{+\infty} \mathrm{d}\tau \, \langle \Delta I(t) \, \Delta I(t+\tau) \rangle$$

$$= \frac{e^2}{h} \sum_k \int \mathrm{d}\epsilon \Big\{ \Gamma_\mathrm{R}^2 \Big| \sum_{k'} \Gamma_\mathrm{L}(\epsilon_{k'}) \, G_{1N}^{(k'-k)}(\epsilon_k) \big[G_{1N}^{(k')}(\epsilon) \big]^* \Big|^2 f_\mathrm{R}(\epsilon) \bar{f}_\mathrm{R}(\epsilon_k)$$

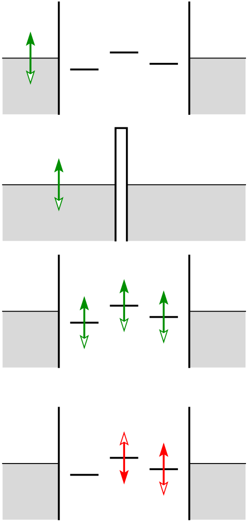
$$+ \Gamma_\mathrm{R}\Gamma_\mathrm{L} \Big| \sum_{k'} \Gamma_\mathrm{L} G_{1N}^{(k'-k)}(\epsilon_k) \big[G_{11}^{(k')}(\epsilon) \big]^* - \mathrm{i} G_{1N}^{(-k)}(\epsilon_k) \Big|^2 f_\mathrm{L}(\epsilon) \bar{f}_\mathrm{R}(\epsilon_k)$$

$$+ \text{same terms with the replacement } (L,1) \leftrightarrow (R,N) \Big\}$$

where $\epsilon_k = \epsilon + k\hbar\Omega$

depends on transmission amplitudes $G_{1N}^{(k)}$

when is Tien-Gordon theory applicable ?



applicable for

- AC bias voltage
- tunnel barriers
  (studied by Tien & Gordon)
- uniform AC gate voltage

but not for

- non-uniform gating or phase lag
- dipole force

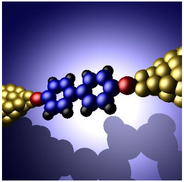
Camalet, SK, Hänggi, PRB **70**, 155326 (2004)

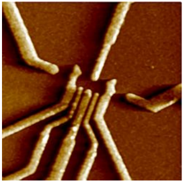
Driven transport through nanoscale conductors

- Floquet transport theory

- **DC current from AC driving**
  - ratchet effect
  - nonadiabatic electron pump
  - pumping heat

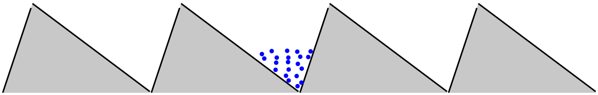
- Coulomb blockade effects





coherent quantum ratchet: motivation

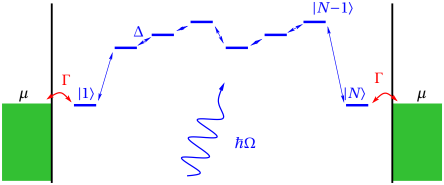
- classical Brownian motion
  in a periodic but asymmetric potential



  - despite asymmetry: zero current in equilibrium
  - asymmetry plus driving $\longrightarrow$ directed transport

- here: coherent quantum dynamics, non-adiabatic driving

coherent quantum ratchet



- no transport voltage

$$\mu_L = \mu_R$$

- finite periodic system consisting of N_g asymmetric groups

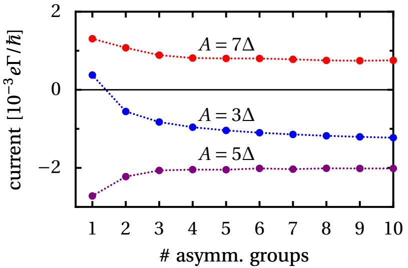
$$H_{nn'}(t) = -\Delta(\delta_{n,n'+1} + \delta_{n+1,n'}) + \Big(E_n + Ax_n \cos(\Omega t)\Big)\delta_{nn'}$$

- length dependence?
- coherent vs. incoherent quantum transport?

Lehmann, SK, Hänggi, Nitzan, PRL **88**, 228305 (2002)

coherent quantum ratchet: length dependence

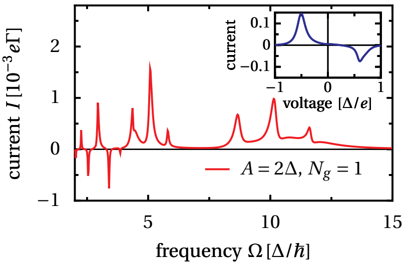
dc current *vs.* length



$\Delta = 1$, $\hbar\Omega = 3$, $\Gamma = 0.1$, $kT = 0.25$, $\mu = 0$, $E_B = 10$, $E_S = 1$

- current converges to non-zero value

coherent quantum ratchet: resonances



$\Delta = 1,\ \mu_L = \mu_R = 0,\ \Gamma = 0.1,\ E_D = E_A = 0, E_B = 10,\ E_S = 1$

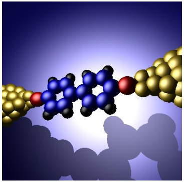
- ratchet current exhibits resonances $\longrightarrow$ coherent transport
- e.g. molecule: $A = e\mathscr{E} d_{\text{site}}$. $d_{\text{site}} \approx 1\,\text{nm}$, $\Delta = 0.1\text{eV}$
  $\Longrightarrow$ electric field strength $\mathscr{E} = 10^6\,\text{V/cm}$

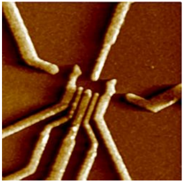
Driven transport through nanoscale conductors

- Floquet transport theory

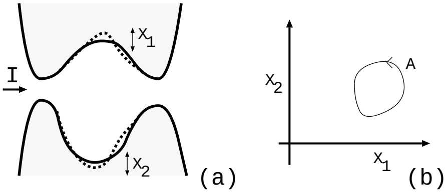
- **DC current from AC driving**
  - ratchet effect
  - nonadiabatic electron pump
  - pumping heat

- Coulomb blockade effects





adiabatic pumping:

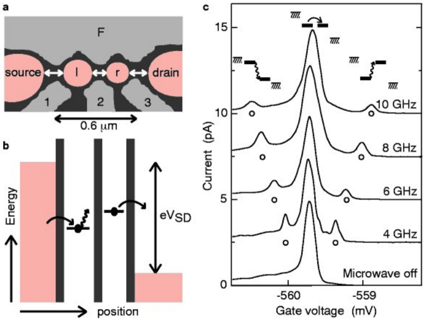


P. Brouwer, PRB **58**, 10135 (1998)

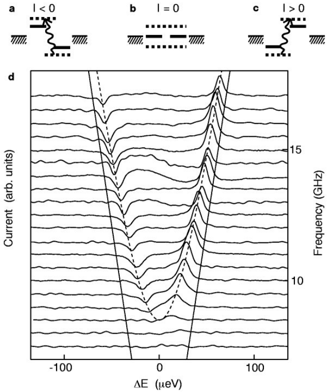
- adiabatic pump current:
  - time-reversal summetry: $I = 0$
  - $I \propto$ frequency
- pumping is more effective beyond the adiabatic limit

pumping with coupled quantum dots

Photon-assisted tunneling



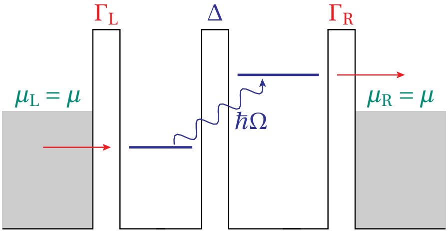
Pumping



T. H. Oosterkamp *et al.*, Nature **395**, 873 (1998)

- current maximum at resonance $\hbar\Omega = \sqrt{(\epsilon_L - \epsilon_R)^2 + \Delta^2}$

non-adiabatic electron pumping



- zero voltage: $\mu_L = \mu_R = \mu$
- coupling to microwaves:

$$H(t) \sim x \cos(\Omega t)$$

- ? behaviour close to resonances
- ? current noise
- ? pumping and time-reversal symmetry

Strass, Hänggi, SK, PRL **95**, 130601 (2005)

rotating-wave approximation at first resonance

- double dot

$$H(t) = -\frac{\Delta}{2}(c_1^\dagger c_2 + c_2^\dagger c_1) + \frac{1}{2}\Big(\underbrace{\delta + \hbar\Omega}_{\text{internal bias}} + A\cos(\Omega t)\Big)(n_1 - n_2)$$

1. interaction picture w.r.t. $H_0(t) \longrightarrow \widetilde{H}(t) = \widetilde{H}(t + 2\pi/\Omega) \ll \hbar\Omega$
2. time-scale separation: replace $\widetilde{H}(t)$ by its time average

$\longrightarrow$ effective static Hamiltonian

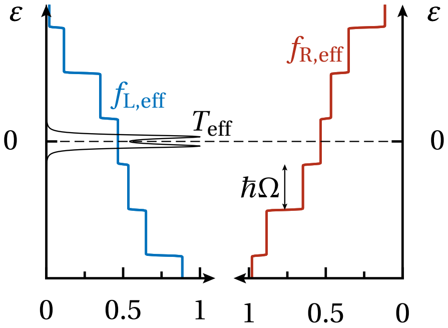
$$H_{\text{eff}} = -\frac{\Delta_{\text{eff}}}{2}(c_1^\dagger c_2 + c_2^\dagger c_1) + \frac{\delta}{2}(n_1 - n_2)$$

renormalized tunnel matrix element $\Delta \longrightarrow \Delta_{\text{eff}} = J_1(A/\hbar\Omega)\Delta$

- leads: t-average of Green's function $g^< = \frac{\mathrm{i}}{\hbar}\langle c_q^\dagger(t-\tau)c_q(t)\rangle$
effective electron distribution

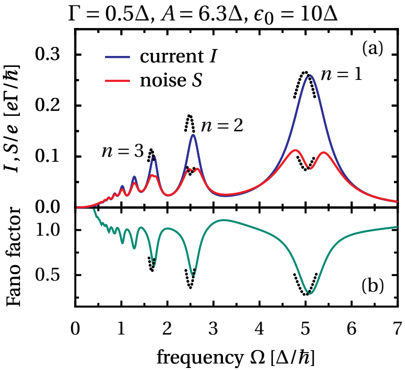
$$f_{\text{eff}}(\epsilon) = \sum_k J_k{}^2(A/2\hbar\Omega)\, f(\epsilon + (k \pm {}^1/_2)\hbar\Omega)$$

- effective static problem



- "voltage": $f_{\mathrm{L,eff}}(0) - f_{\mathrm{R,eff}}(0) = J_0^2(A/2\hbar\Omega)$ (as in Tien-Gordon theory!)
- total transmission at $\epsilon = 0$ determined by
  - inter-well coupling $\Delta_{\mathrm{eff}} = J_1(A/\hbar\Omega)\Delta$ (beyond Tien-Gordon!)
  - dot-lead coupling Γ

non-adiabatic electron pumping



- current maximum and noise minimum
- Fano factor $F = S/eI$

  noise strength considerably below shot noise level $F = 1$
- at n-photon resonance
  - intra-well coupling

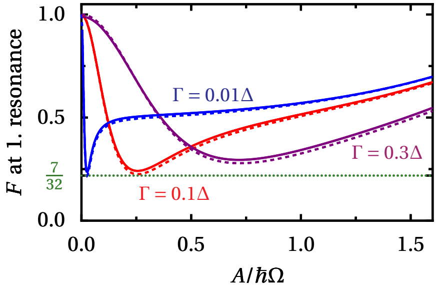
    $$\Delta_{\mathrm{eff}} = J_n(A/\hbar\Omega)$$

  - "voltage"

    $$f_{\mathrm{L,eff}}(0) - f_{\mathrm{R,eff}}(0) = J_0^2(A/2\hbar\Omega)$$

optimizing the pump

- goal: large current and low noise



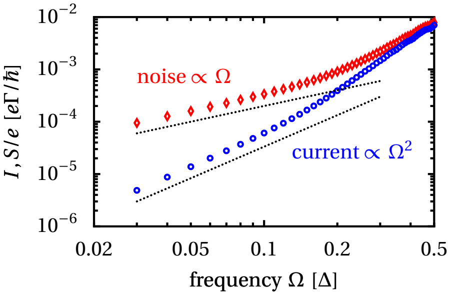
double dot,
wide-band limit: $F_{\min} = \frac{7}{32}$

- ideal conditions:
  - large bias ϵ_0
  - resonant driving $\hbar\Omega \approx \epsilon_0$
  - weak wire-lead coupling $\Gamma \lesssim 0.1\Delta$
- typical parameters: $\Delta = 10\,\mu\text{eV}$, $\Omega = 2\pi \times 15\,\text{GHz}$

$$\Longrightarrow \boxed{I \approx 40\,\text{pA} \text{ with } F \approx 0.23}$$

- adiabatic pumps:
  - cyclic evolution in parameter space
  - current determined by enclosed area $\longrightarrow$ $I_{\text{adiabatic}} \propto \Omega$
- here:
  - one parameter, area = 0 $\longrightarrow$ $I_{\text{adiabatic}} = 0$
  - $I \propto \Omega^2$?

current & noise in the adiabatic limit?



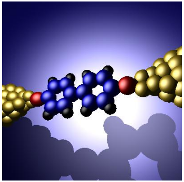
current	$\propto \Omega^2$ (as expected)
noise	$\propto \Omega$
Fano factor	$\propto \Omega^{-1}$

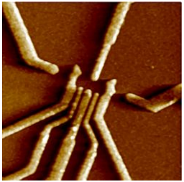
Driven transport through nanoscale conductors

- Floquet transport theory

- **DC current from AC driving**
  - ratchet effect
  - nonadiabatic electron pump
  - pumping heat

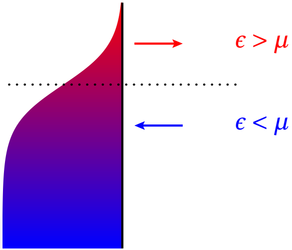
- Coulomb blockade effects



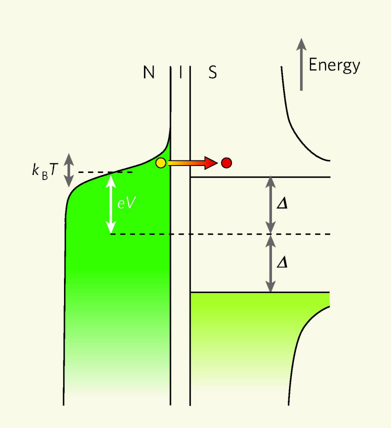


Can one extract heat/energy from the lead ?

- "heat":
  energy w.r.t. ground state
- "cooling":
  - remove electrons with $\epsilon > \mu$
  - fill holes with $\epsilon < \mu$
  - ensure $I = 0$
    to avoid charging of the leads

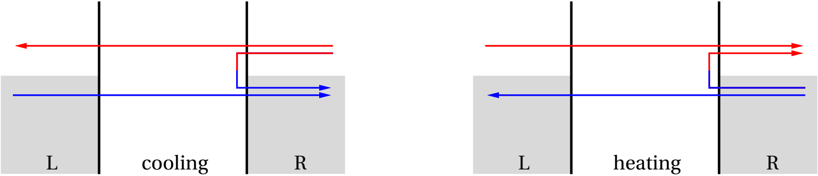


NIS interface



J. P. Pekola, Nature **435**, 889 (2005) heat balance for right lead:

$$\frac{d}{dt}E_R = \sum_q (\epsilon_q - \mu_R)\frac{dN_{R,q}}{dt} = \ldots = \frac{1}{h}\sum_{k=-\infty}^{\infty}\int d\varepsilon \Big[(\mu_R - \varepsilon)\, T_{LR}^{(k)}(\varepsilon) f_R(\varepsilon) + k\hbar\Omega\, T_{RR}^{(k)}(\varepsilon) f_R(\epsilon) + (\varepsilon + k\hbar\Omega - \mu_R)\, T_{RL}^{(k)}(\varepsilon) f_L(\varepsilon)\Big]$$



Rey, Strass, SK, Hänggi, Sols, PRB **76**, 085337 (2007)

inelastic backscattering

- time-reversal symmetry: $T_{\mathrm{RR}}^{(k)}(\epsilon) = T_{\mathrm{RR}}^{(-k)}(\epsilon + k\hbar\Omega)$

- thermal occupation $f(\epsilon)$ monotonously decaying

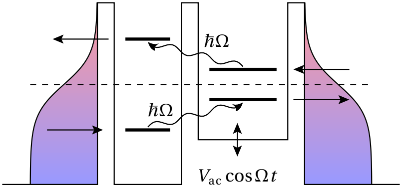
➔ inelastic backscattering always leads to heating

$$\frac{\mathrm{d}E_{\mathrm{R}}}{\mathrm{d}t} < 0 \text{ nevertheless possible ?}$$

heat pumping

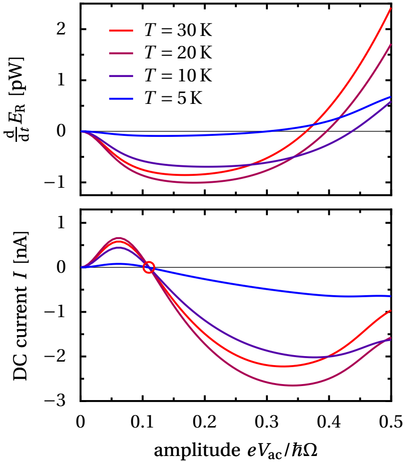
- GaAs heterostructure

  size ~ 10 nm, $\hbar\Omega = 2\,\text{meV}$, amplitude $V_{\text{ac}} \sim 100\mu\text{V}$



- dominant channels: $\Delta E = \pm\hbar\Omega$
- numerical computation of the transmission:
  Floquet scattering theory & transfer matrices

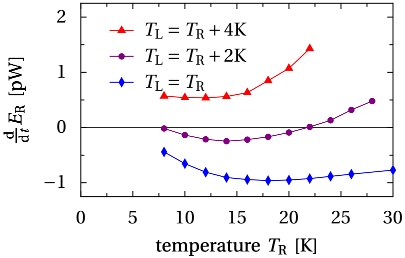
heat pumping



- $\frac{d}{dt}E_R < 0$, cooling!
- $I = 0$ possible
- cooling power: ~ pW

Rey, Strass, SK, Hänggi, Sols, PRB **76**, 085337 (2007)

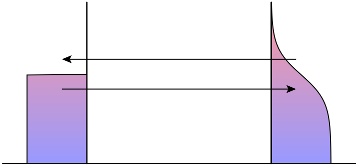
temperature dependence



for vanishing current:

- heat pumping:
  from cold to warm
- operating regime:
  - $T \gtrsim 10\,K$
  - $\Delta T \lesssim 1\,K$

efficiency



optimal cooling:

- contact with lead at zero temperature
- fully transparent 1d channel: $T(\epsilon) \equiv 1$

$$\frac{\mathrm{d}E_\mathrm{R}}{\mathrm{d}t} = \frac{1}{h}\int \mathrm{d}\epsilon\, (\epsilon - \mu) f(\epsilon) = \frac{\pi k_\mathrm{B}^2 T^2}{12\hbar}$$

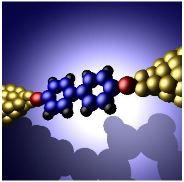
➔ “quantum of cooling power”

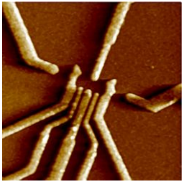
$$\approx 50\,\mathrm{pW} \quad (\text{at } T = 10\mathrm{K})$$

nonadiabatic heat pump rate ≈ 2% of optimal value

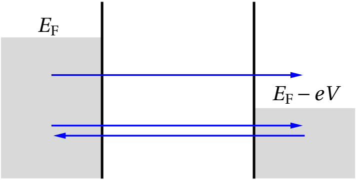
Driven transport through nanoscale conductors

- Floquet transport theory
- DC current from AC driving
  - ratchet effect
  - nonadiabatic electron pump
  - pumping heat
- Coulomb blockade effects





more on noise: counting statistics



- electron distribution $P(N,t)$?
- moments:

$$\langle N \rangle = N_0 + \frac{It}{e}$$

$$\langle N^2 \rangle = \langle N \rangle^2 + Dt$$

➔ "charge diffusion coefficient"

$$D = \lim_{t\to\infty} \frac{\langle \Delta N^2 \rangle}{t}$$

$$\boxed{S(\omega \to 0) = e^2 D}$$

D. K. C. MacDonald, 1949

strong Coulomb repulsion

- e-e interaction U
- master equation approach:
  - perturbation theory for weak wire-lead coupling
  - master equation for reduced density operator

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_{\mathrm{wire}} = \frac{\mathrm{d}}{\mathrm{d}t}\mathrm{tr}_{\mathrm{leads}}\rho, \quad I \sim \frac{\mathrm{d}}{\mathrm{d}t}\mathrm{tr}_{\mathrm{leads}} N_{\mathrm{L}}\rho, \quad S \sim \frac{\mathrm{d}}{\mathrm{d}t}\mathrm{tr}_{\mathrm{leads}} N_{\mathrm{L}}^2\rho$$

➔ counting statistics, Mac Donald formula

Elattari & Gurvitz, Phys. Lett. (2002)

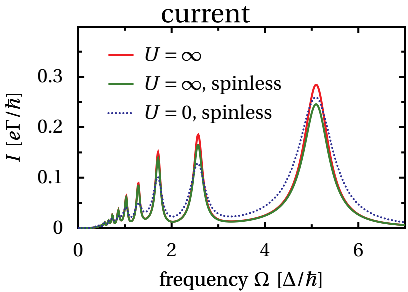
Bagrets & Nazarov, PRB (2003)

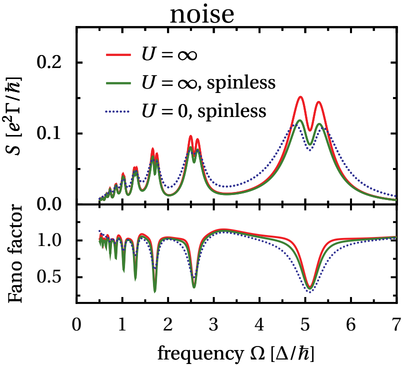
Flindt, Novotný & Jauho, PRB (2004)

  - $U \to \infty$: at most one excess electron
  - decompostion into Floquet basis
  - spin *vs.* spinless electrons

strong Coulomb repulsion

two-level electron pump:





- close to resonance: no significant changes
- finite U: work in progress

Kaiser, Hänggi, SK, EPJ B **54**, 201 (2006)

Kaiser & SK, Ann. Phys. (in press); cond-mat/0705.4204

summary

- Floquet transport theory for driven conductors
  - photon-assisted tunneling
  - ratchets, pumps, rectification
  - heat pumps
  - Coulomb repulsion
  - current and noise control
- current projects
  - disorder & pumping — Kaiser, Hänggi, SK, NJP **10** 065013 (2008)
  - stochastic driving
  - coupling to molecule vibrations
- review article

  S. Kohler, J. Lehmann, and P. Hänggi, Phys. Rep. **406**, 379 (2005)

thanks to . . .

- Franz-Josef Kaiser
  Michael Stark
  Peter Hänggi (Augsburg)
- Jörg Lehmann (Basel)
- Sébastien Camalet (CNRS Paris)
- Michael Strass (Nagler & Co)
- Abraham Nitzan (Tel Aviv)
- Miguel Rey
  Fernando Sols (Madrid)

nanosystems initiative munich

SFB 484

SFB 631

SPP 1243