Chapter 11
Electronic transport measurements

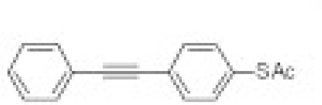
1. Measurements of the linear conductance (DC)
2. Measurement of current-voltage characteristics
3. Measurements of differential conductance dI/dV and IETS/PCS (d^2I/dV^2)
4. Thermopower
5. Shot Noise

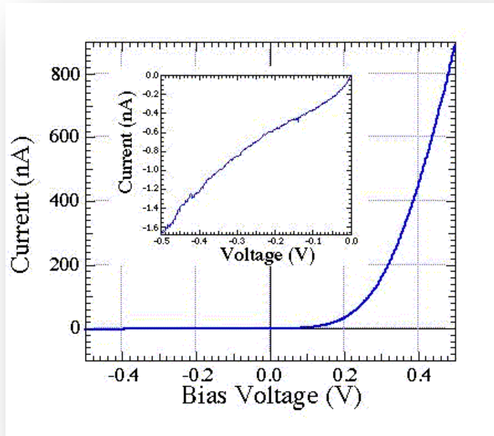
REFERENCES

1) Chapter 5 of the lecture script

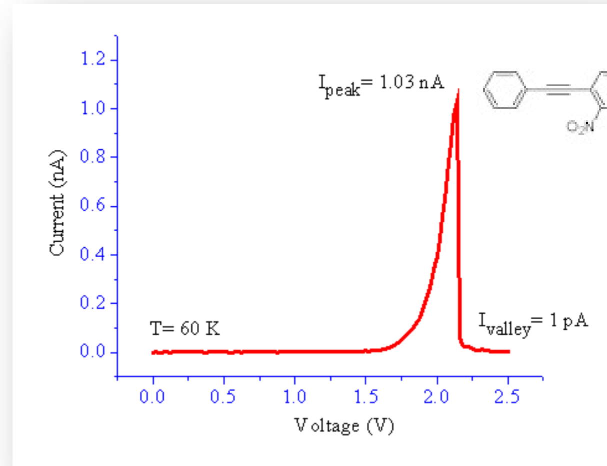
Typical Properties of Functional Structures

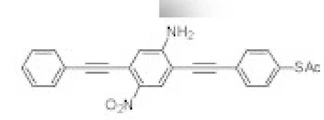
1. Diode: Au-SAM-Thiol-Au (Nanopore)
4-thioacetatebiphenyl, M. Reed, APL (1997)



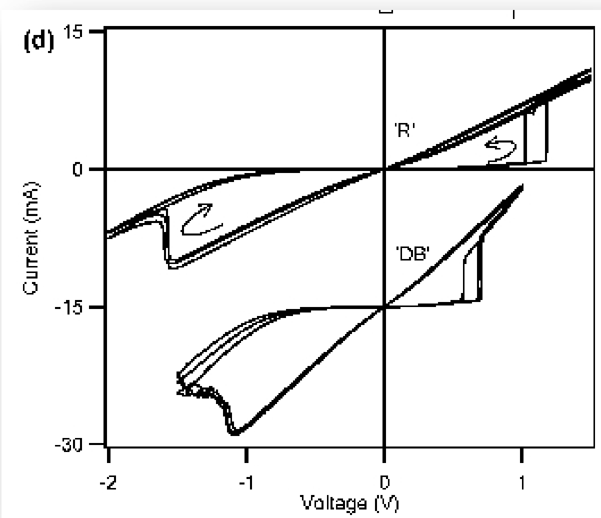


2. Switch: Nanopore (60 K)
M. Reed et al., Science (1999)

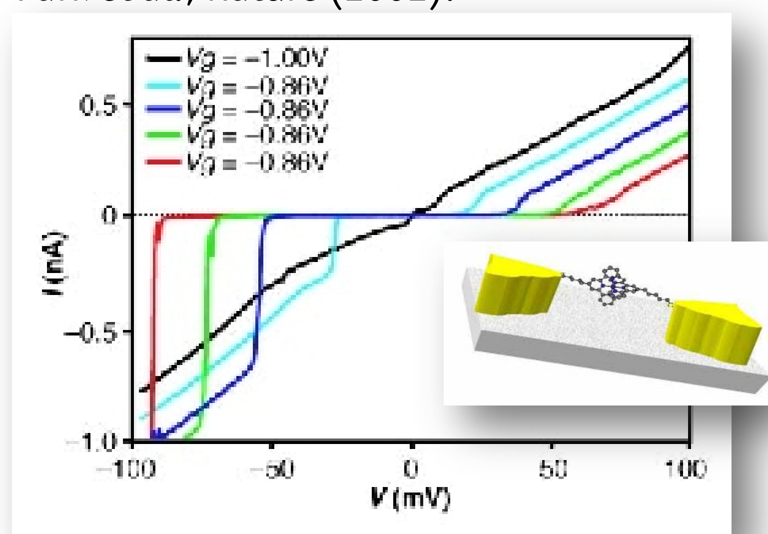




3. Reconfigurable Switch: Catanane
J.R. Heath et al., Science (2000)



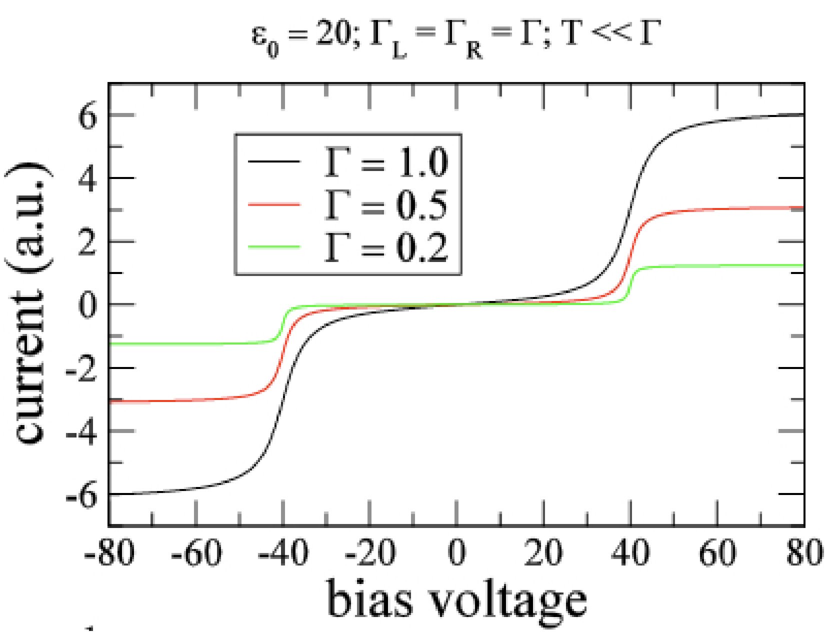
4. Single-electron transistor:
Park et al., Nature (2002).



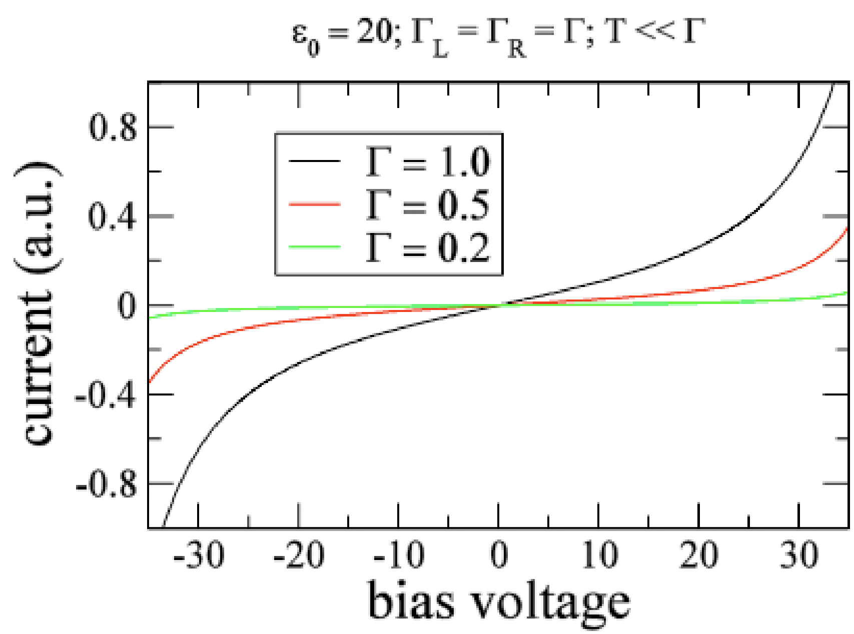
Reminder : IVs of molecular contacts

Non-linear IVs: Current bias vs. Voltage bias

Resonant case:
transport through
molecular level



Off resonant case: molecules
as tunneling junctions



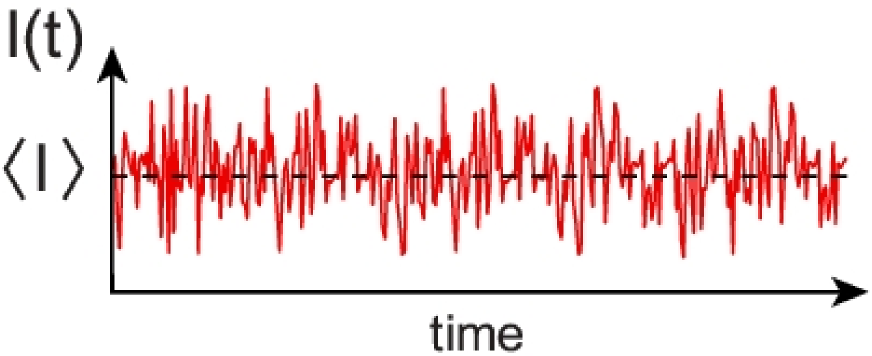
L. Zotti et al, Small (2010)

Design of the electrical measurement circuit

- What is the property to measure?
- Energy resolution? Which voltage range?
- Absolute & relative amplitude of signal?
- Small relative signal on constant background?
  -> Lock in technique
- Large variations of the background (high dynamic range)
  -> I & V have to be measured
- Which absolute resistance/conductance regime?
- Expected size of voltage signal:
  ~ 1 nV or 1 pA possible with RT electronics (thermal noise),
  for smaller voltages: SQUID or cold amplifiers

Reminder: 10.1 Shot noise

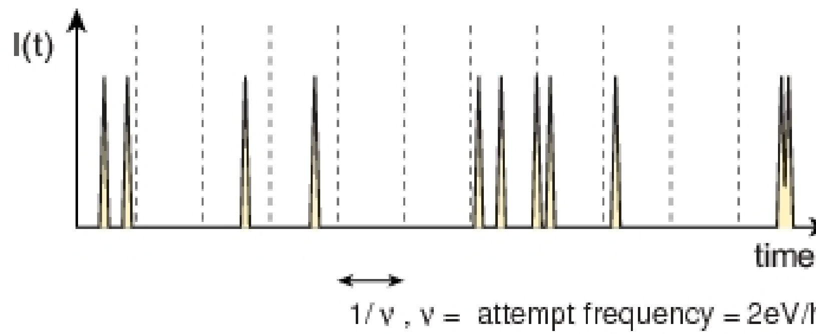
Intrinsic current fluctuations of an electrical resistor:



* Thermal fluctuations:

Johnson /Nyquist noise: $S_I = 4k_BTG$

* Non-equilibrium fluctuations (shot noise): randomly distributed tunneling of *q* discrete charges.



$$S_{Poisson} = 2q\,|\,I\,|$$

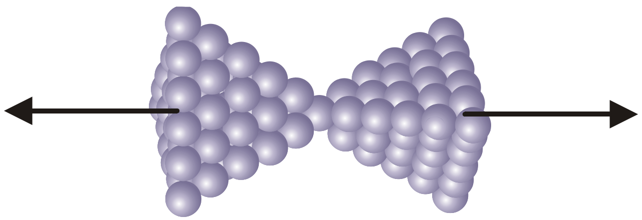
(W. Schottky 1918)

Noise: Definition

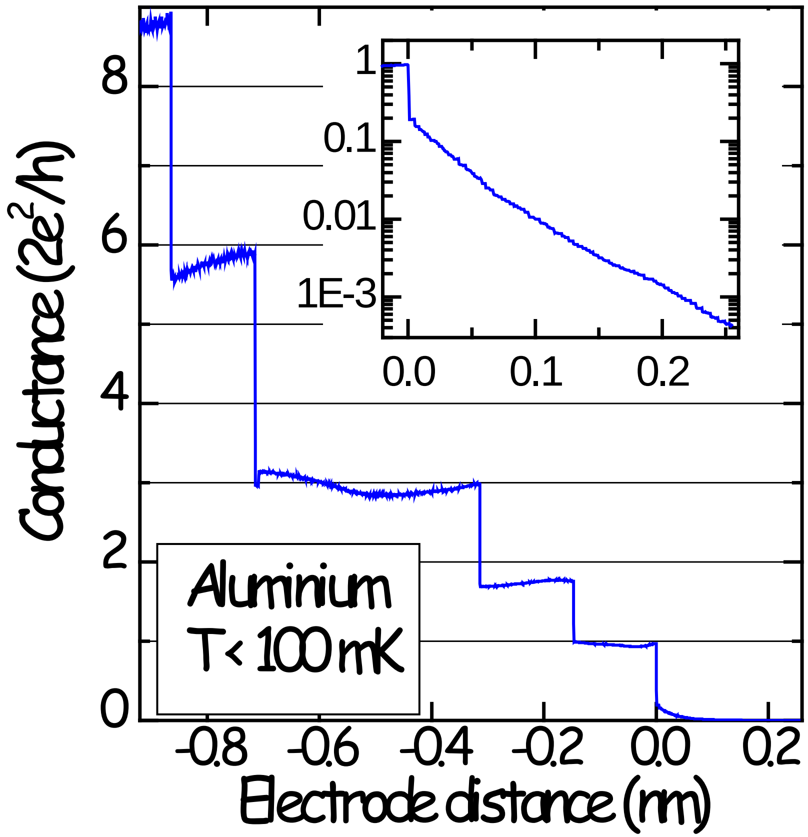
$$S_I(\omega) = 2\int dt e^{i\omega t}\left\langle \Delta\hat{I}(t+t_0)\Delta\hat{I}(t_0)\ \right\rangle$$

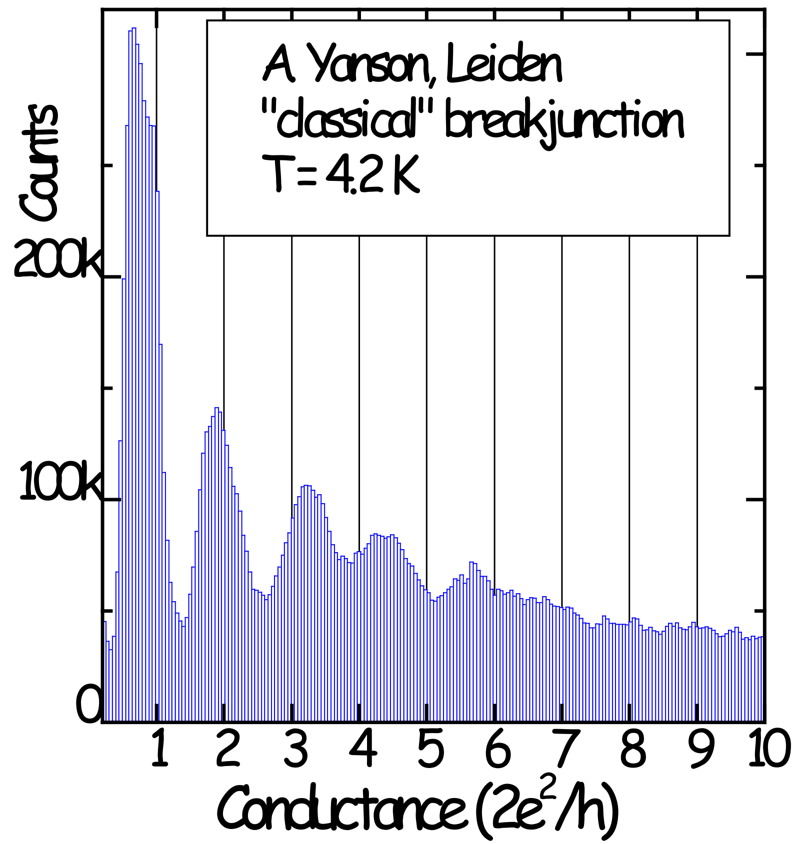
$$\Delta\hat{I}(t) \equiv \hat{I}(t) - \left\langle \hat{I}(t) \right\rangle$$

Typical Measurement Task in Molecular Electronics:



ALUMINUM
FEW-ATOMS CONTACTS





Histogram calculated from >6200 scans

Particular requirements for electronic measurement of atomic and molecular contacts

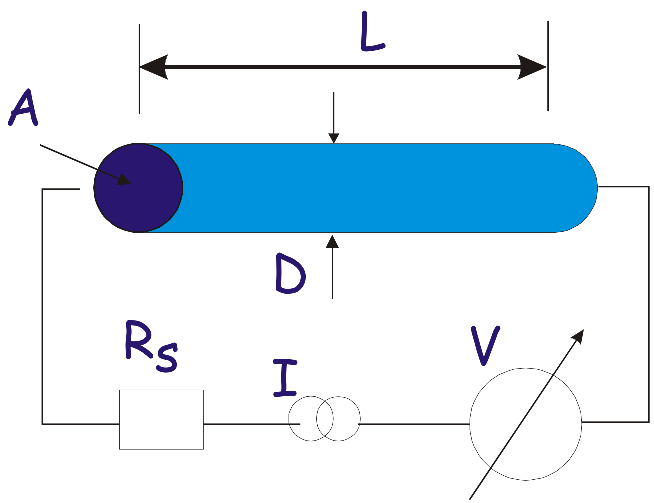
- Wide range of conductance from 10^{-5} G_0 (10 GΩ) to few G_0 (kΩ)
- Measurement of linear conductance, e.g. for conductance histograms: Correct choice of bias voltage to assure working in the linear regime - difficult because nonlinearities appear on varying voltage scales (μV to V)
- Self-heating of the contacts due to Joule dissipation is difficult to detect & to discriminate from intrinsic properties
- Sudden voltage spikes and jumps may destroy the sample.
- Extreme variation of the differential conductance within small changes of the bias.
- Strongly nonlinear IVs with negative differential resistance NDR or hysteresis -> voltage bias vs. current bias -> hysteresis effects
- Limited lifetime of the junctions.

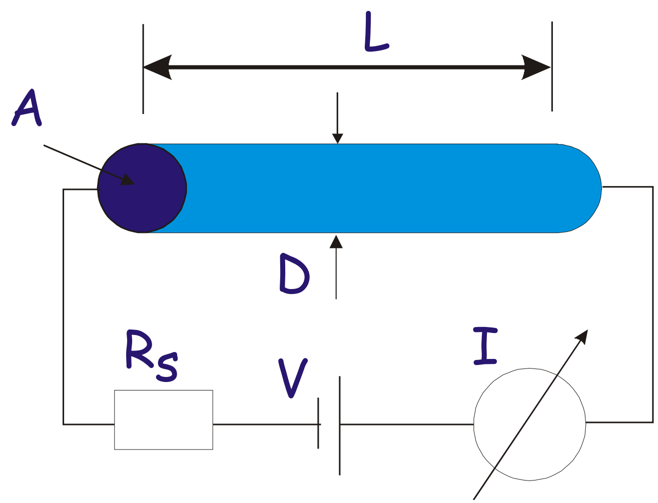
11.1 Conductance vs. Resistance measurements

Ohm's law – linear conductance

$$\text{Conductance } G = \frac{\text{Current } I}{\text{Voltage } V}$$

$$\text{Differential Conductance} = \frac{dI}{dV}$$

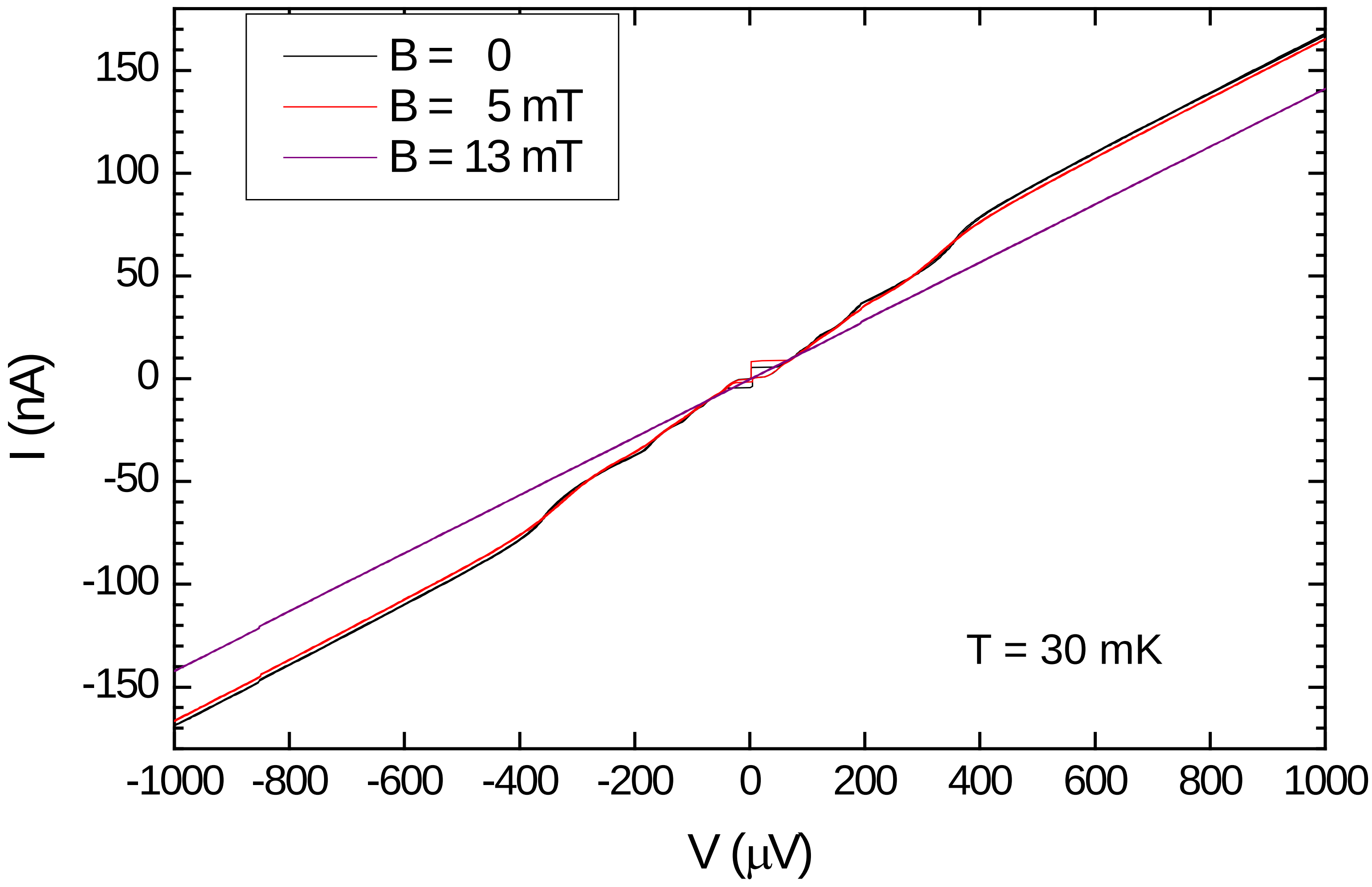




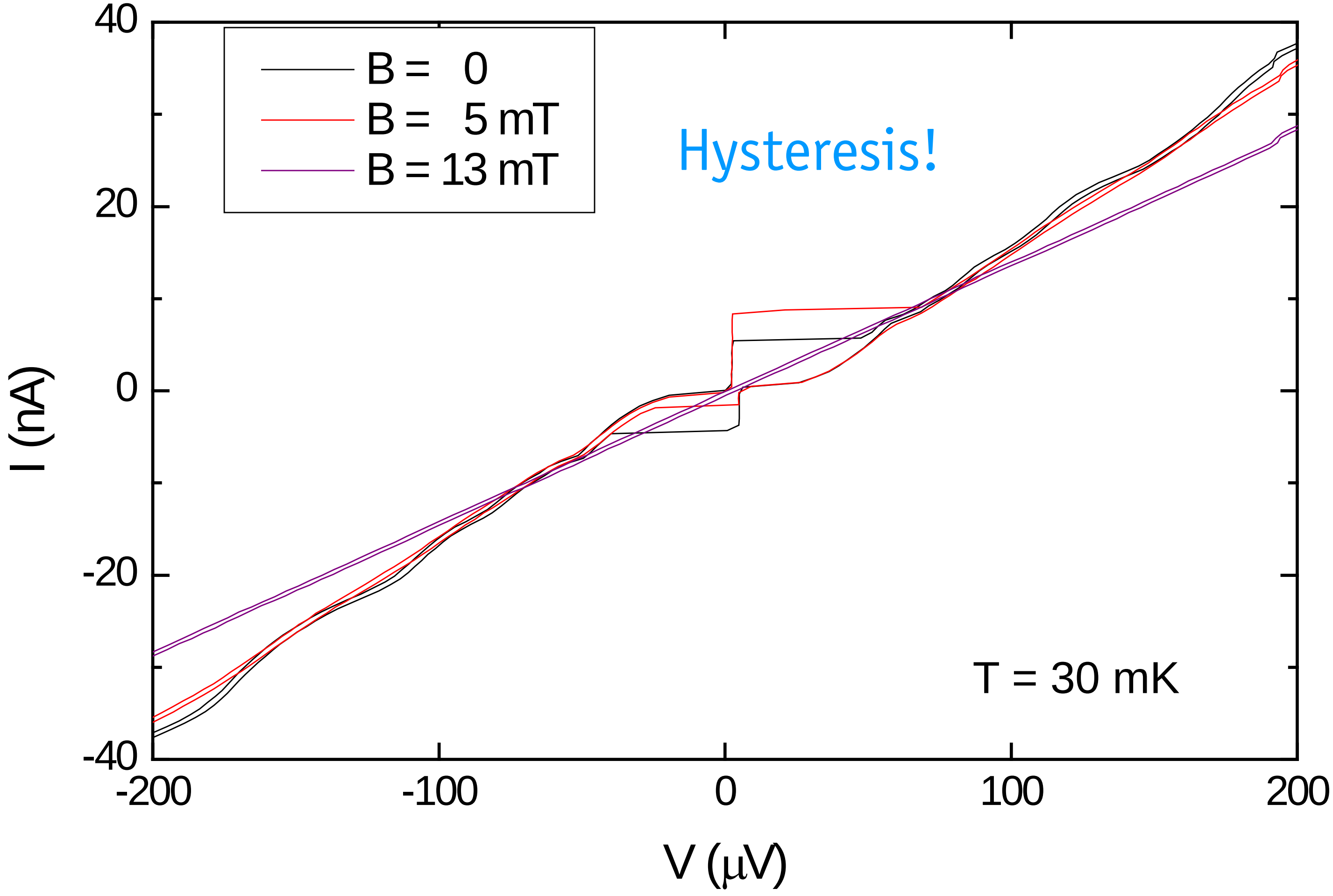
<u>High resistance $R_x > R_s$: (Low conductance $G_x < G_s$)</u>
Voltage bias, current measurement – effective conductance measurement

<u>Low resistances $R_x < R_s$: (High conductance $G_x > G_s$)</u>
Current bias, voltage measurement – effective resistance measurement

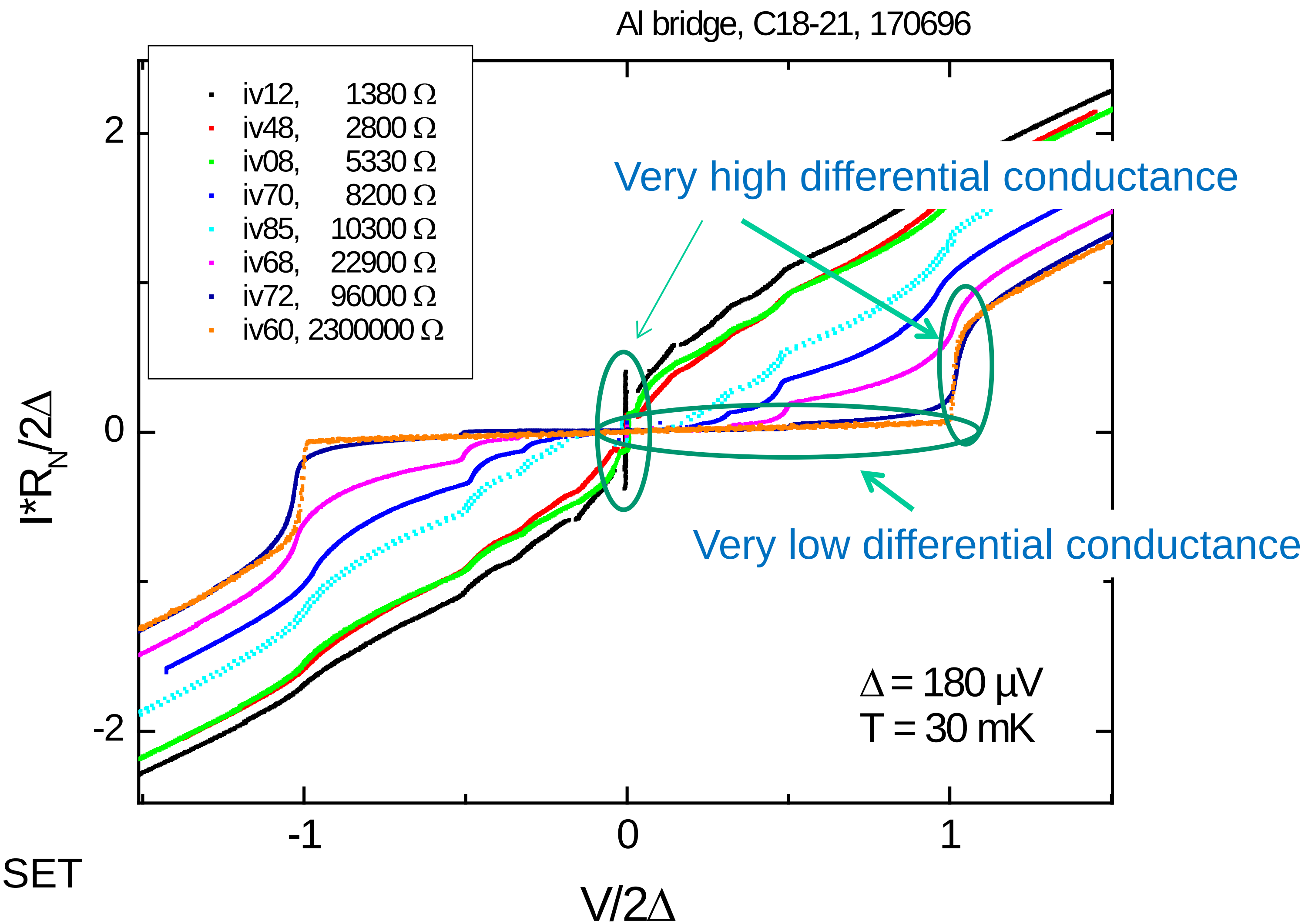
Example: IVs of a superconducting atomic contact



Example: IVs of a superconducting atomic contact

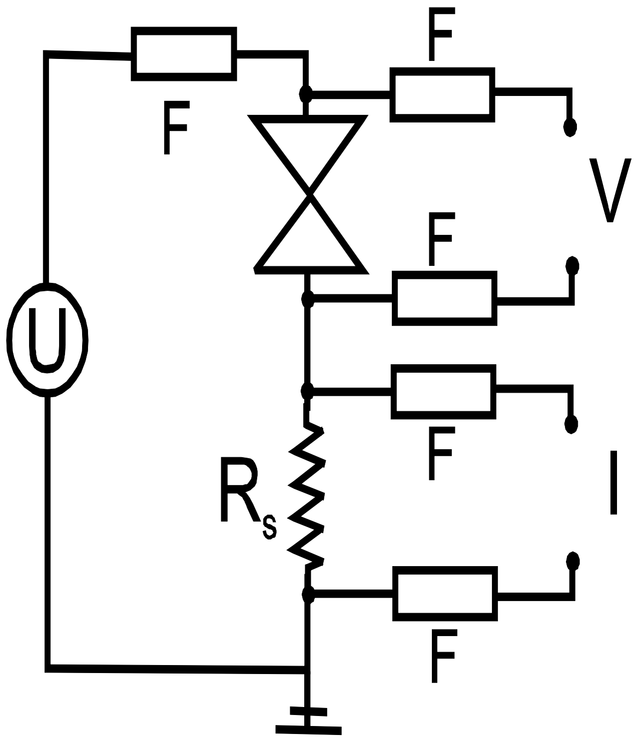


Example: IVs of a superconducting atomic contact



Similar: SET

11.2 Nonlinear-Conductance Measurements with high dynamic range & high spectral resolution



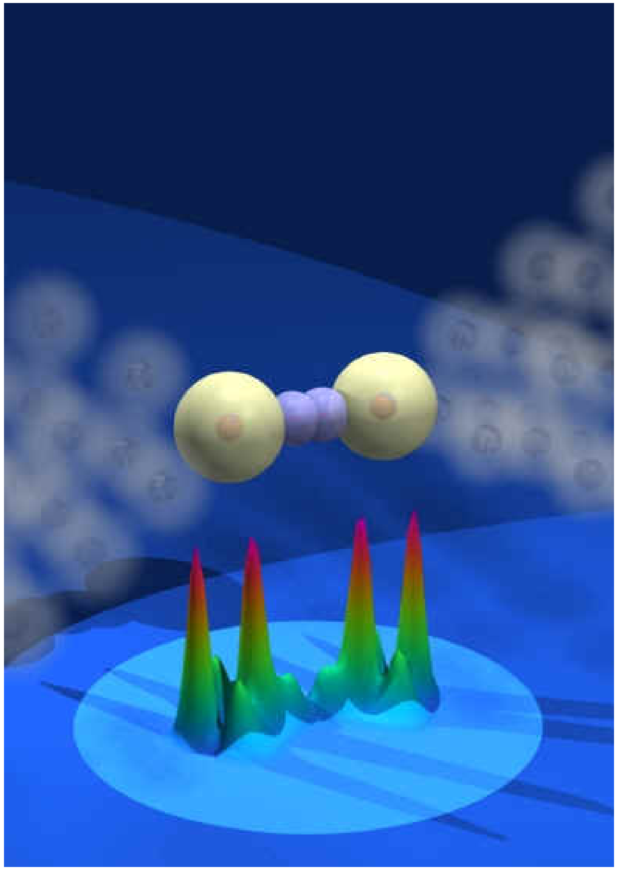
After amplification :
V -> x-channel, I -> y-channel of oscilloscope or analogue-digital converter

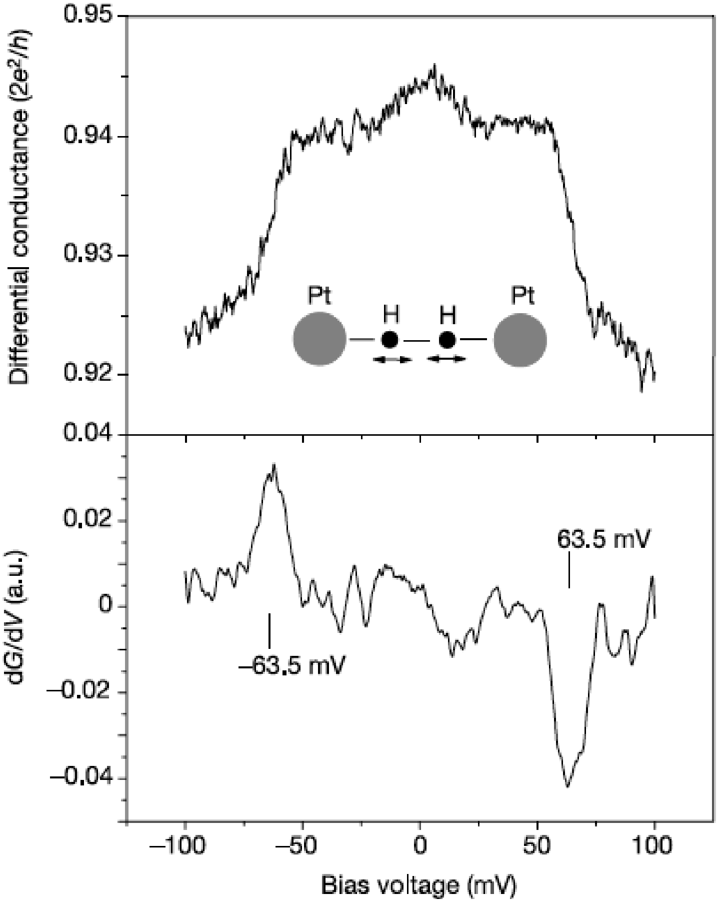
Set-up for measuring IVs of atomic contacts with strongly varying linear and differential conductance (corresponding to differential resistances of kΩ to MΩ)

V : Bias voltage applied to series of $R_s + R_x$
R_s: series resistance, chosen according to expected differential resistance
For $R_s < R_x$: Voltage bias, for $R_s > R_x$: effective current bias
Minimum R_s necessary for current limitation
F: Low-pass filters, necessary for achieving high energy resolution

Reminder: 9.1 Point contact spectroscopy (PCS) of Pt-H_2-Pt junctions

*R.H.M. Smit, Y. Noat, C. Untiedt, N.D. Lang, M.C. van Hemert, J.M. van Ruitenbeek, Nature **419**, 906 (2002)*

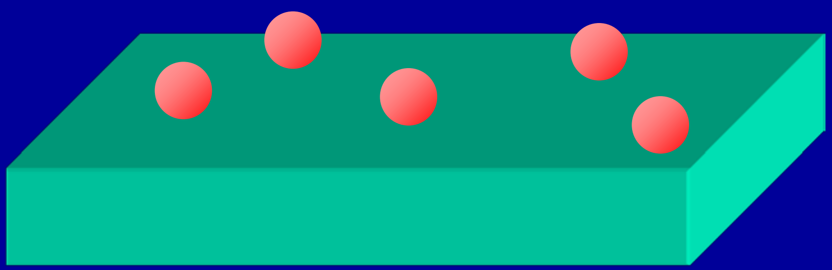




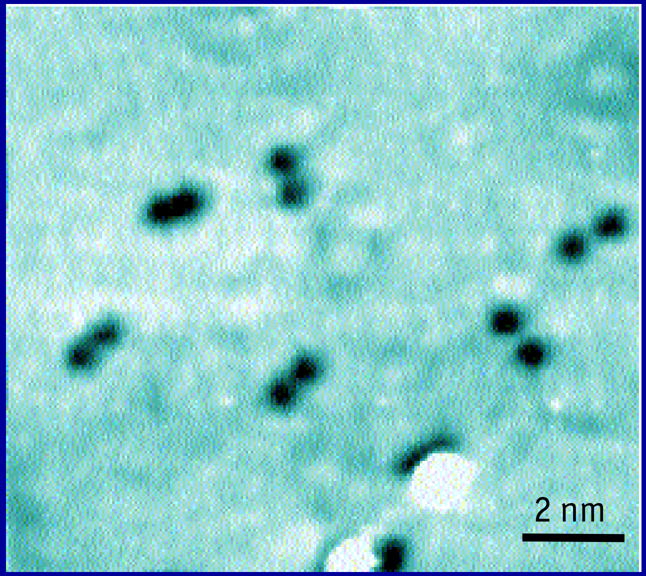
Measurement of the conductance of a hydrogen molecule between Pt leads with the break-junction technique

II.3 Scanning probe spectroscopy and Inelastic Electron Tunneling Spectroscopy

„negative“ atoms ?



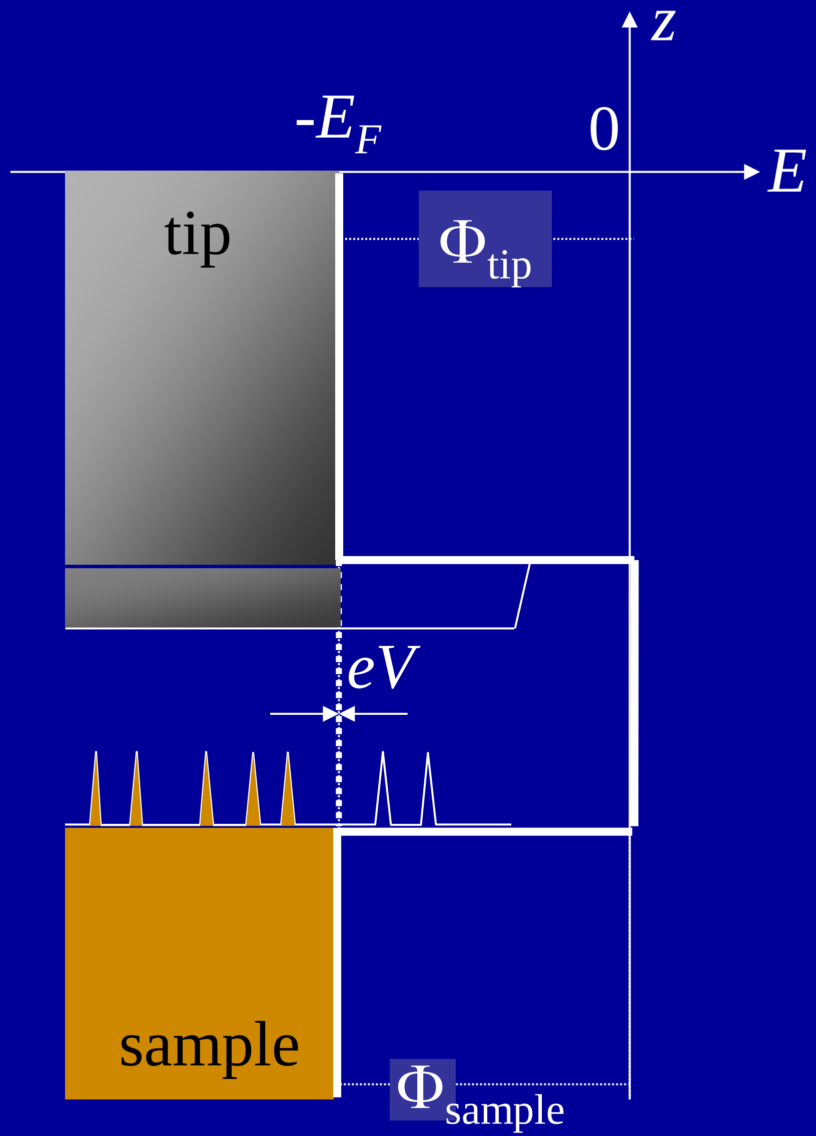
oxygen / Pt(111)

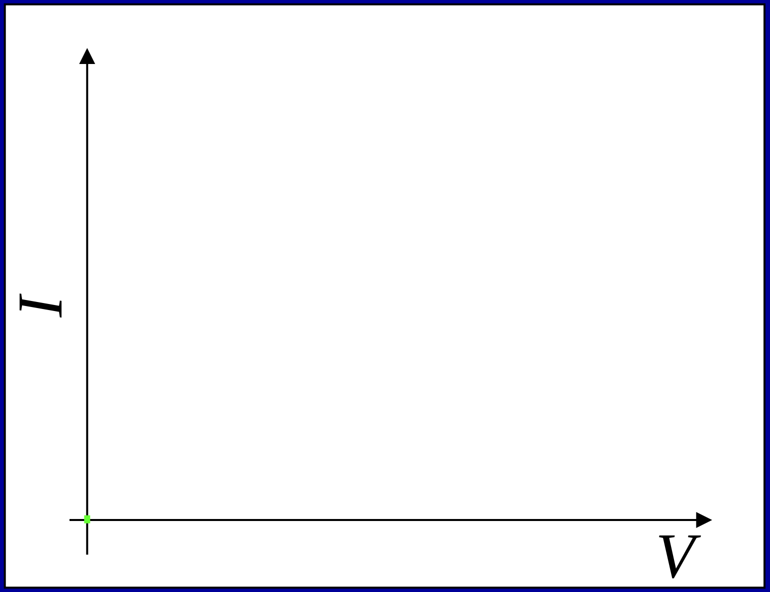


J.Wintterlin, Berlin

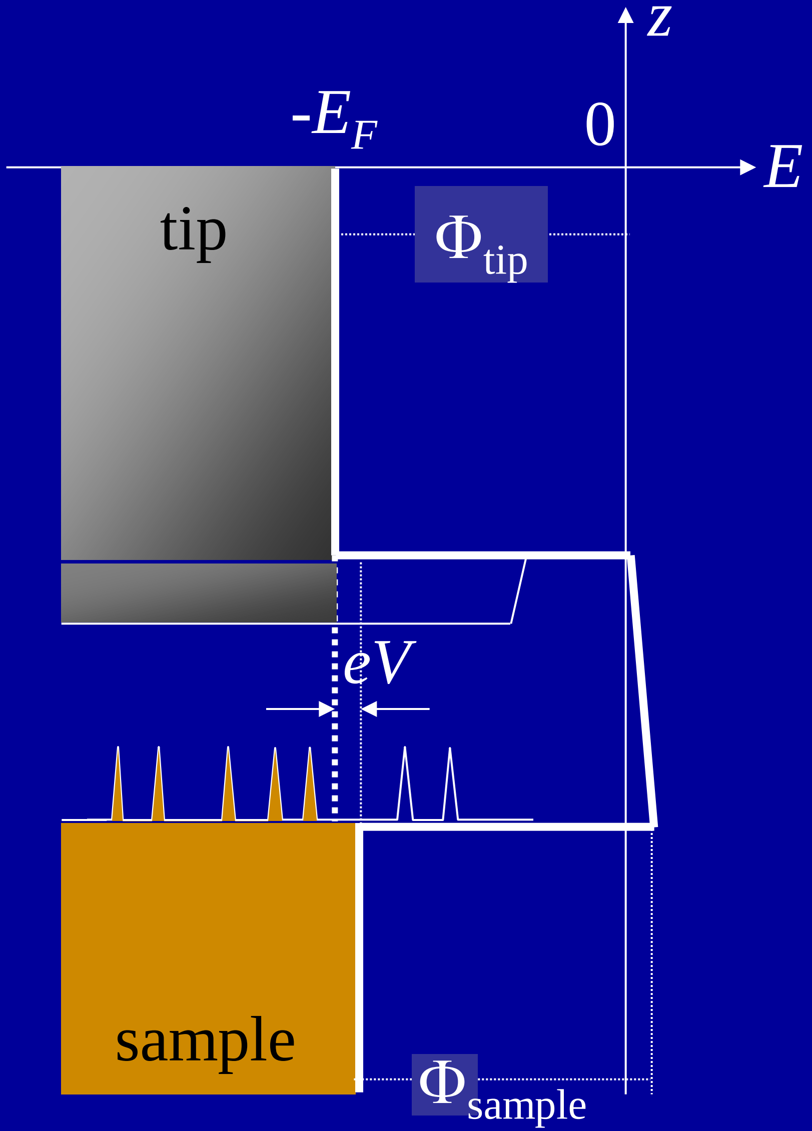
Courtesy: G. Costantini, MPI Stuttgart

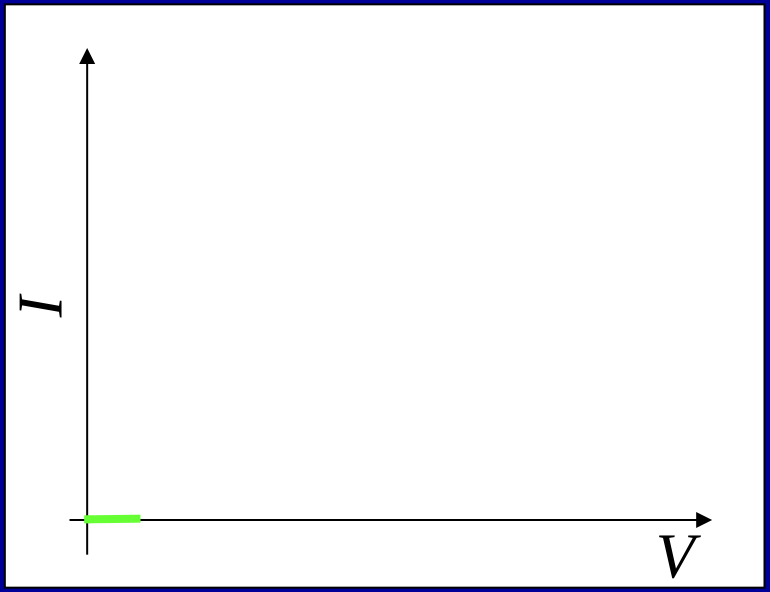
Tunneling spectroscopy





Tunneling spectroscopy





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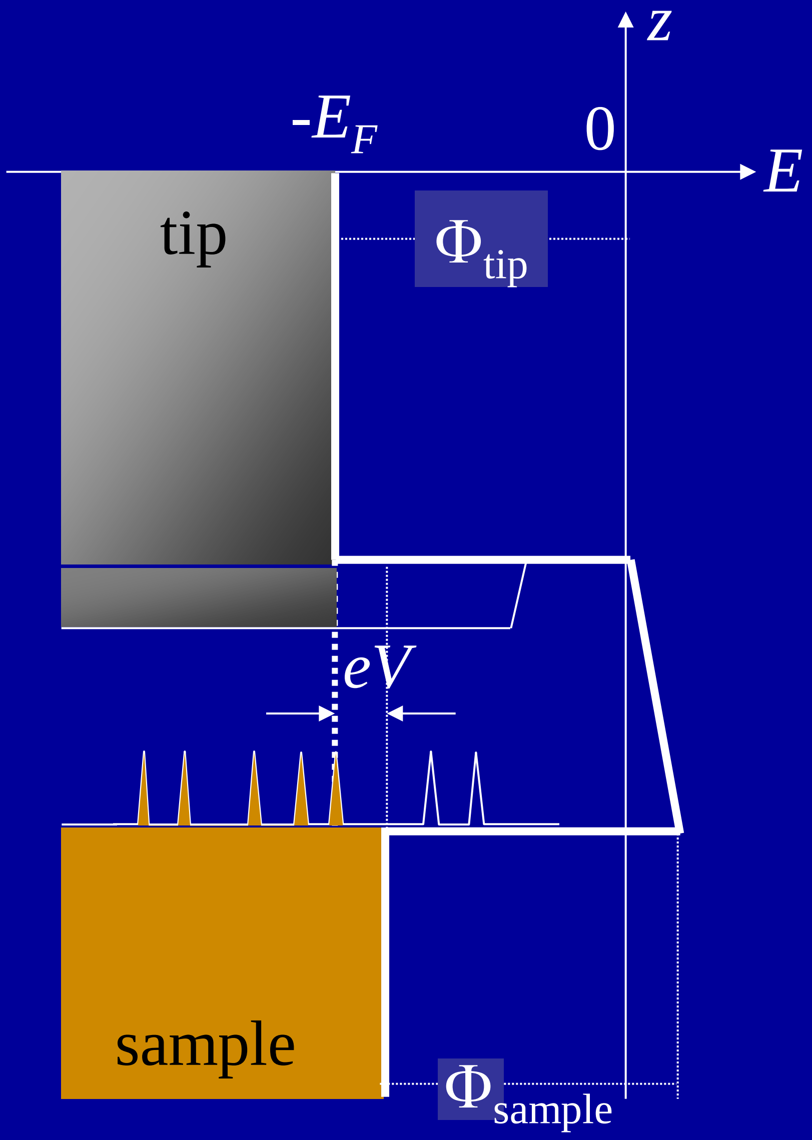

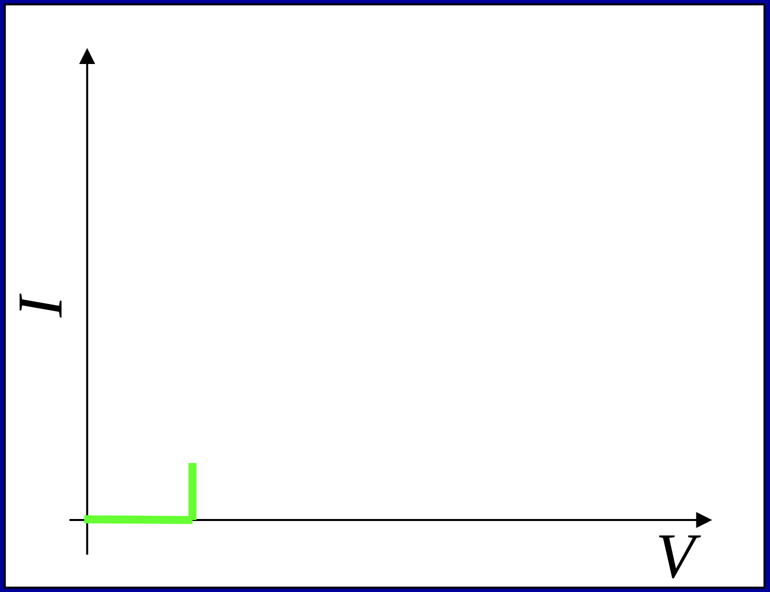

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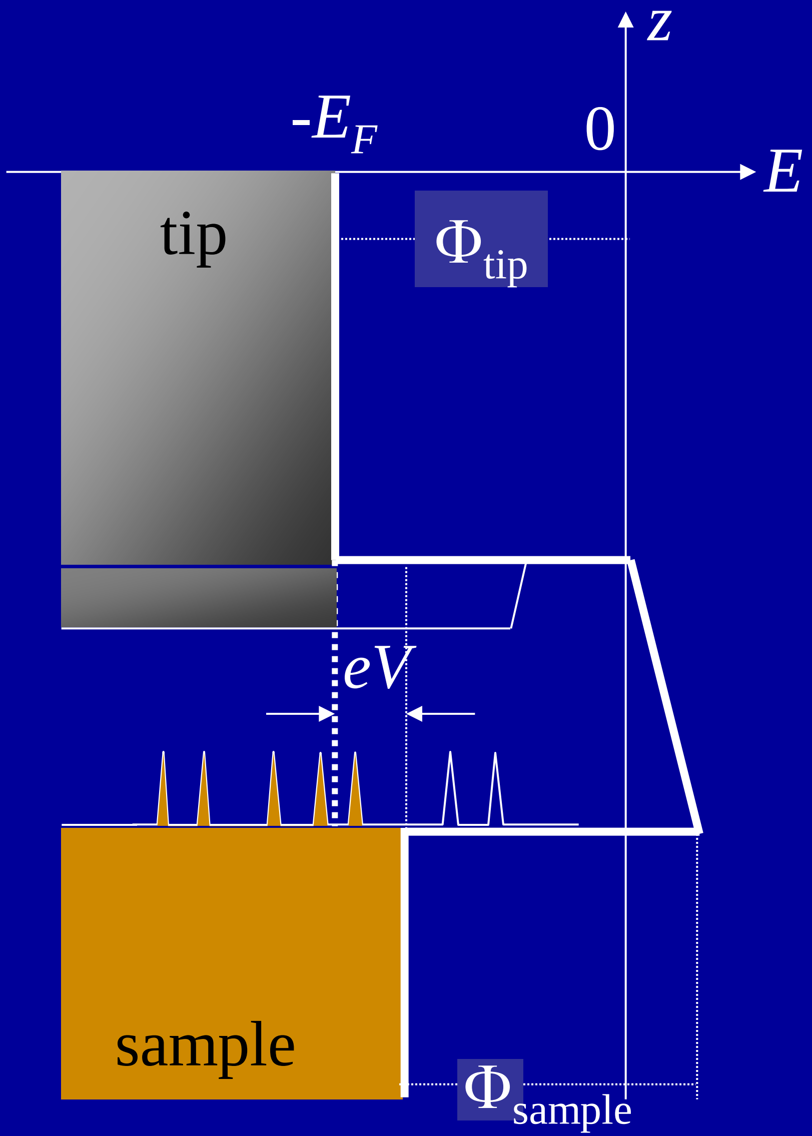
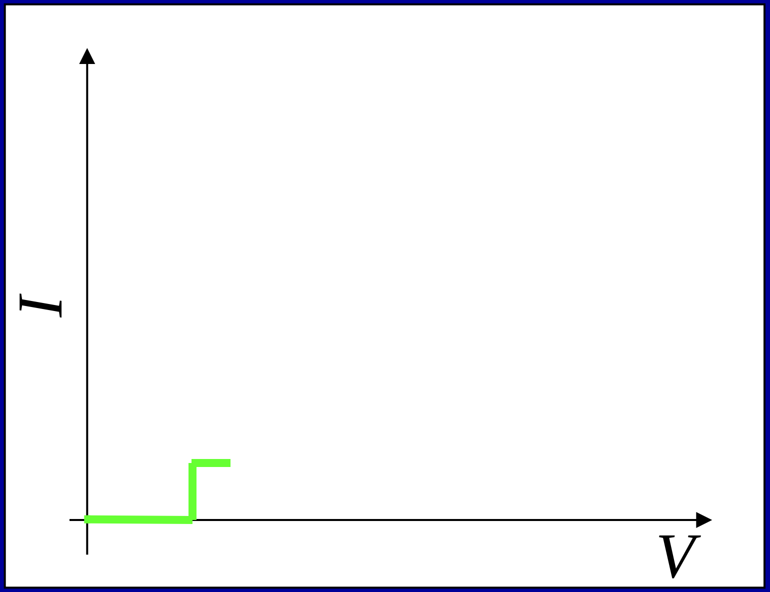
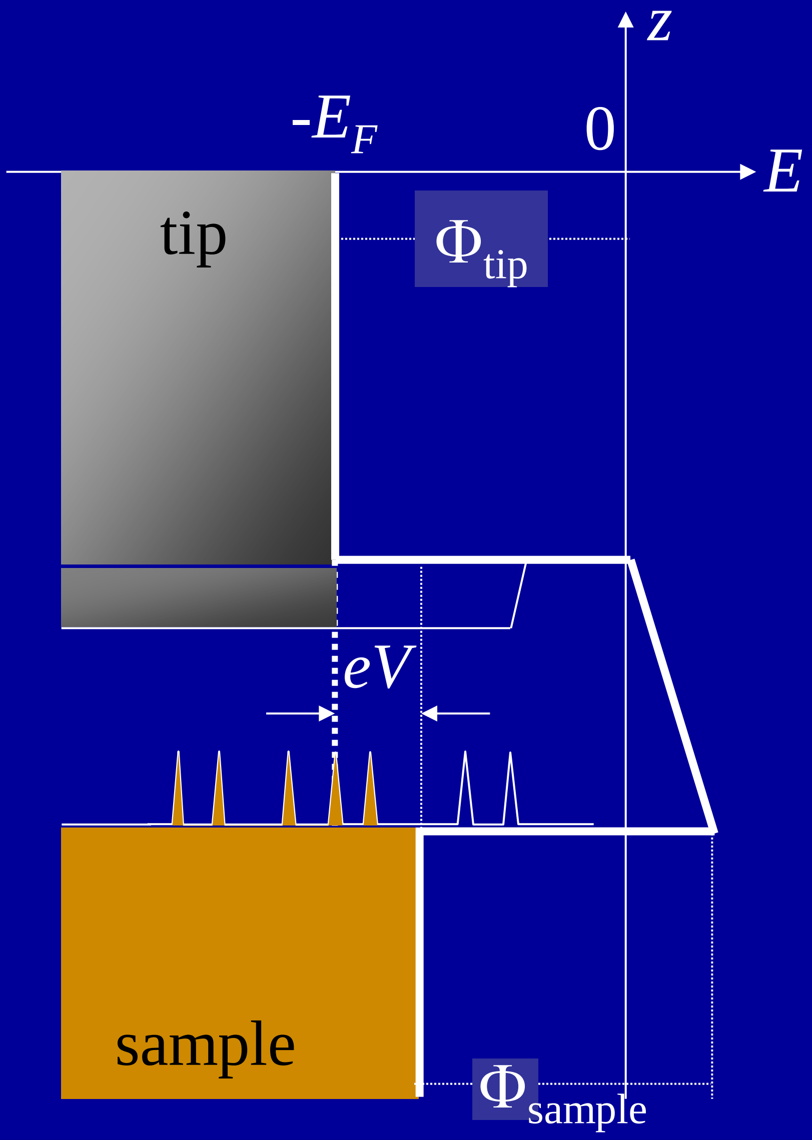
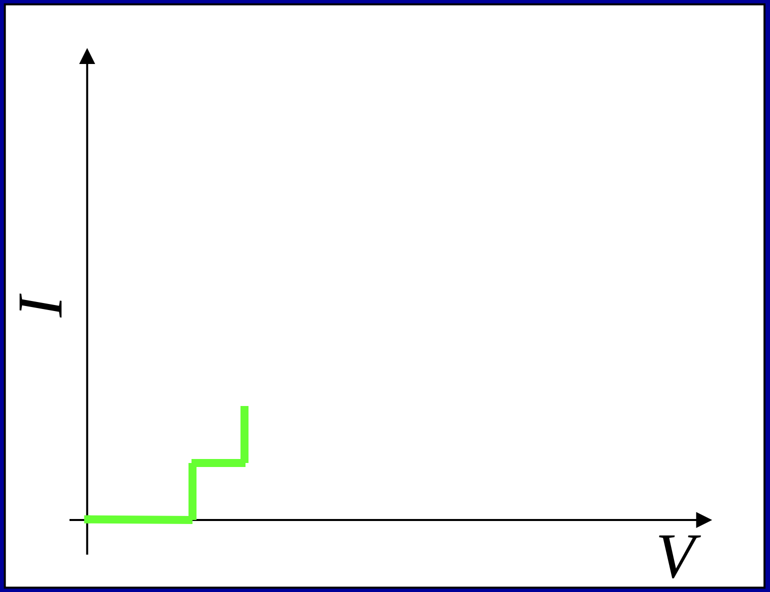


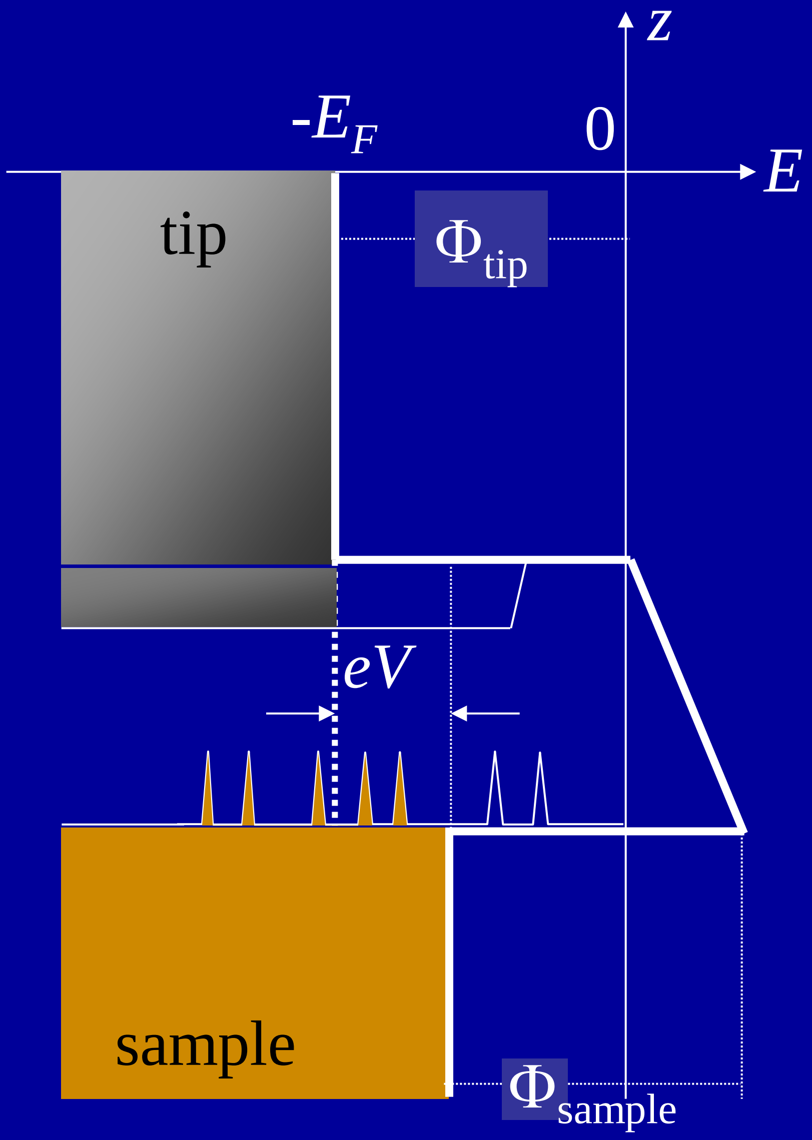


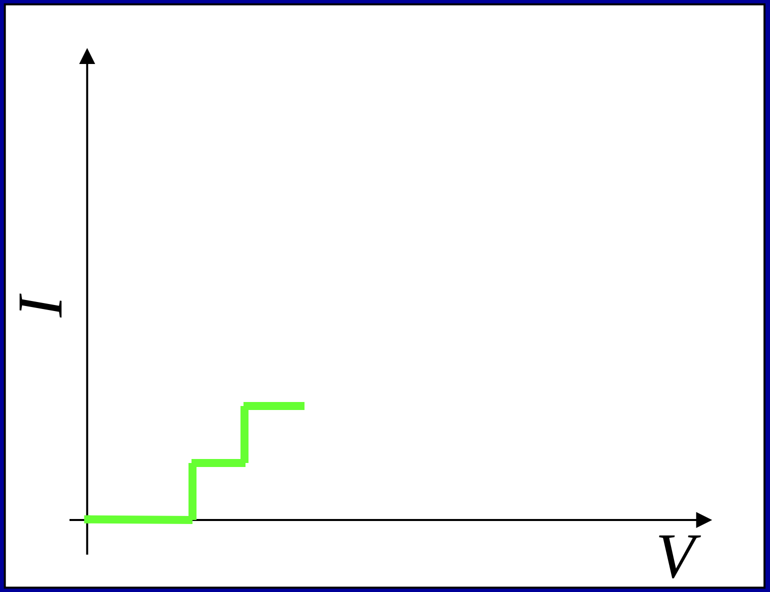
Tunneling spectroscopy



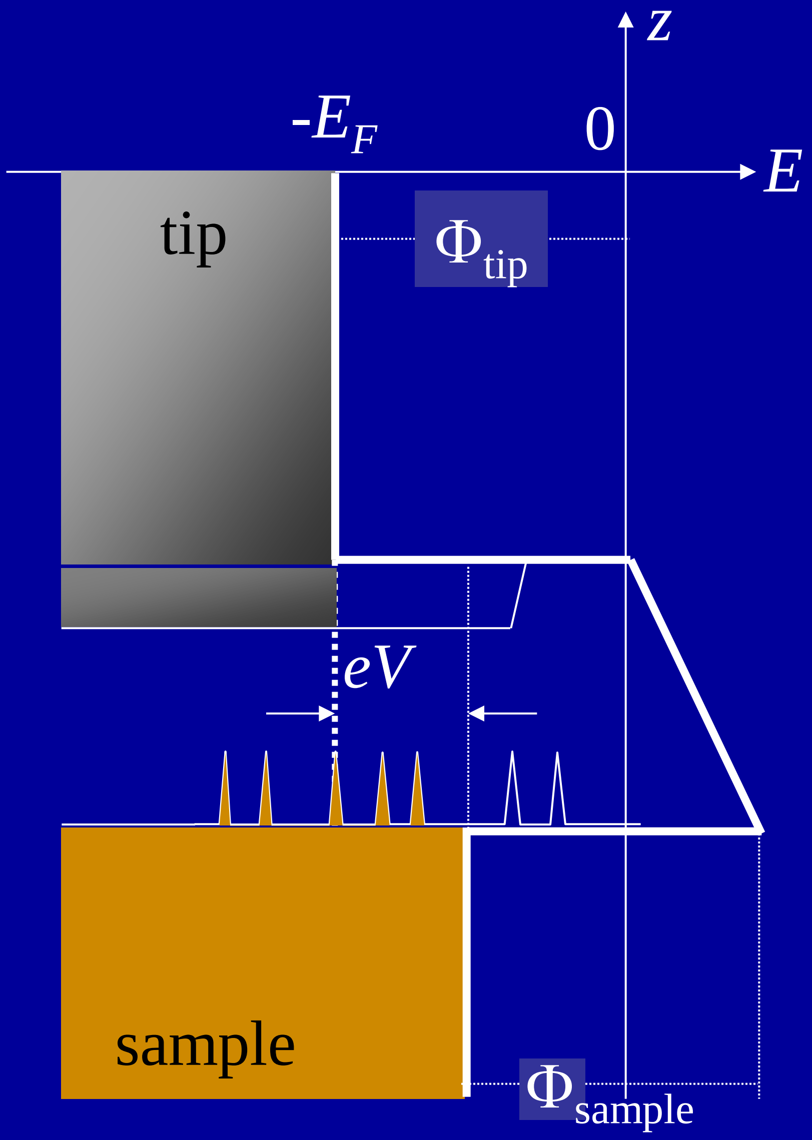


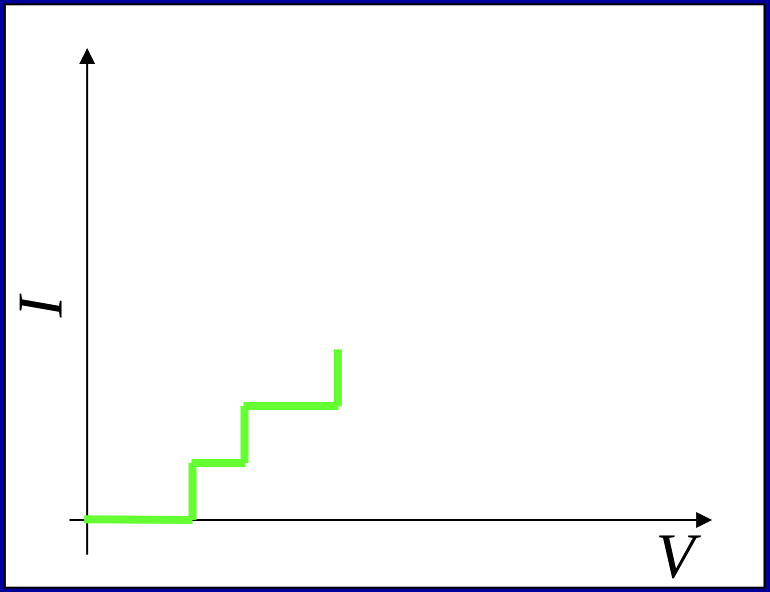
Tunneling spectroscopy





Tunneling spectroscopy





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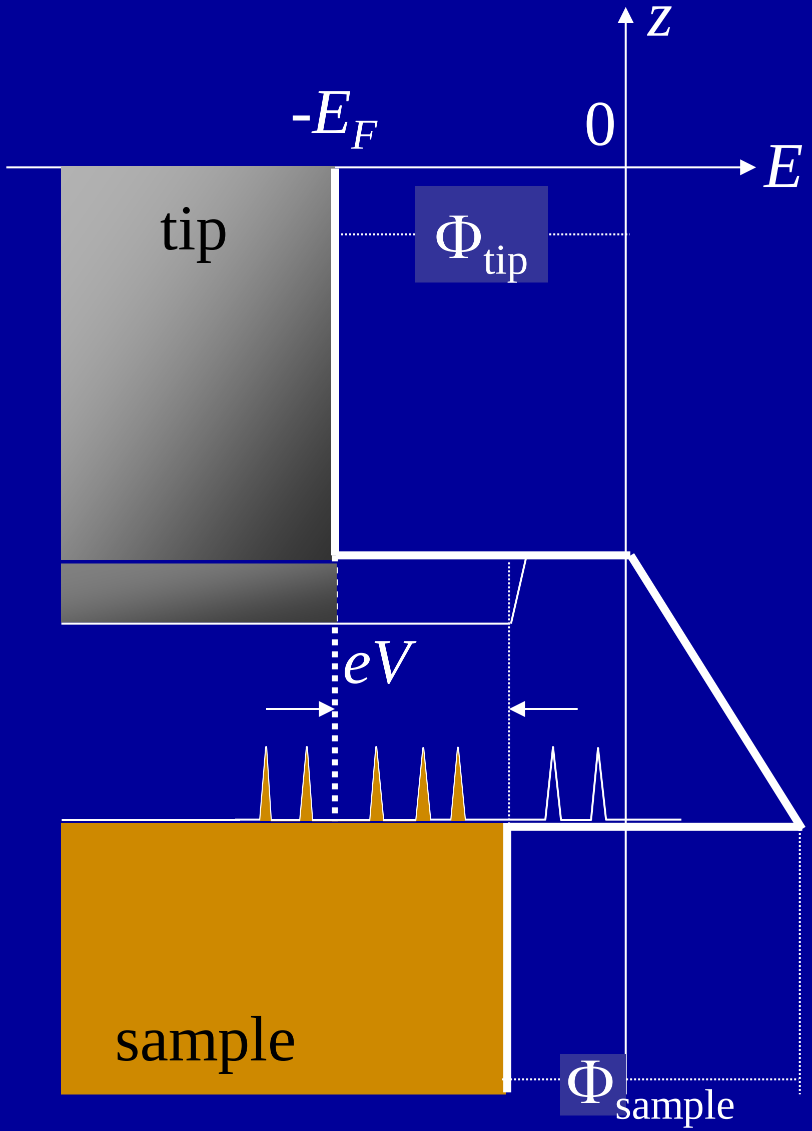
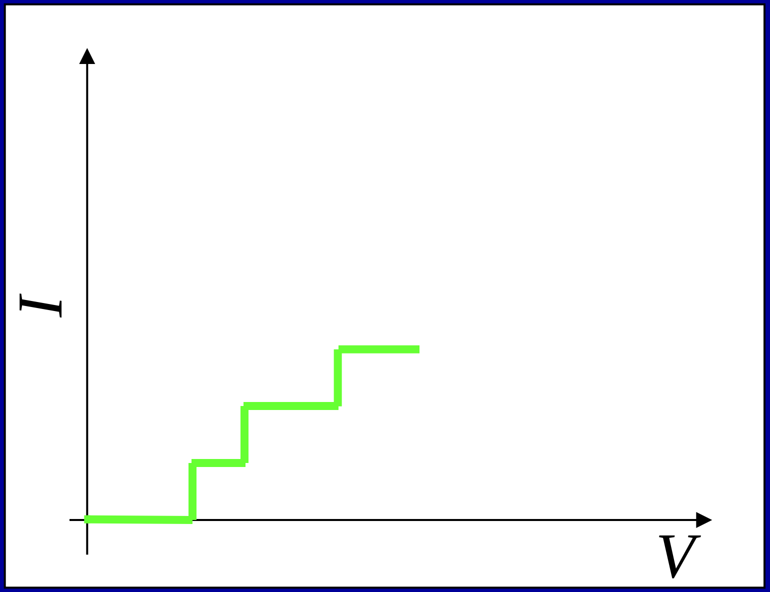
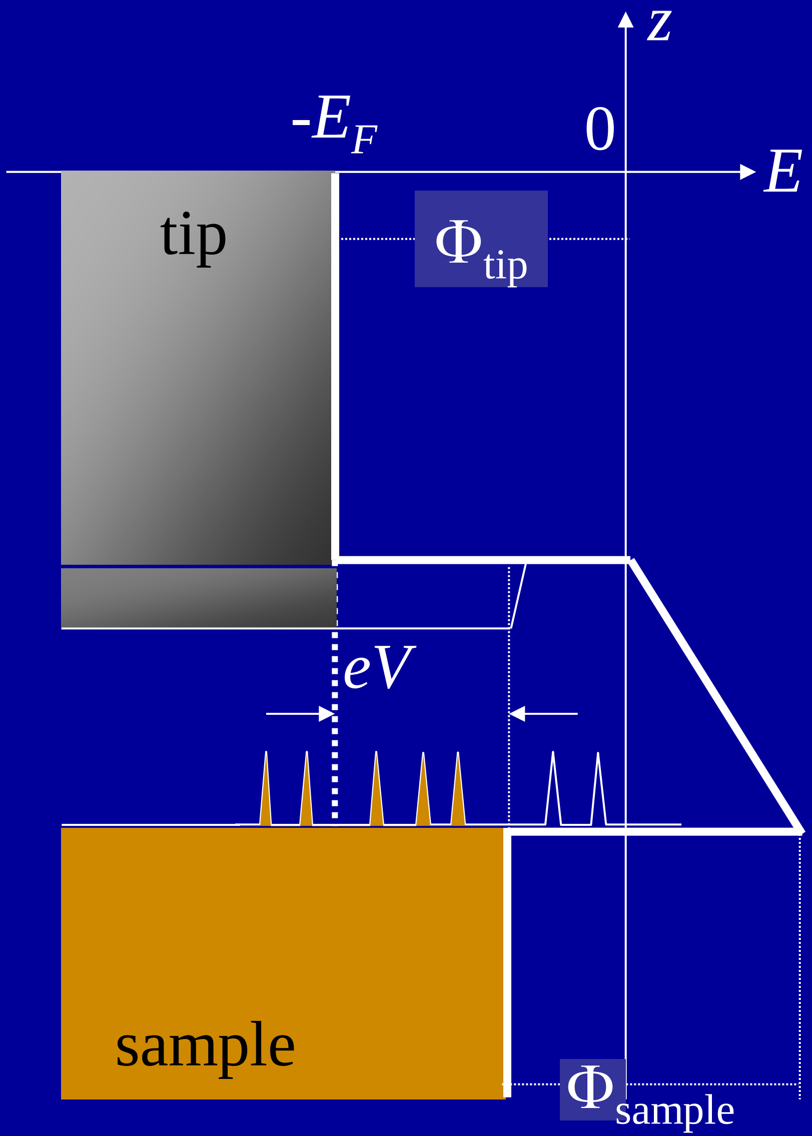
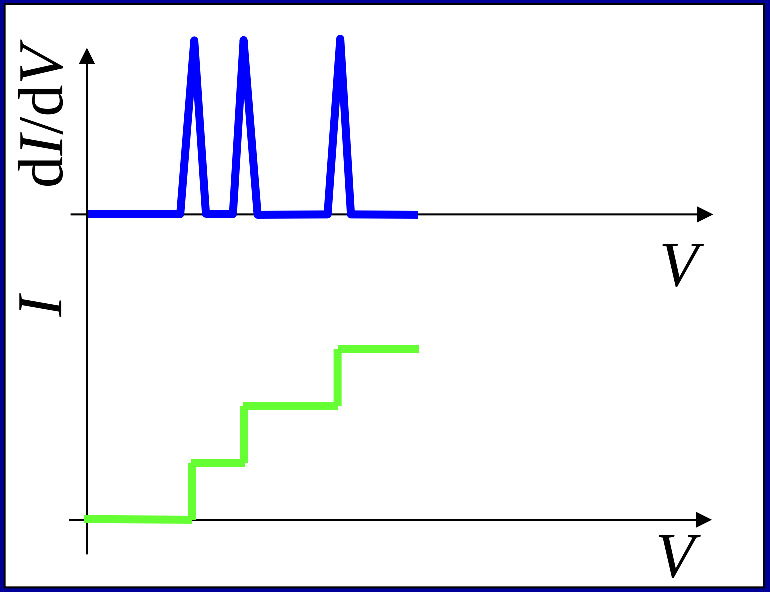


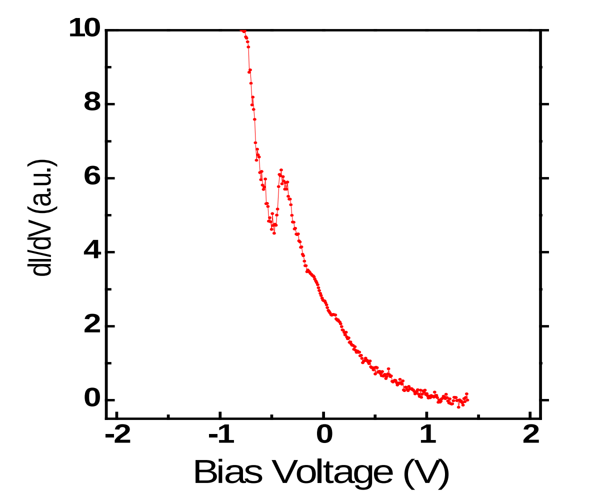


Tunneling spectroscopy



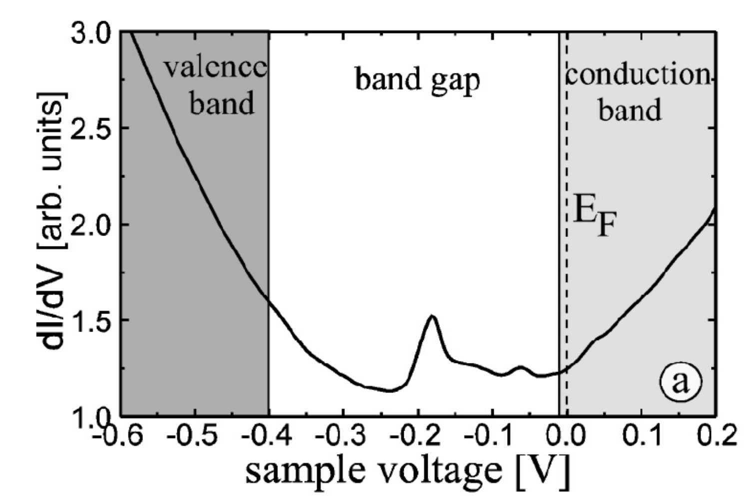


surface state Cu(111)



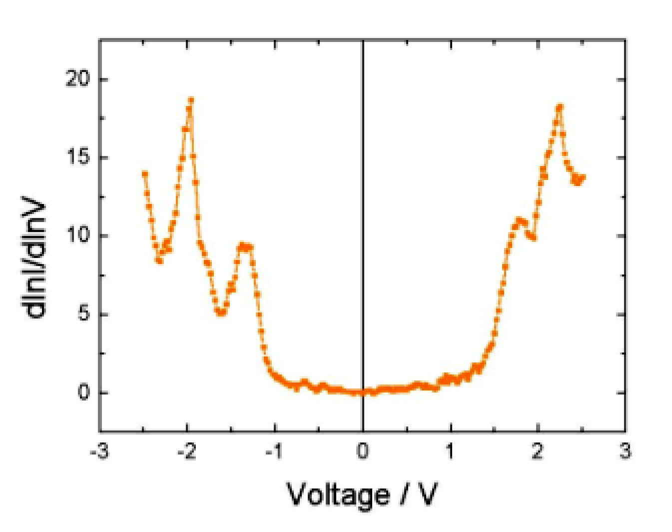
L. Vitali, MPI-FKF

n-InAs(110)



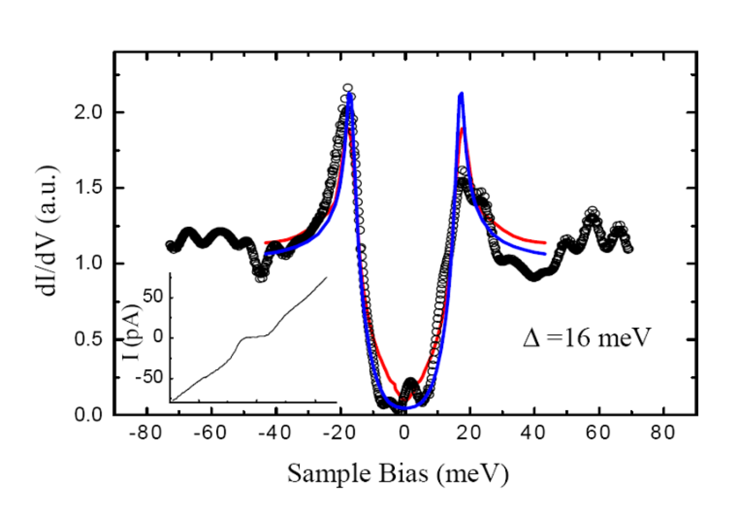
R. Dombrowski et al., Phys. Rev. B **59**, 8043 (1999)

HBC molecules on Au(001)



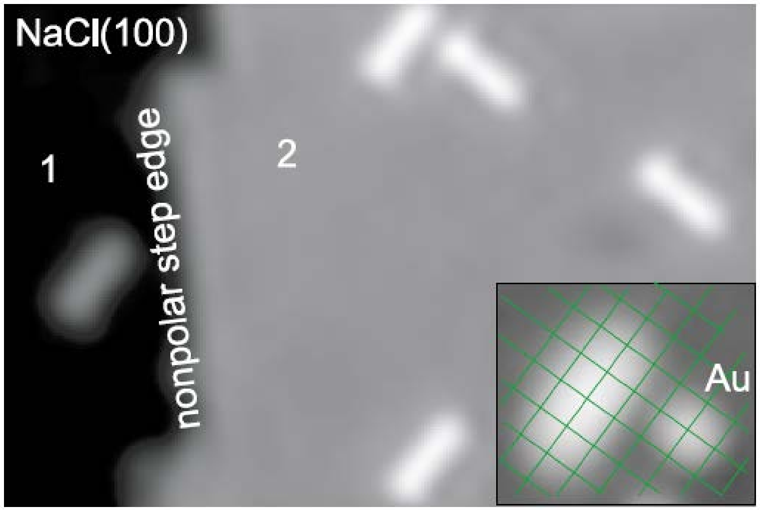
T. Fritz, TU Dresden

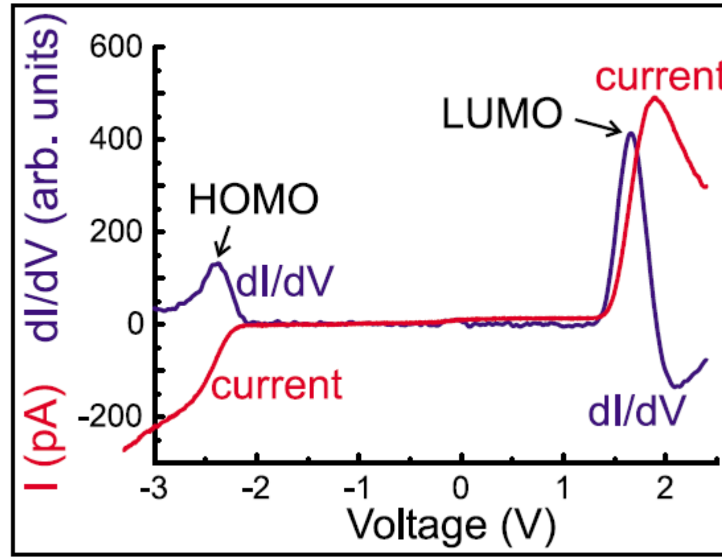
YBCO films

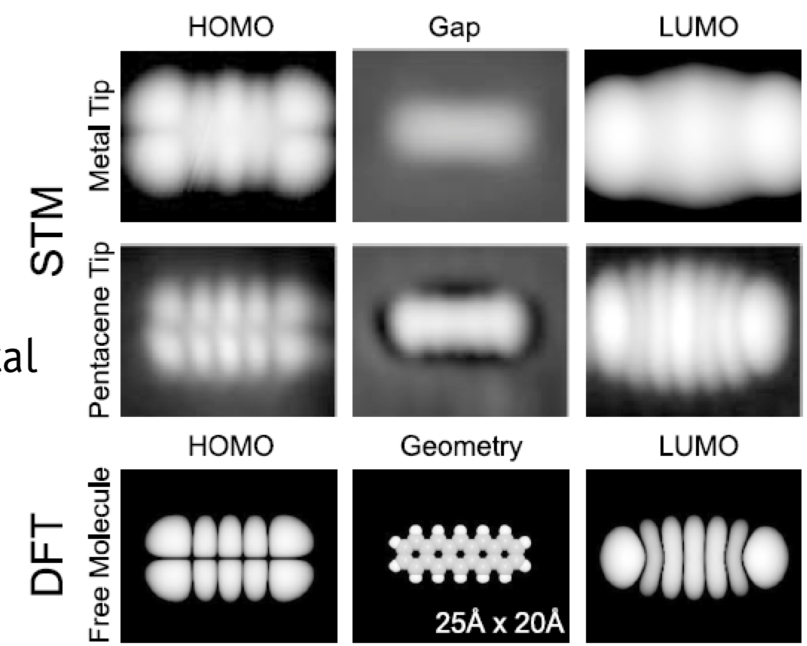


A. Sharoni, et al., Europhys. Lett. **62**, 883 (2003)

Imaging of Molecules



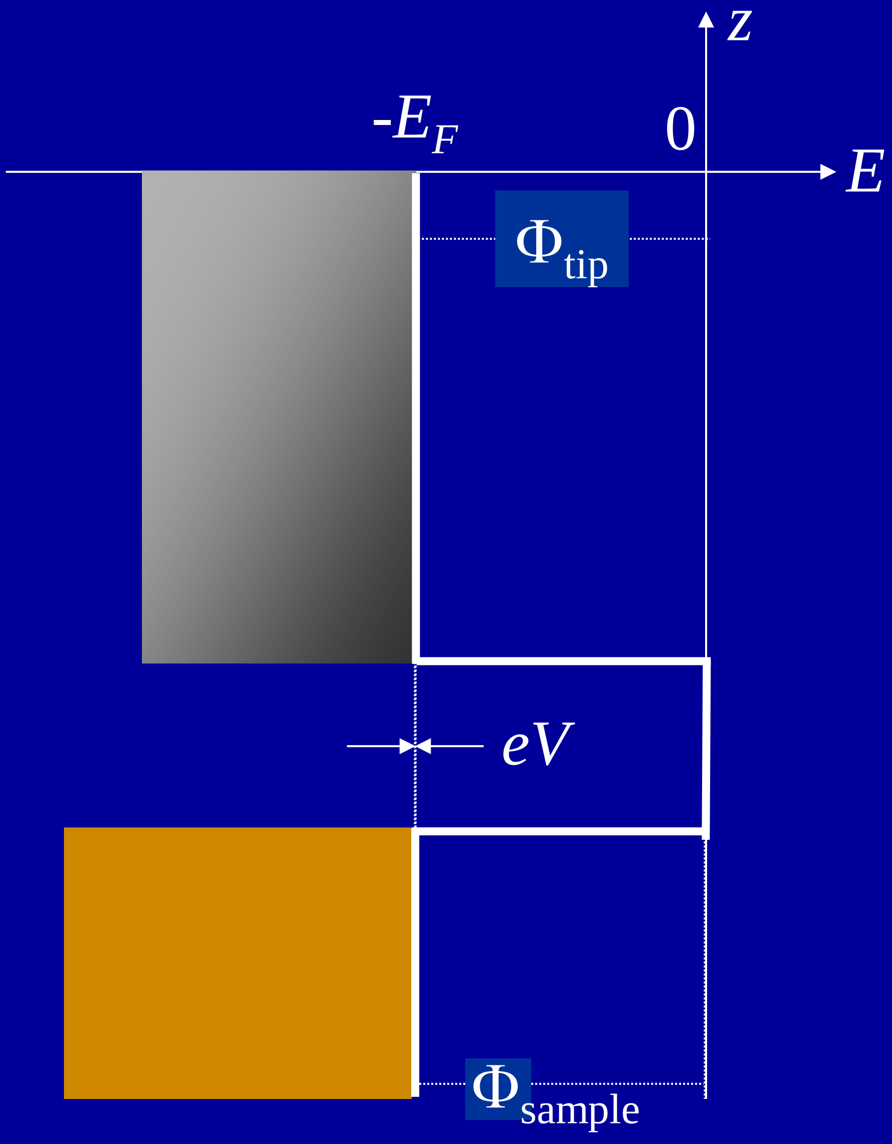


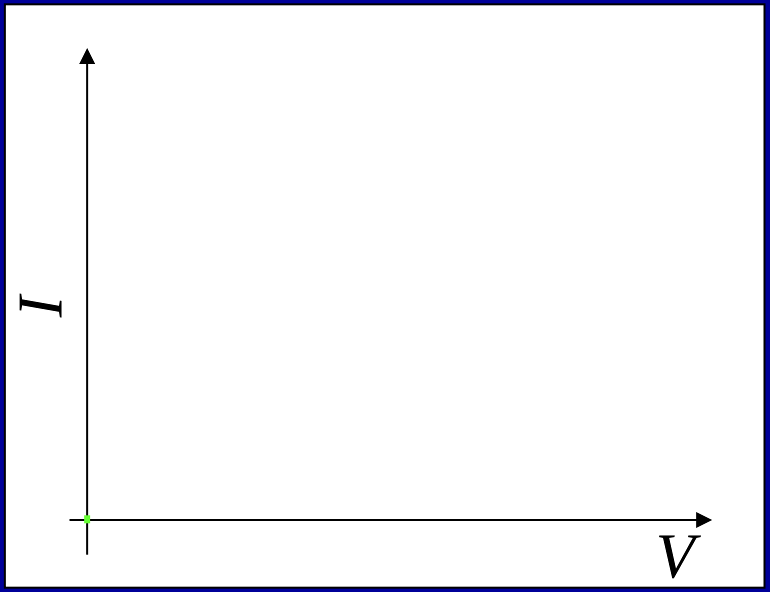


- Pentacene on thin layer of NaCl on Cu (111) for decoupling the molecular states from the electronic states of the metal surface (reducing Γ_s)
- Functionalization of tip changes the “active“ orbital

Repp et al., PRL 94,026803 (2005)

Elastic tunneling





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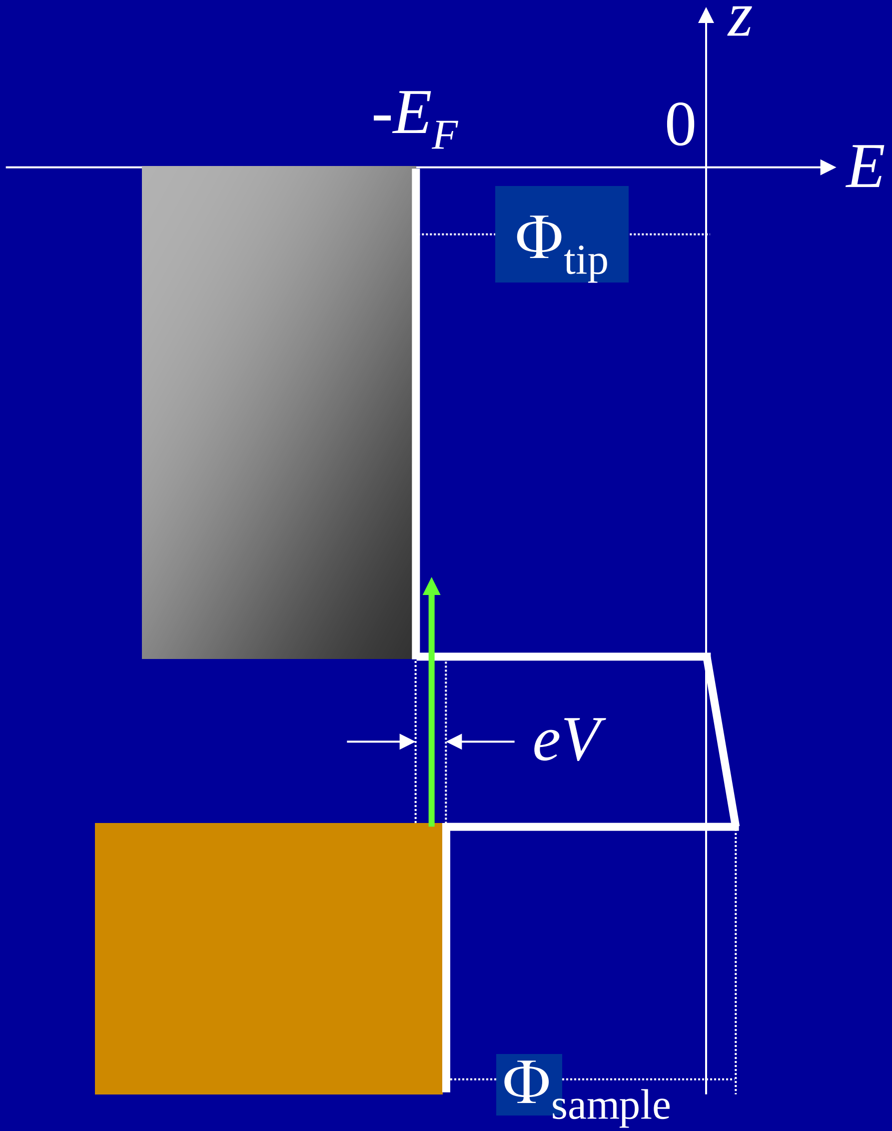
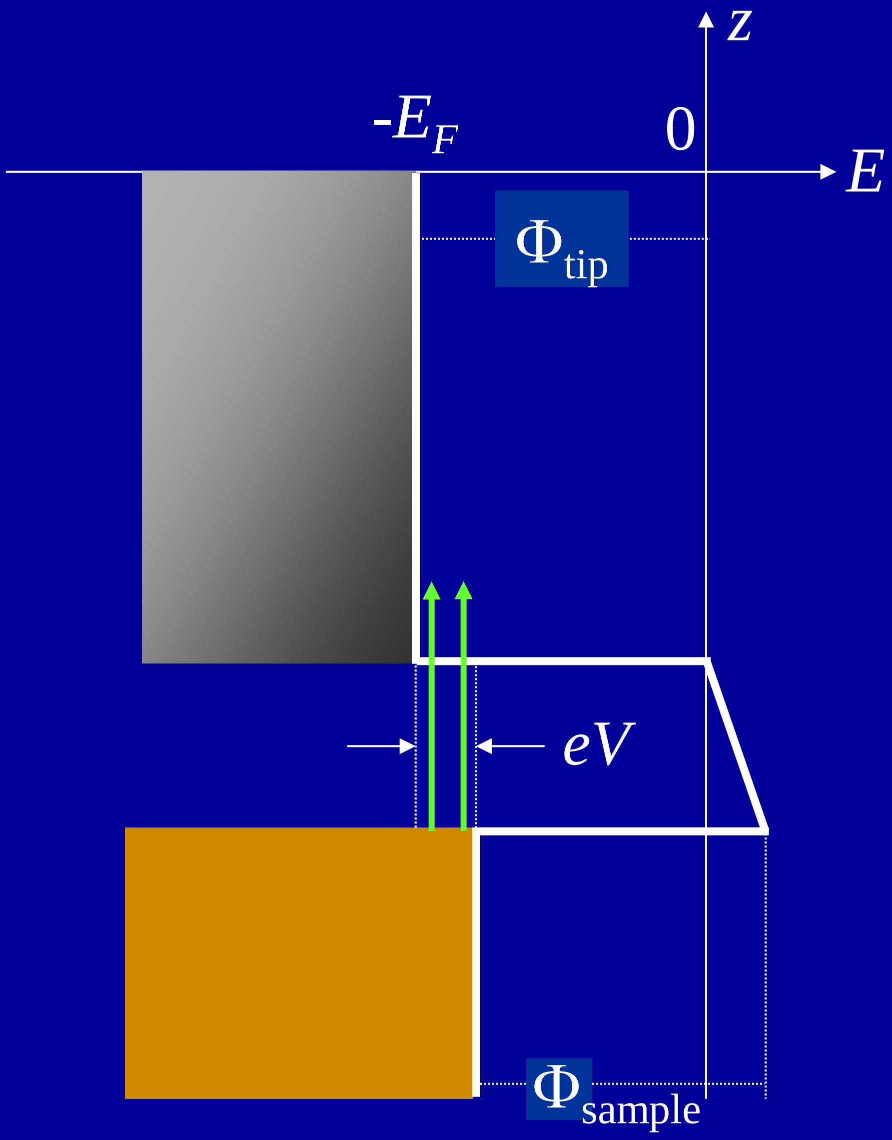
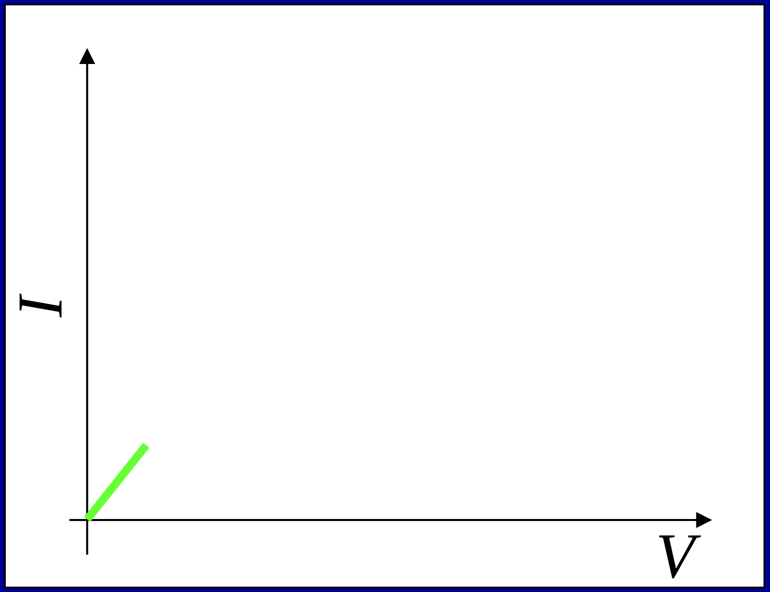
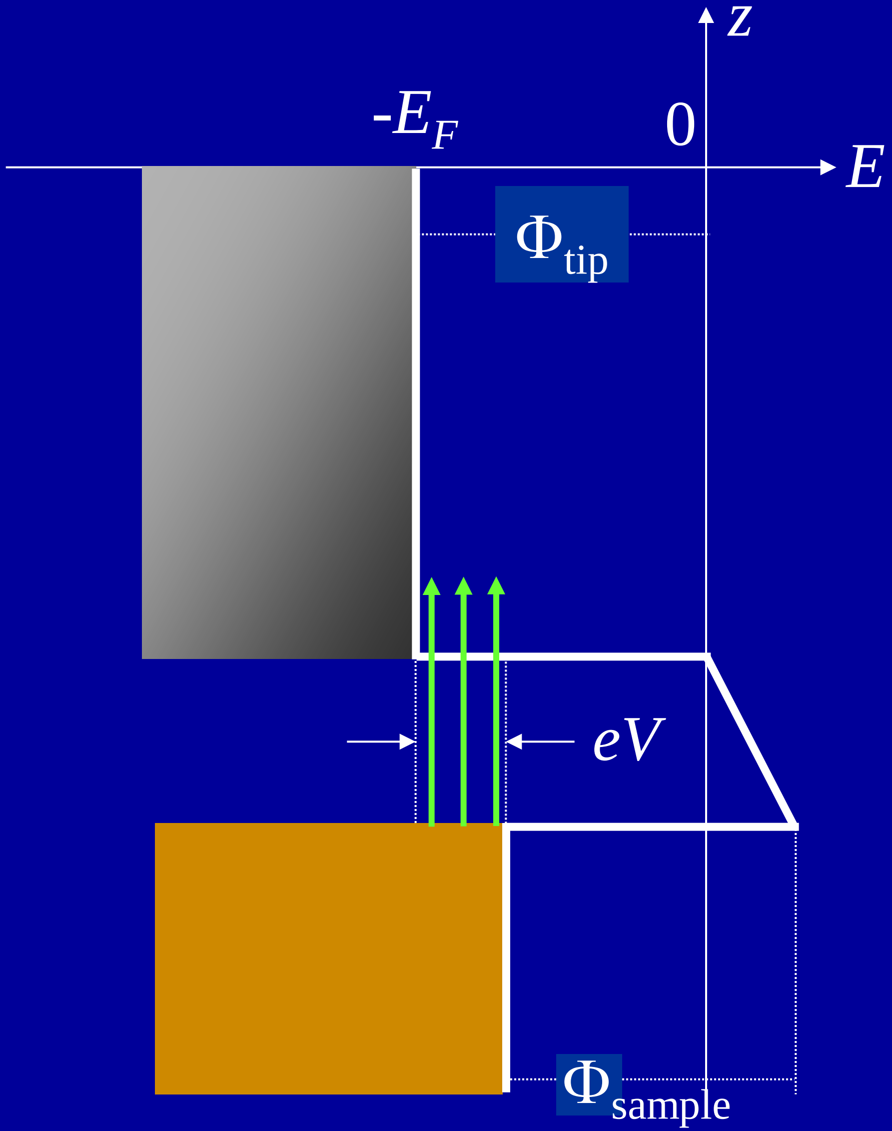


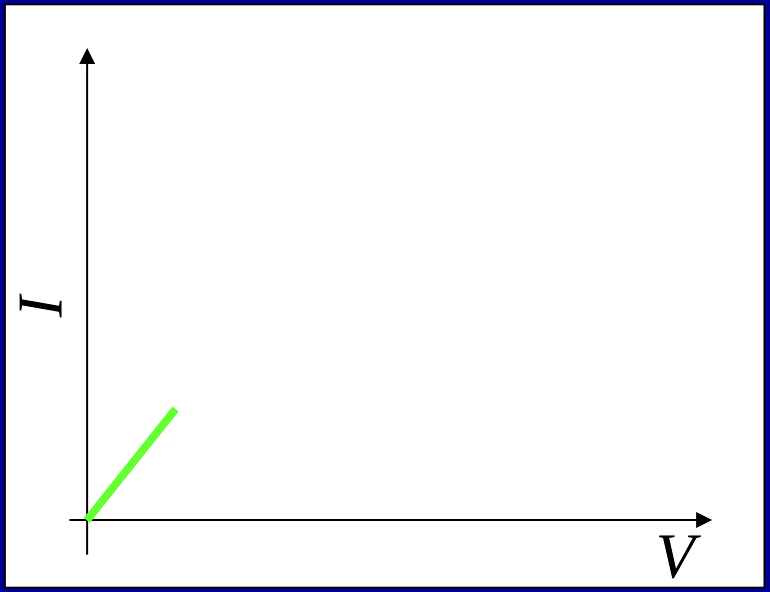
Elastic tunneling



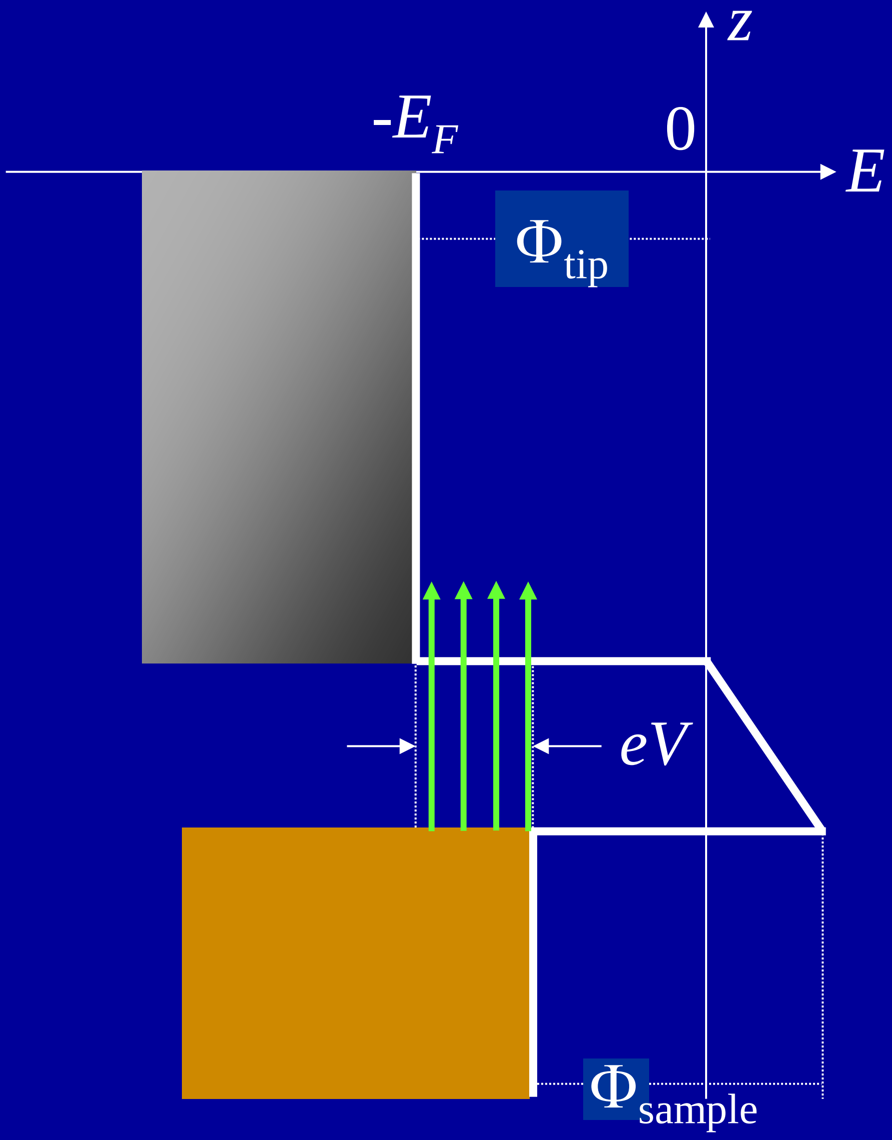


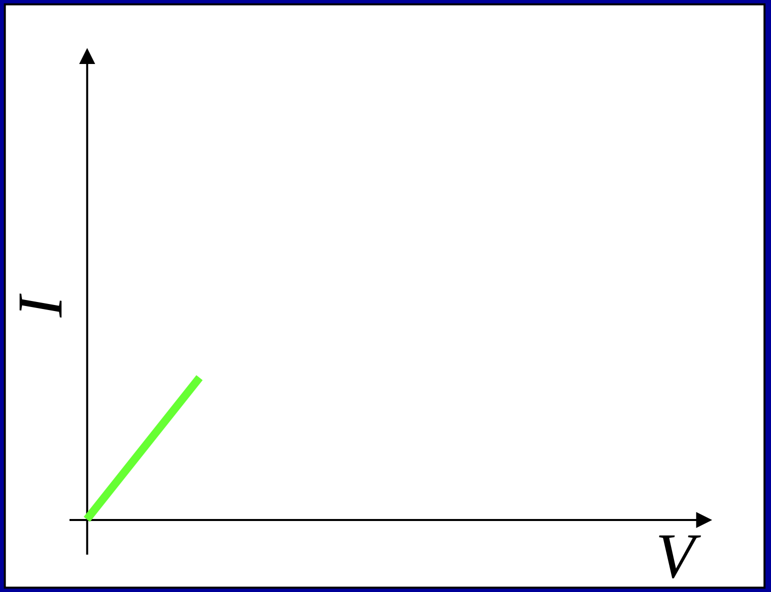
Elastic tunneling



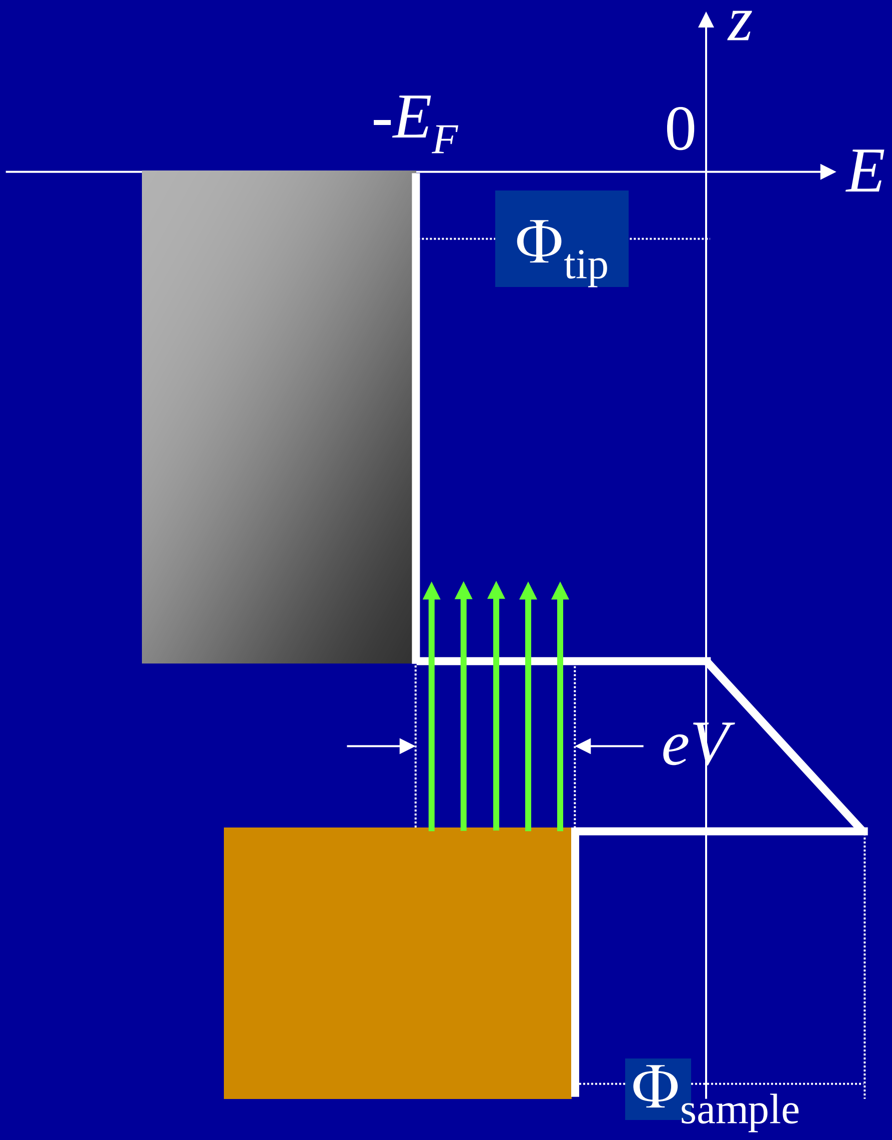


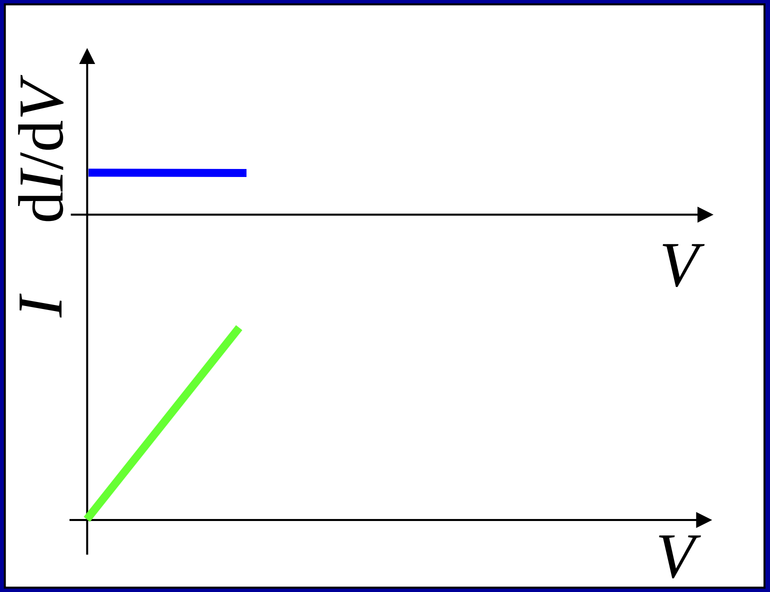
Elastic tunneling



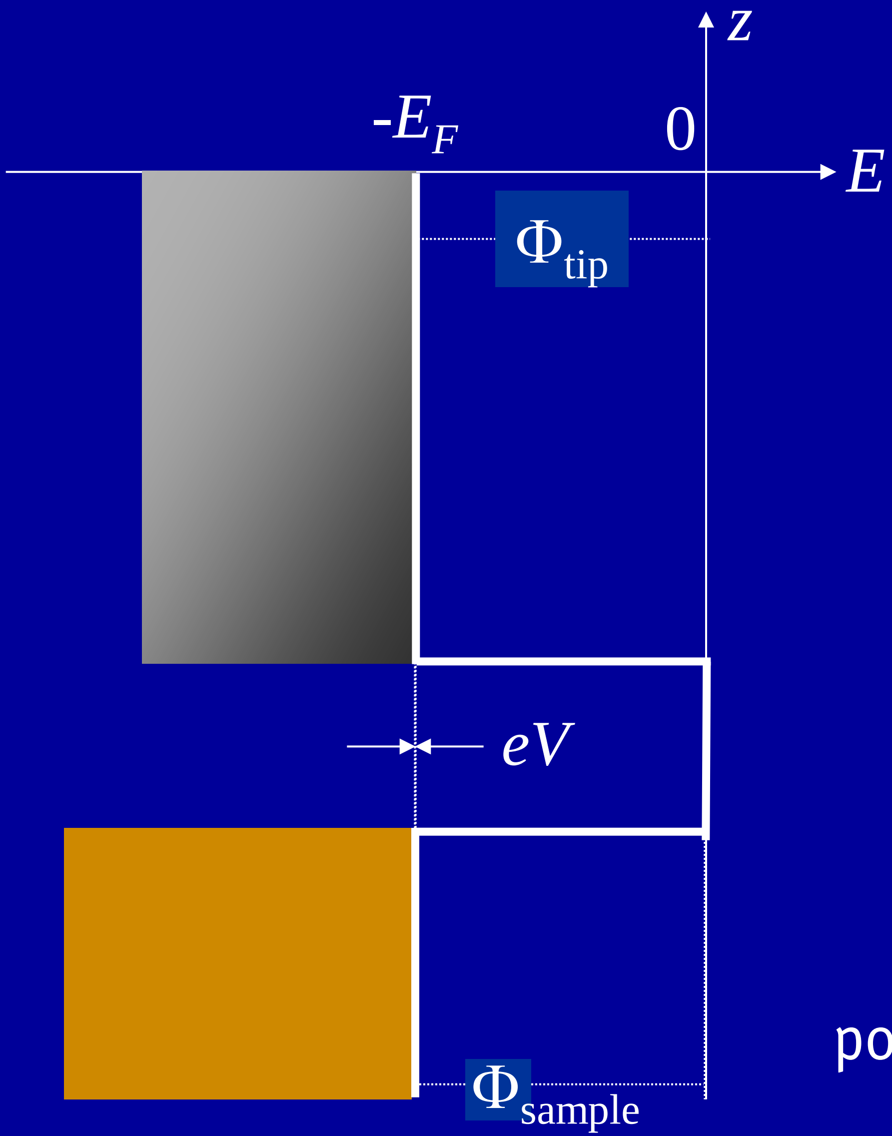


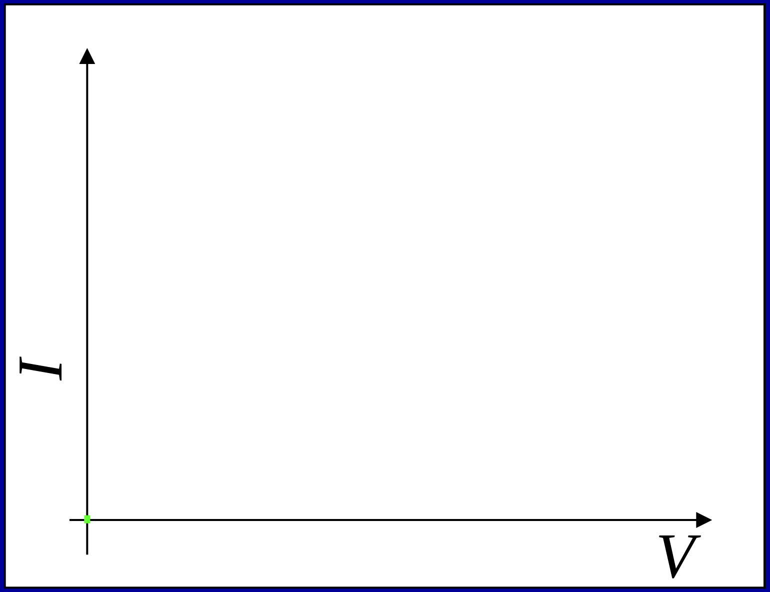
Elastic tunneling





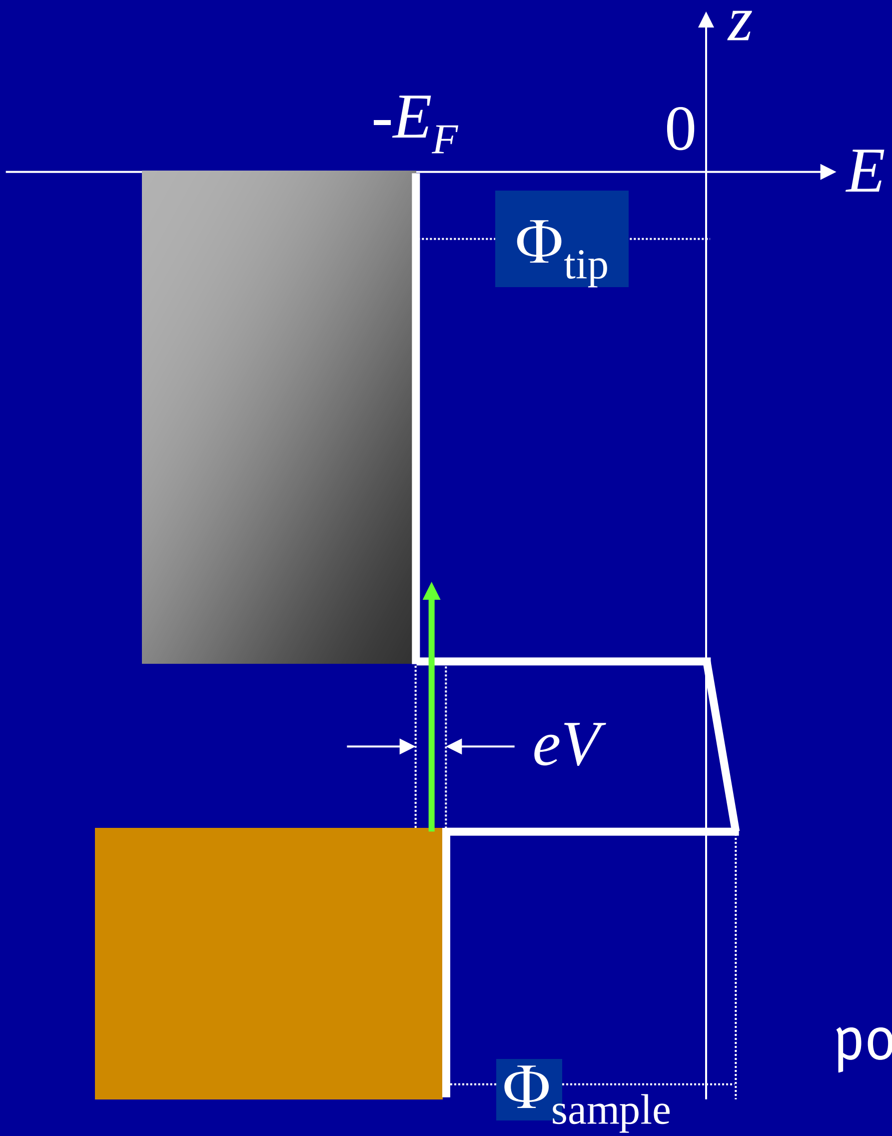
Inelastic tunneling

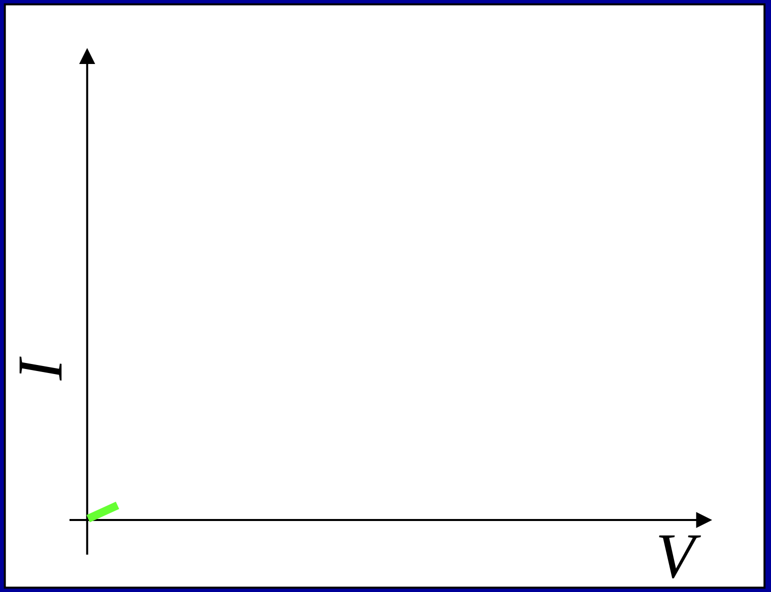




possibility of vibrational "channels"

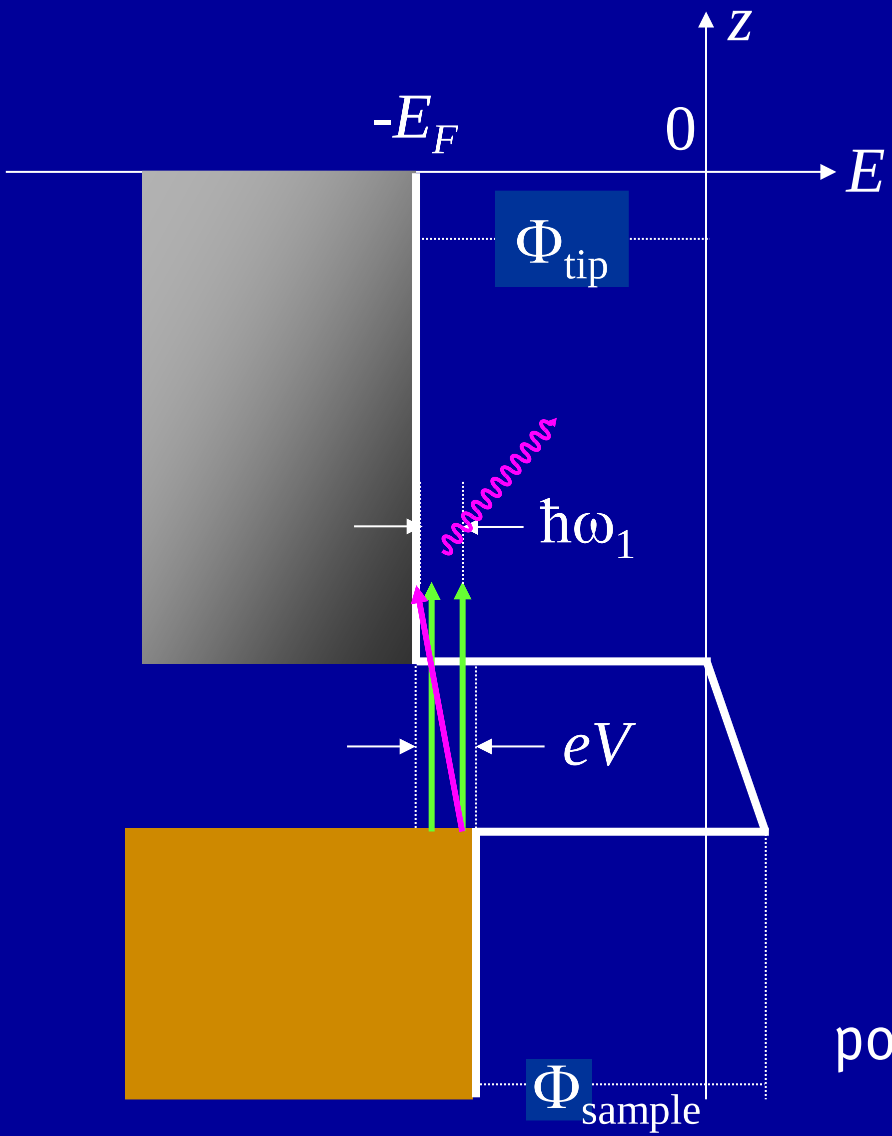
Inelastic tunneling

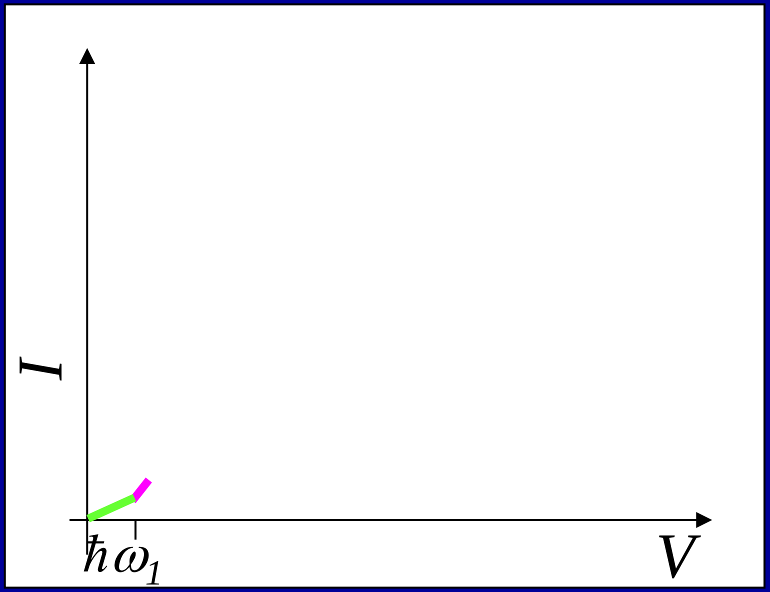




possibility of vibrational “channels”

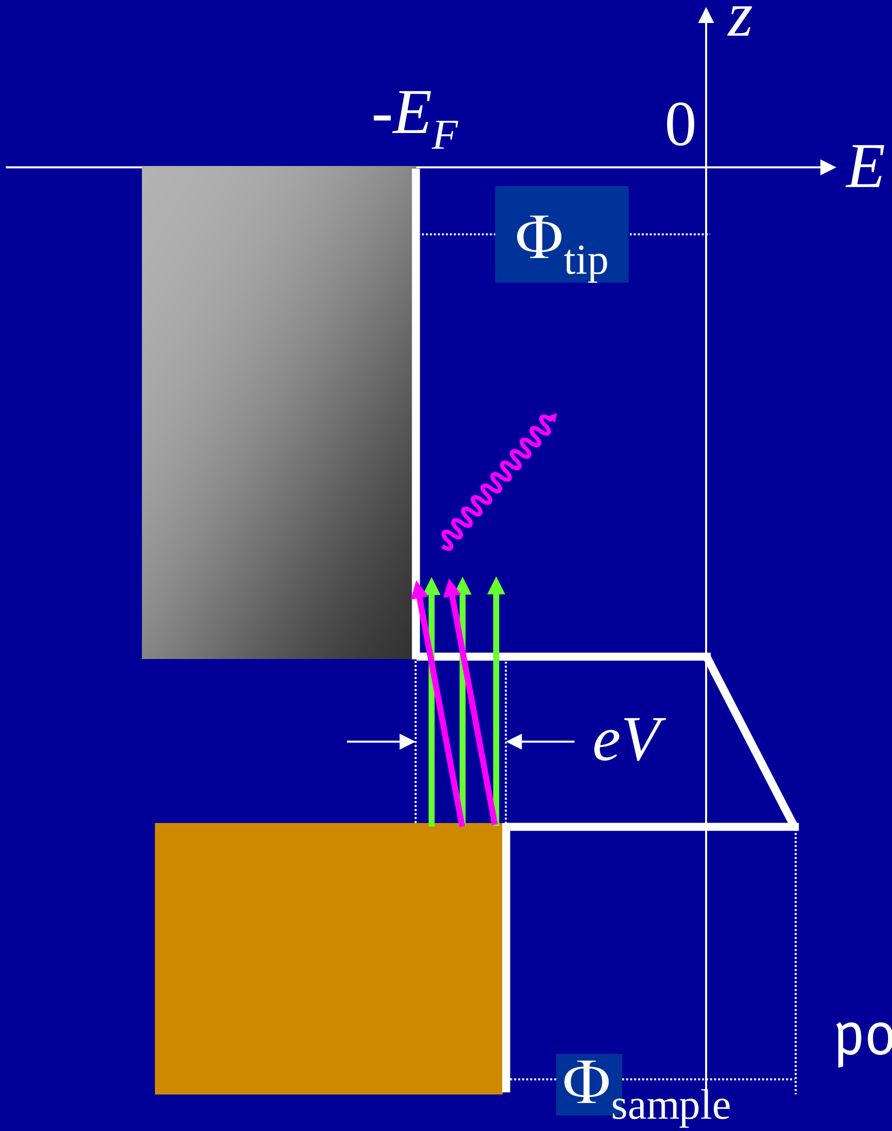
Inelastic tunneling

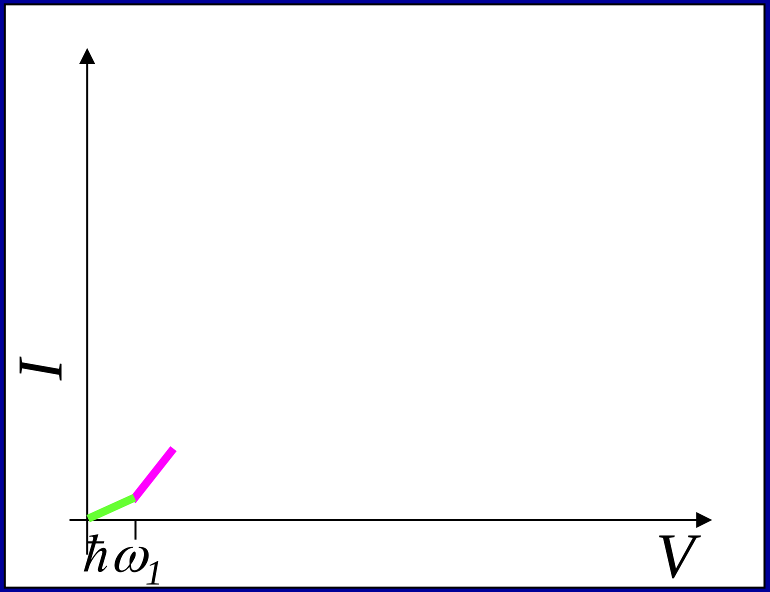




possibility of vibrational “channels”

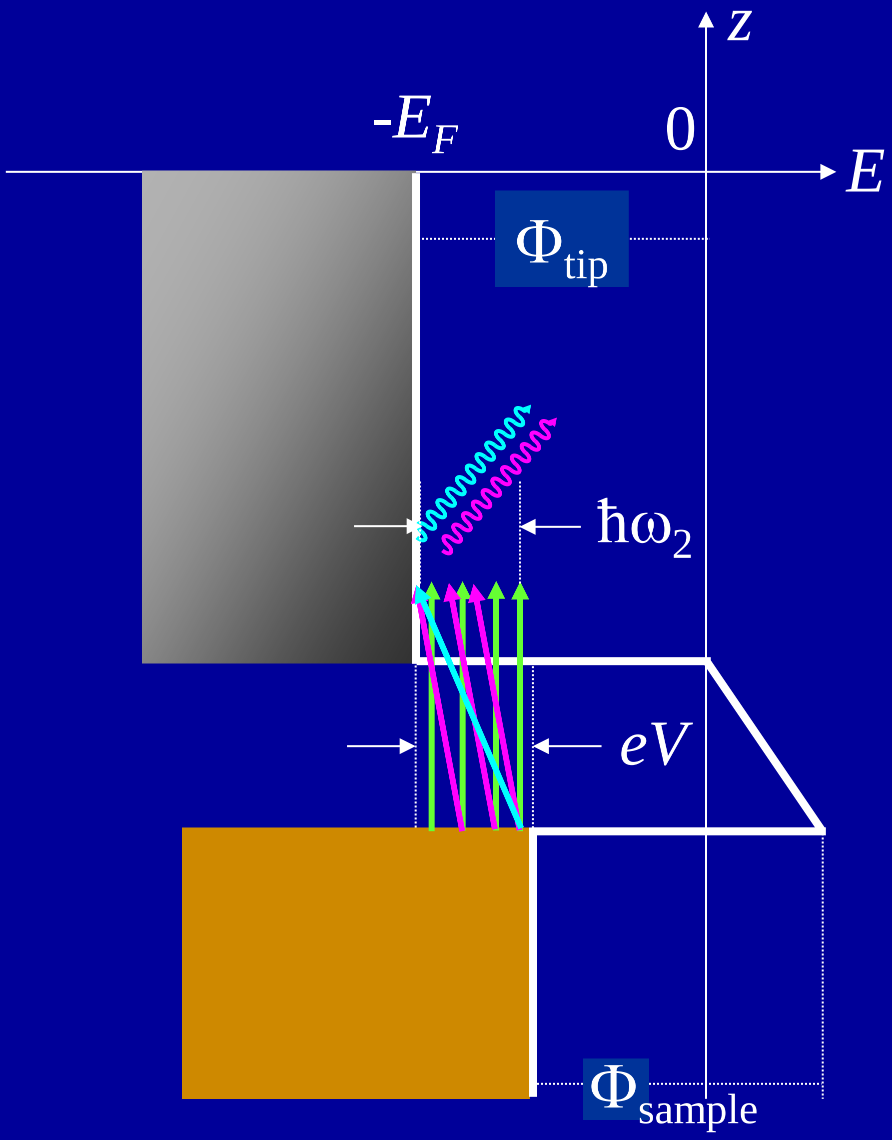
Inelastic tunneling

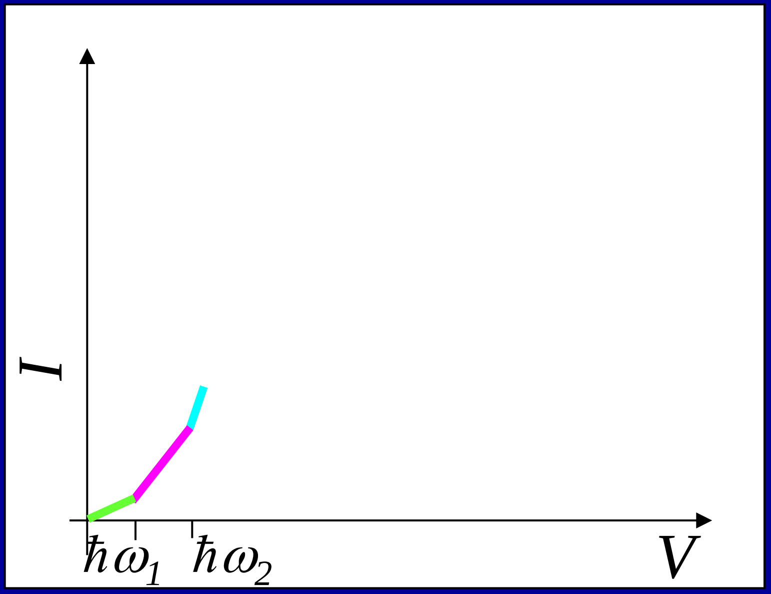




possibility of vibrational “channels”

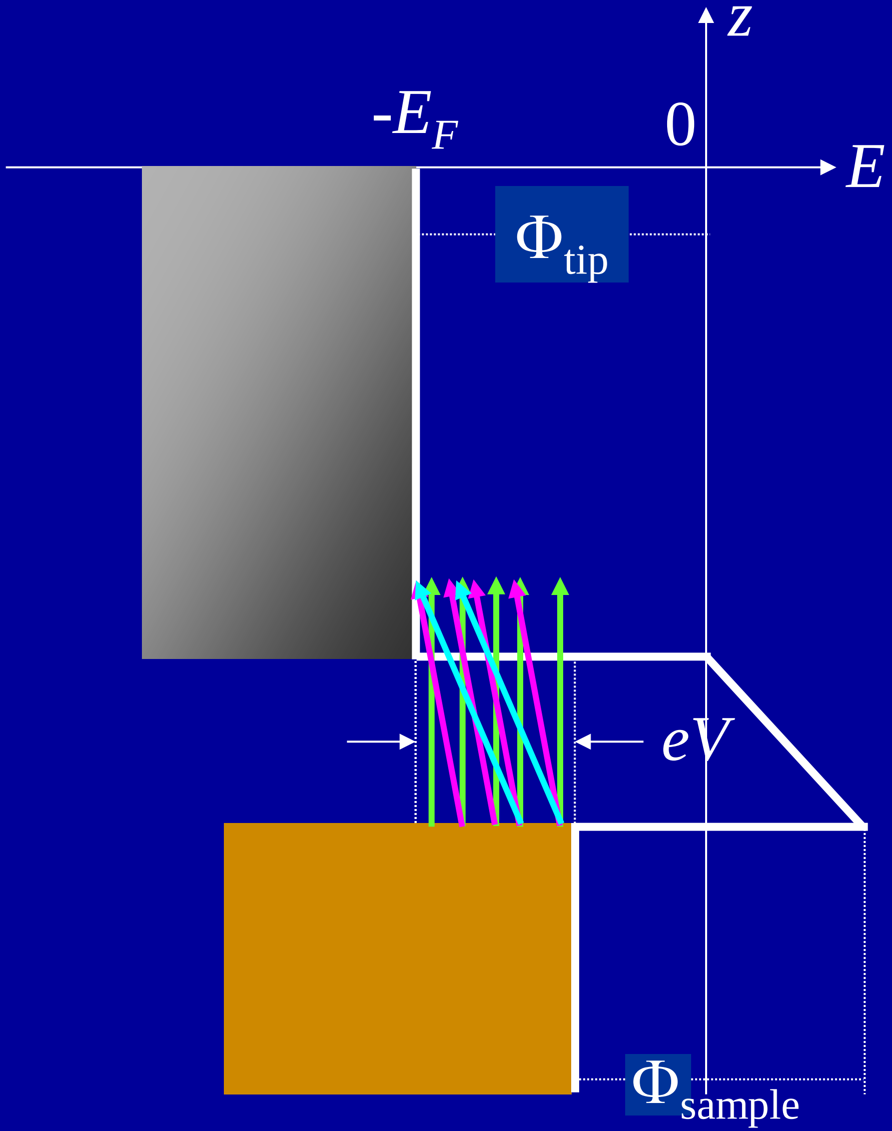
Inelastic tunneling

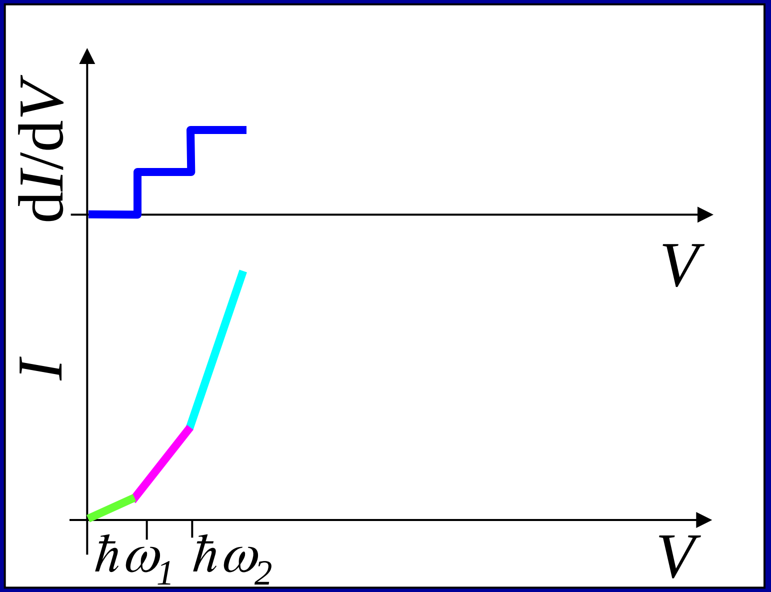




possibility of vibrational “channels”

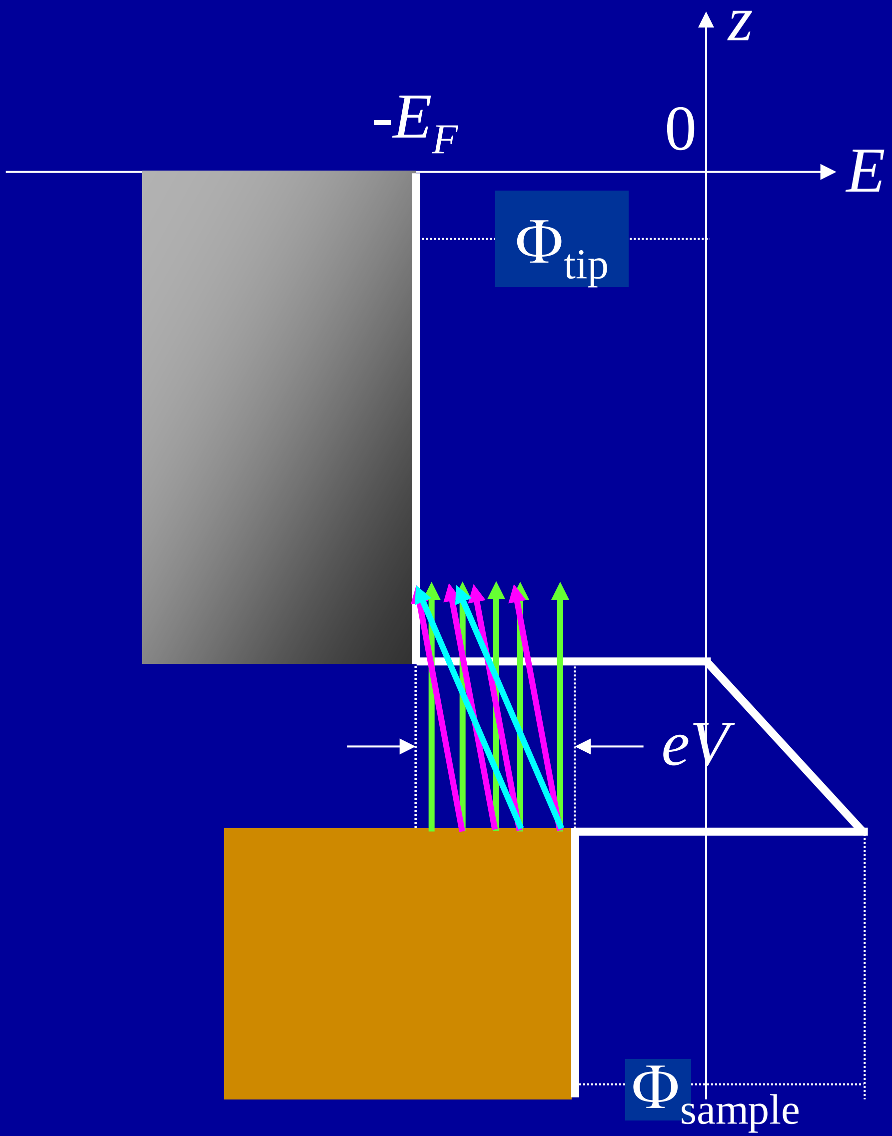
Inelastic tunneling

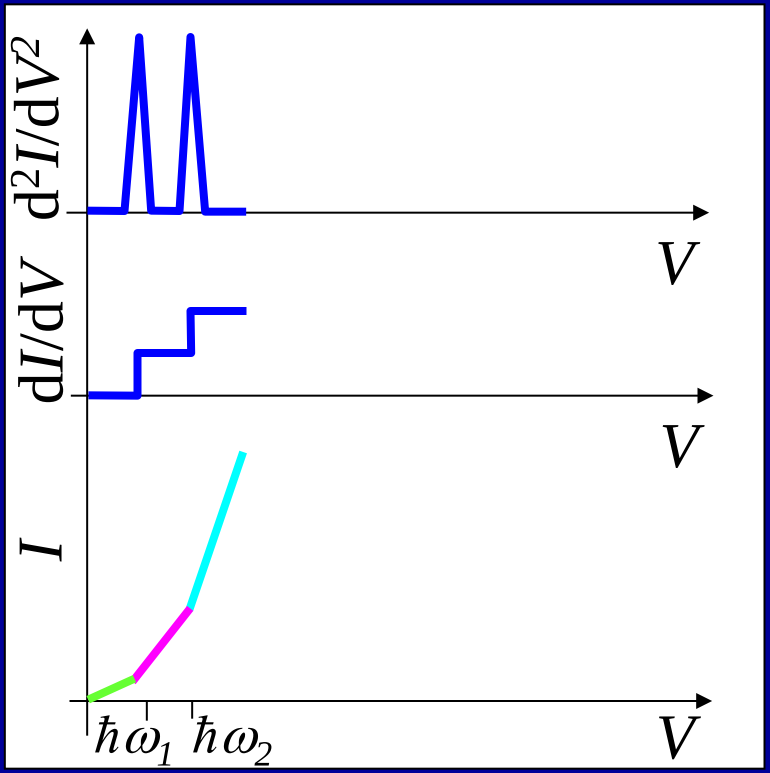




possibility of vibrational “channels”

Inelastic tunneling

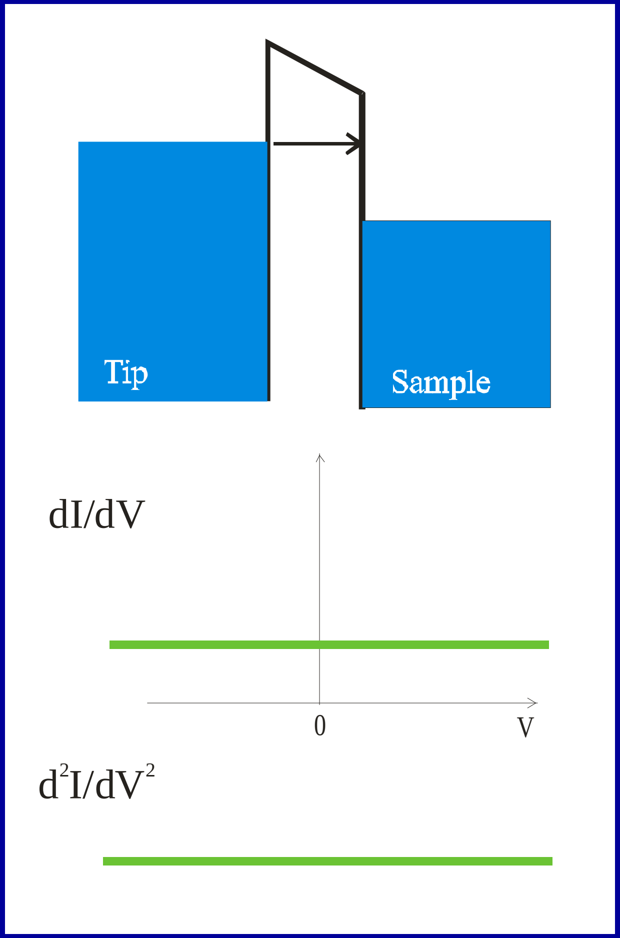


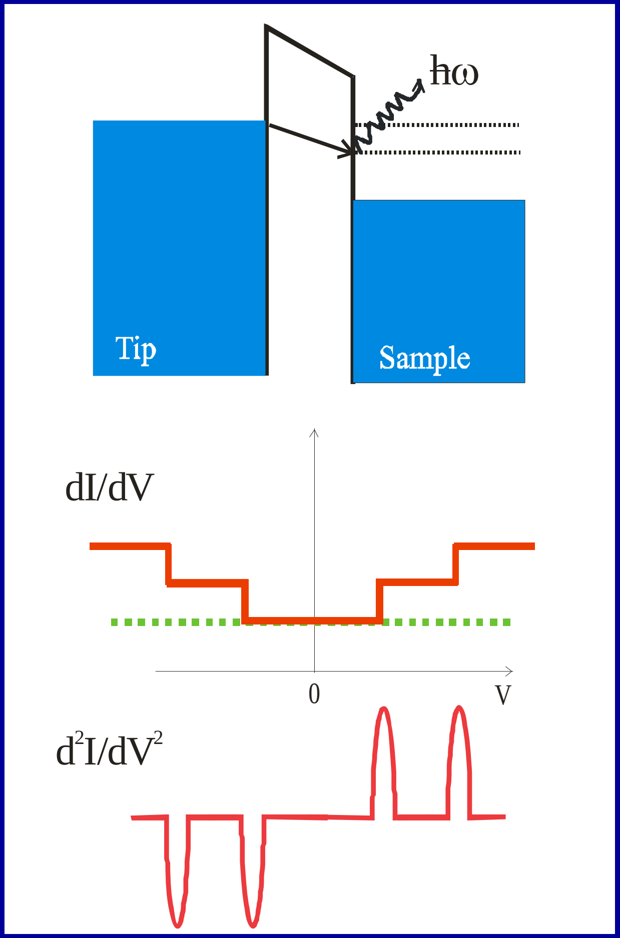


possibility of vibrational “channels”

Elastic tunneling

Inelastic Electron Tunneling Spectroscopy

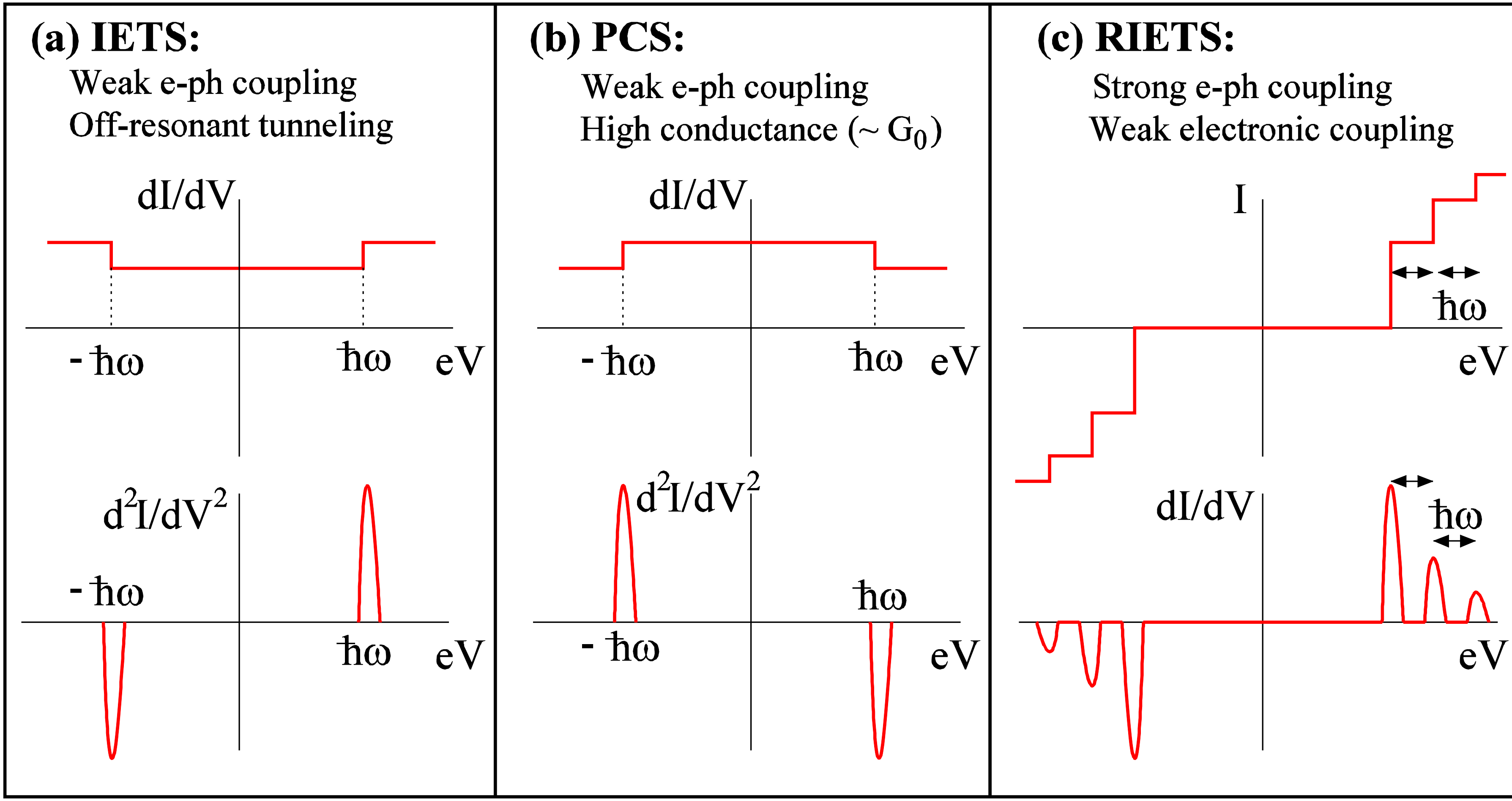




dI/dV ~LDOS

$d^2I/dV^2 \sim \nu DOS$

Reminder: 9.1 Signatures of vibrational modes



Measurement of LDOS by STS

1. Order perturbation theory

$$I = \frac{2me}{h} \int_{-\infty}^{\infty} [f(E_F - eV + \varepsilon) - f(E_F + \varepsilon)] \rho_S (E_F - eV + \varepsilon) \rho_T (E_F + \varepsilon) M_{\mu\nu} d\varepsilon$$

mit $\rho_{T,S}$ Density of states of tip and sample

$M_{\mu\nu}$ Tunnel matrix element: overlap of wave functions of tip and sample, depends on chemical nature and geometry

Für $M_{\mu\nu} = const.$ and low temperature it follows:

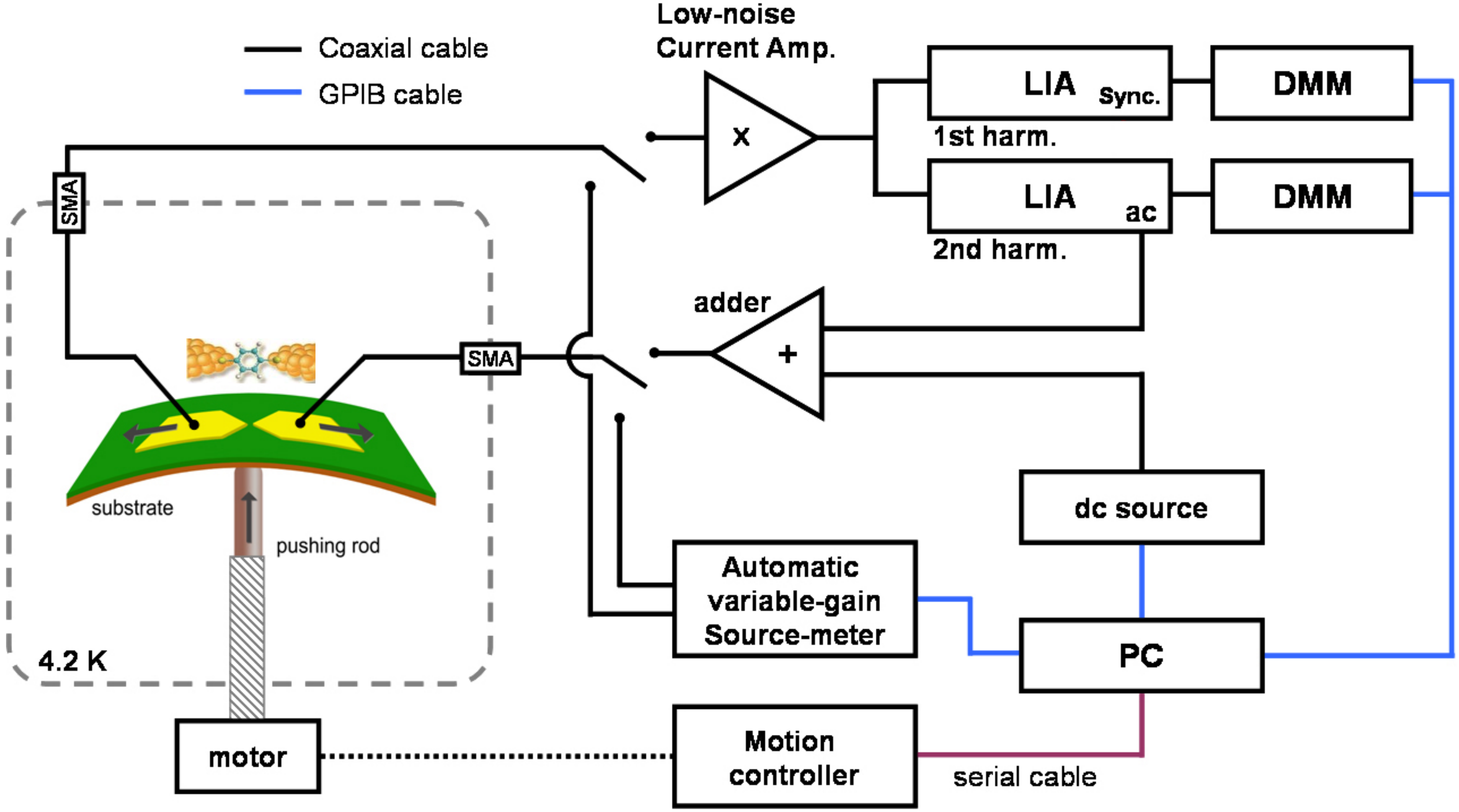
$$I \propto \int_0^{eV} \rho_s (E_F - eV + \varepsilon) \rho_T (E_F + \varepsilon) d\varepsilon$$

If $\rho_T = const.$:

$$\frac{dI}{dV} \propto \rho_S$$

Local Density of States (LDOS) of the sample

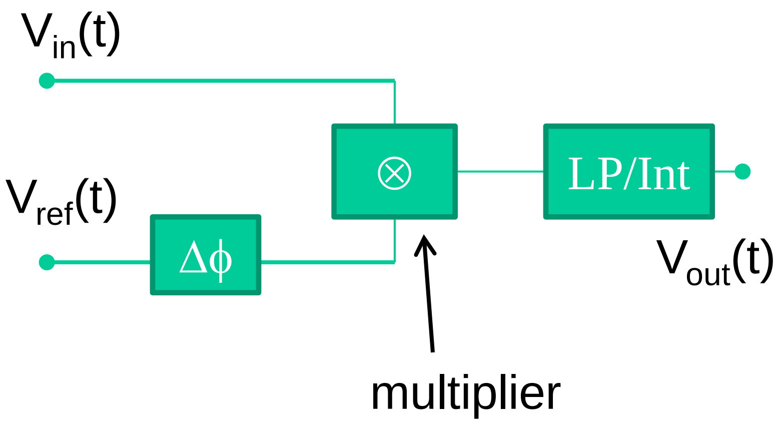
9.3 Measurement of IV, dI/dV and d^2I/dV^2 of molcelular contacts



Source: Y. Kim, A. Karimi, D. Weber

11.3 Lock-in technique

Purpose: low-noise amplification by phase sensitive detection in a narrow frequency band and cross-correlation (narrow band pass filter)



<u>Main ingredients of a lock-in amplifier:</u>

- Preamplifier for input signal $V_{in}(t)$
- Channel for reference signal (internal or external)
- Phase shifting unit (Δϕ), applies phase between signal and reference
- Mixer (multiplier)
- Low pass for integrating the cross correlation

$$V_{out}(t) = \frac{1}{T}\int_{t-T}^{t} \sin[2\pi f_{ref} \cdot s + \Delta\phi] V_{in}(s) ds$$

Cross correlation: $V_{out}(t)$ non-zero if $f_{in} = f_{ref}$
=> frequency sensitive, suppression of noise at other frequencies

For sinusoidal signals: $V_{out}(t) \propto V_{in}(t) \cos(\Delta\phi)$

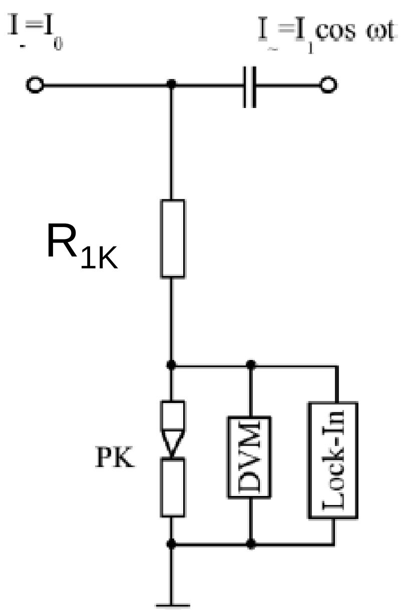
11.3 Lock-in Technique for measuring dV/dI and d^2V/dI^2

Superposition of DC signal and harmonic AC signal : $I = I_0 + I_1 \cos\omega t$

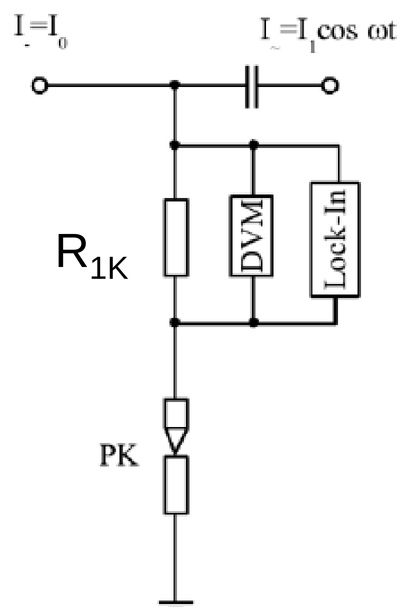
$$V_{PK}(I) = V_{PK}(I_0) + \underbrace{\left.\frac{\mathrm{d}V_{PK}}{\mathrm{d}I}\right|_{I_0}}_{V_1^{PK}} I_1 \cos\omega t + \frac{1}{4}\left.\frac{\mathrm{d}^2V_{PK}}{\mathrm{d}I^2}\right|_{I_0} I_1^2 \cos 2\omega t + \cdots .$$

1st harmonic $\propto$ dV/dI

2nd harmonic $\propto$ d^2V/dI^2



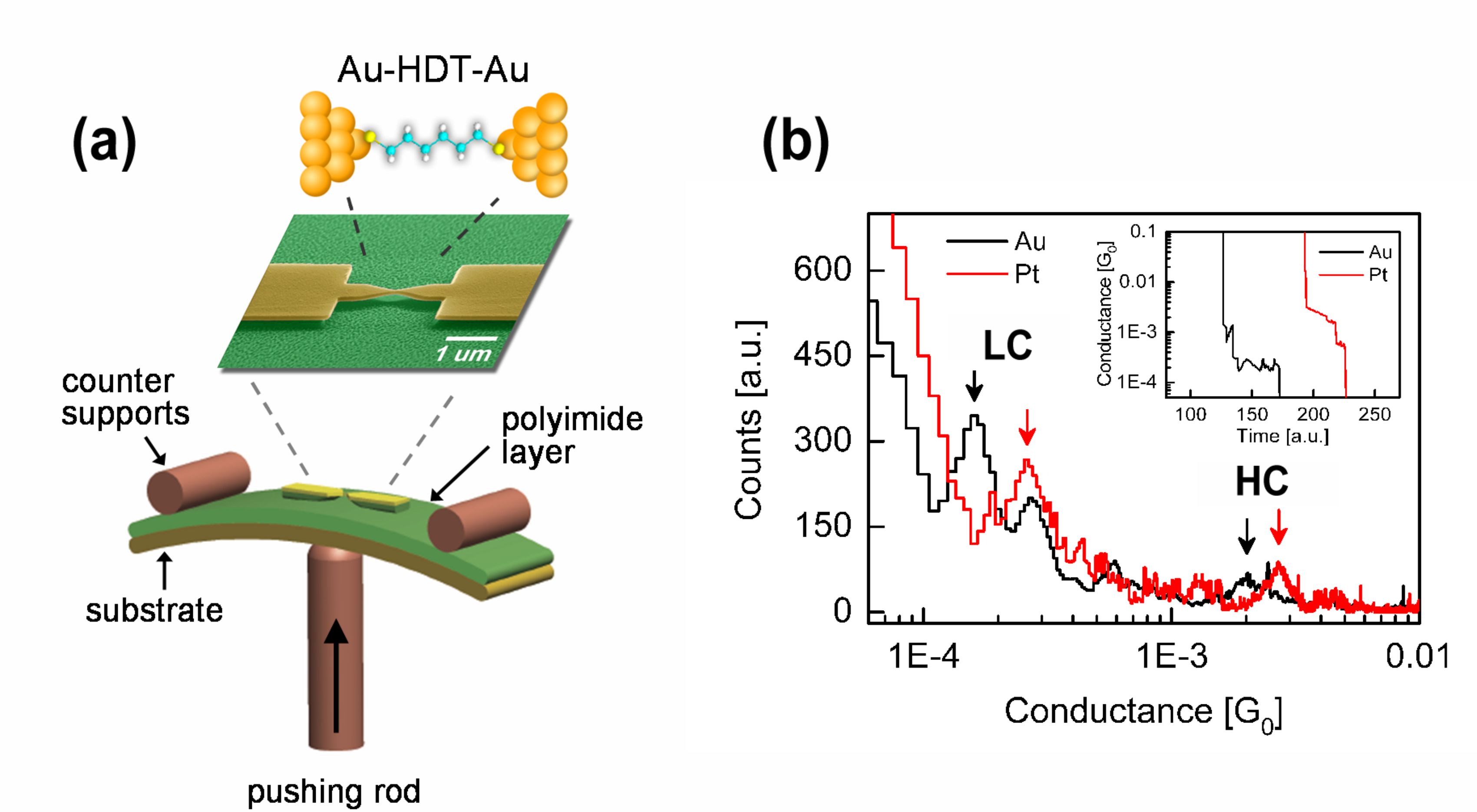
Measurement of V_{PK}



Calibration

Source: Th. Brugger, Diploma Thesis, Uni Karlsruhe

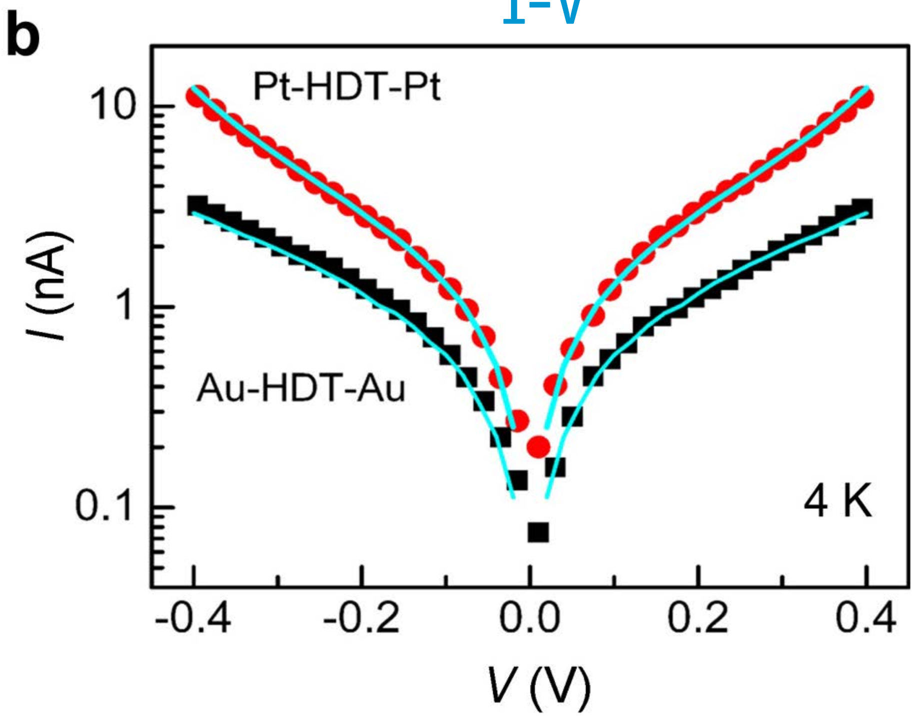
MCBJ at low temperature: Opening Traces and Conductance Histograms



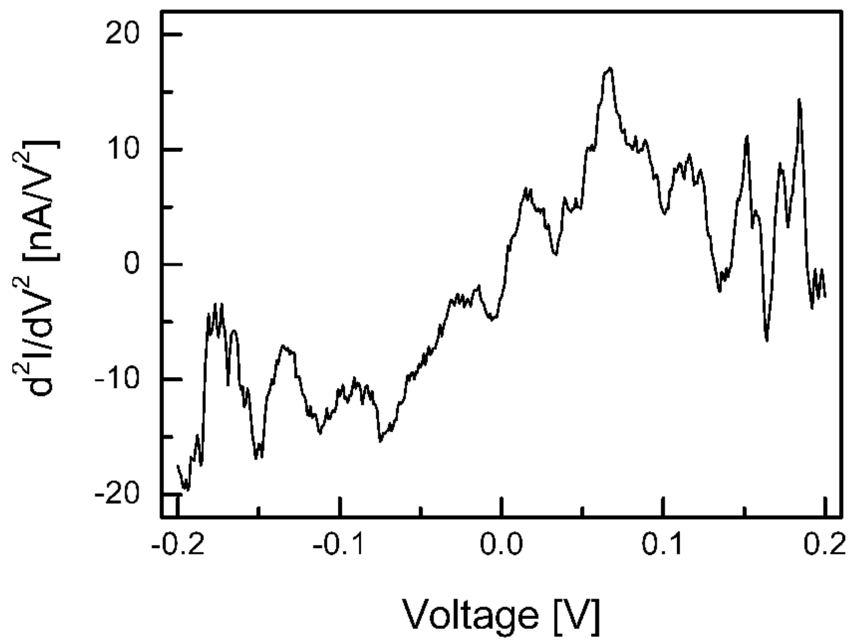
Y. Kim, F. Strigl, H.-F. Pernau, H. Song, T. Lee, E. Scheer, Phys. Rev. Lett. **106**, 196804 (2011)

Examples:
I-Vs and IETS of HDT@ 4.2 K

I-V



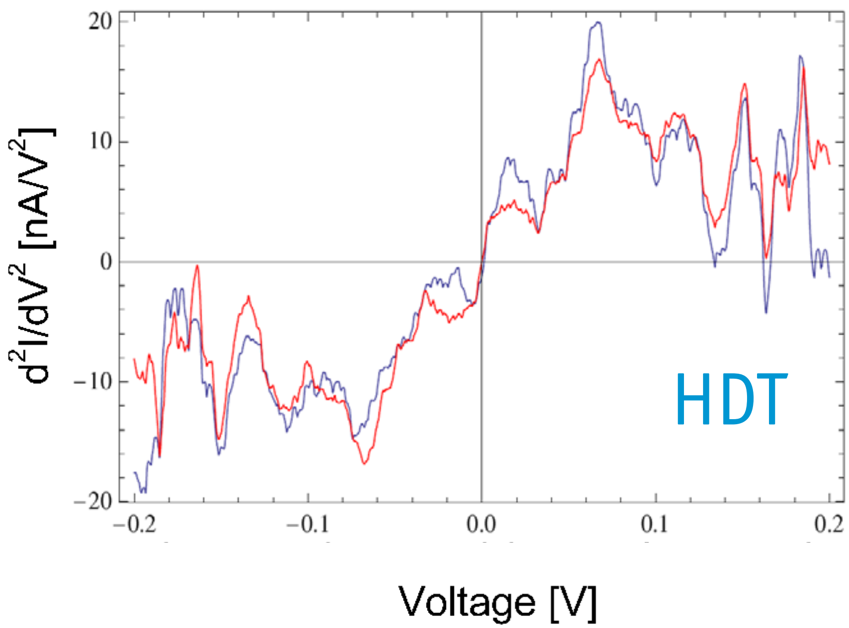
d^2I/dV^2

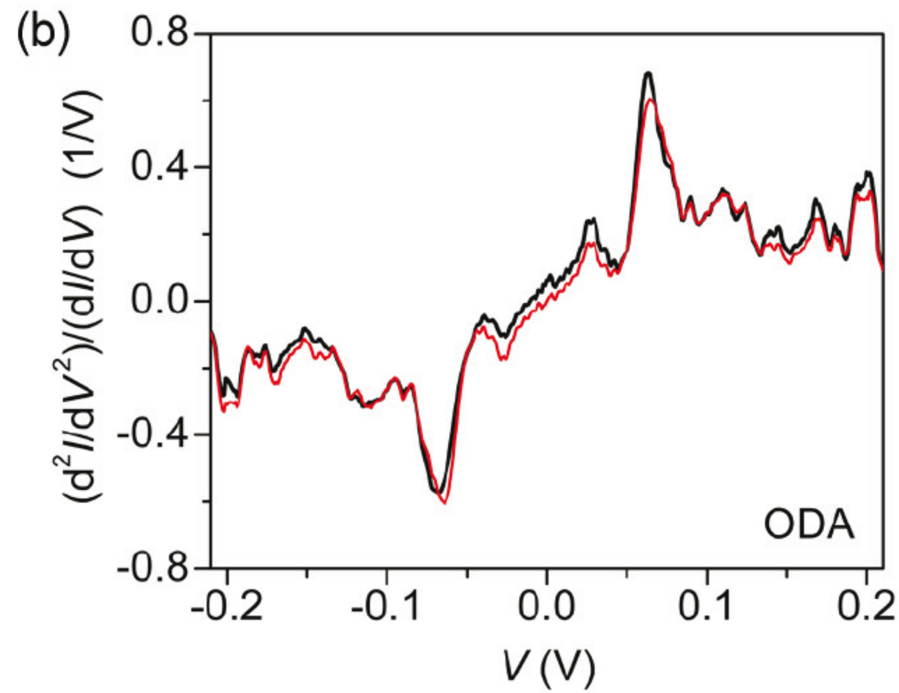


Au-HDT-Au: $\Gamma_{L,R}$ = 120 meV, E_0 = 2.35 eV

Pt-HDT-Pt: $\Gamma_{L,R}$ = 110 meV, E_0 = 1.93 eV

Symmetry of IETS spectra





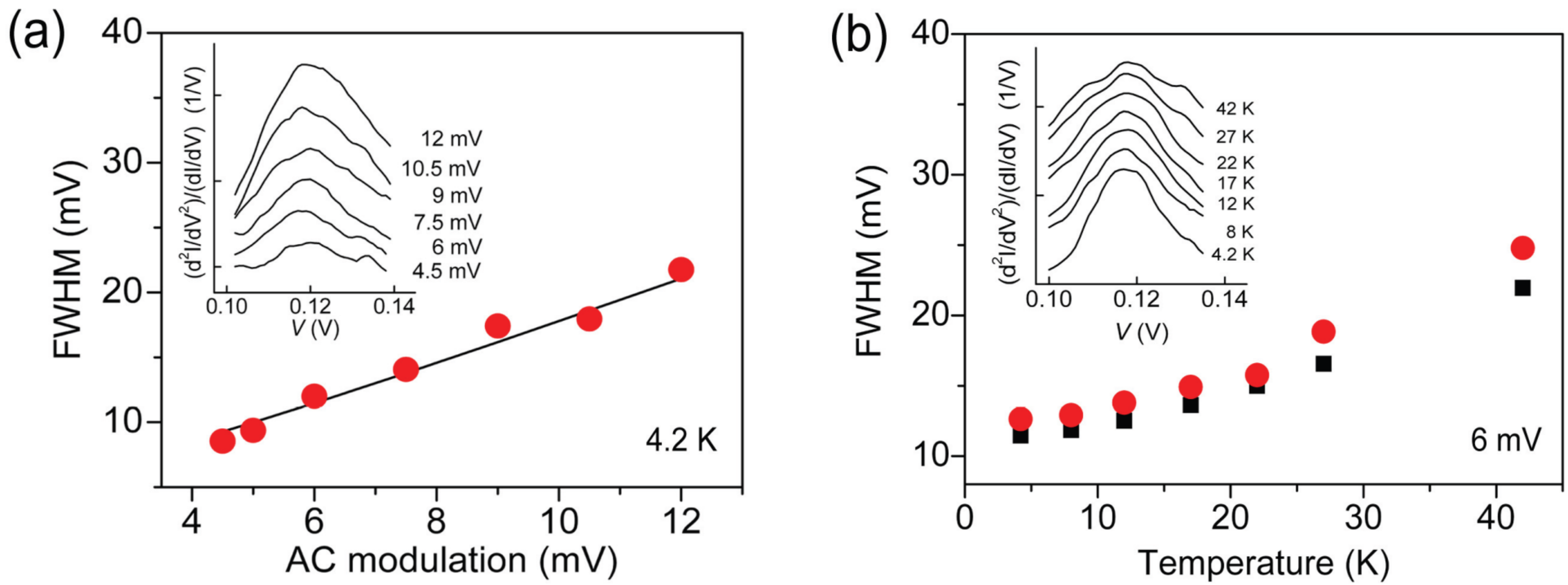
Black/blue: as measured

Red: symmetrized signal

$$y = [f(x) - f(-x)]/2$$

Y. Kim, Th. Hellmuth, M. Bürkle, F. Pauly, E. Scheer, ACS Nano **5**, 4104 (2011).

Linewidth broadening of IETS of ODA



Experimental linewidth W_{exp} given by intrinsic linewidth W_I, thermal broadening k_BT and modulation voltage V_{ac} :

$$W_{exp} = [(5.4k_BT)^2 + (1.7V_{ac})^2 + (W_I)^2]^{1/2}$$

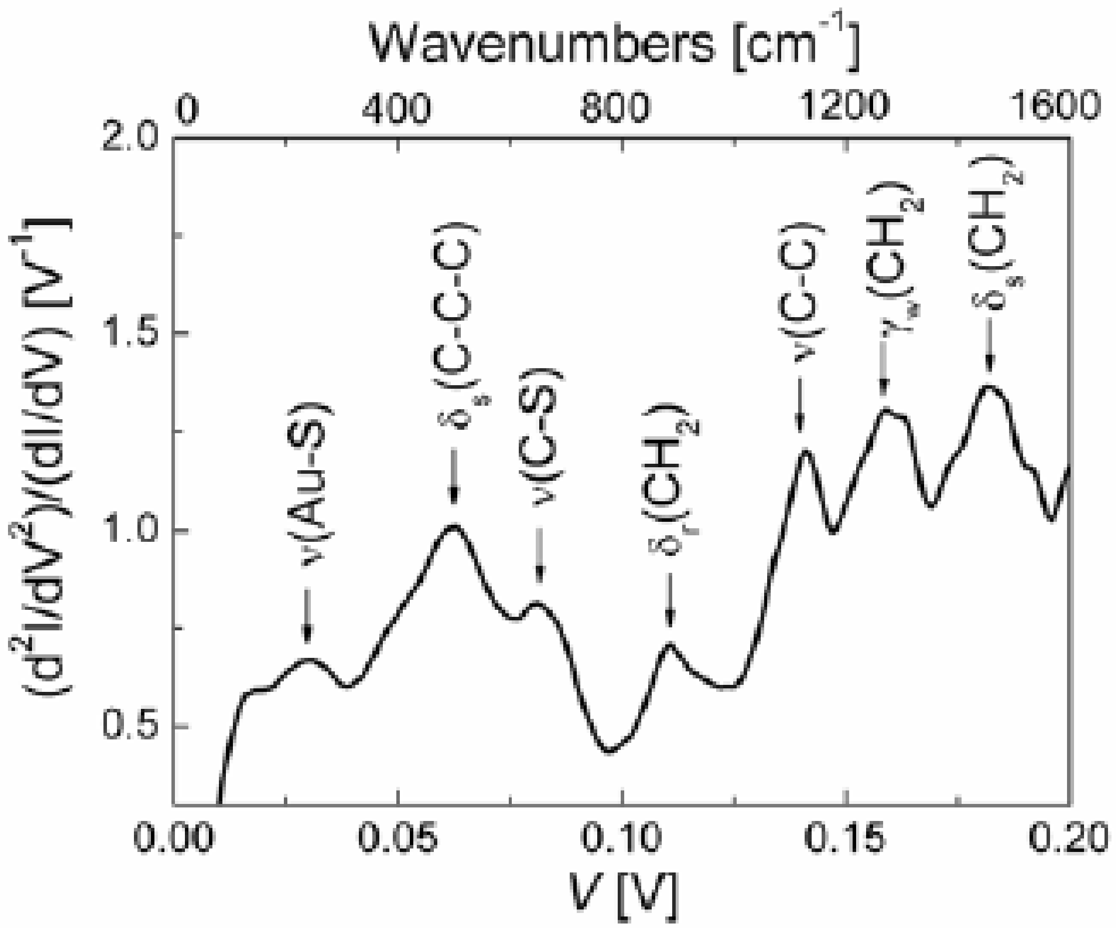
a) V dependence at fixed T: black line: linear fit ->$W_I = 4.9 \pm 0.8$ meV

b) T dependence at fixed V_{ac}: black squares: theoretical expectation, red dots: experimental findings

T = 4K: $5.4k_BT = 1.8$meV

Assignment of modes

Vibrational modes of free molecules known from Raman spectroscopy



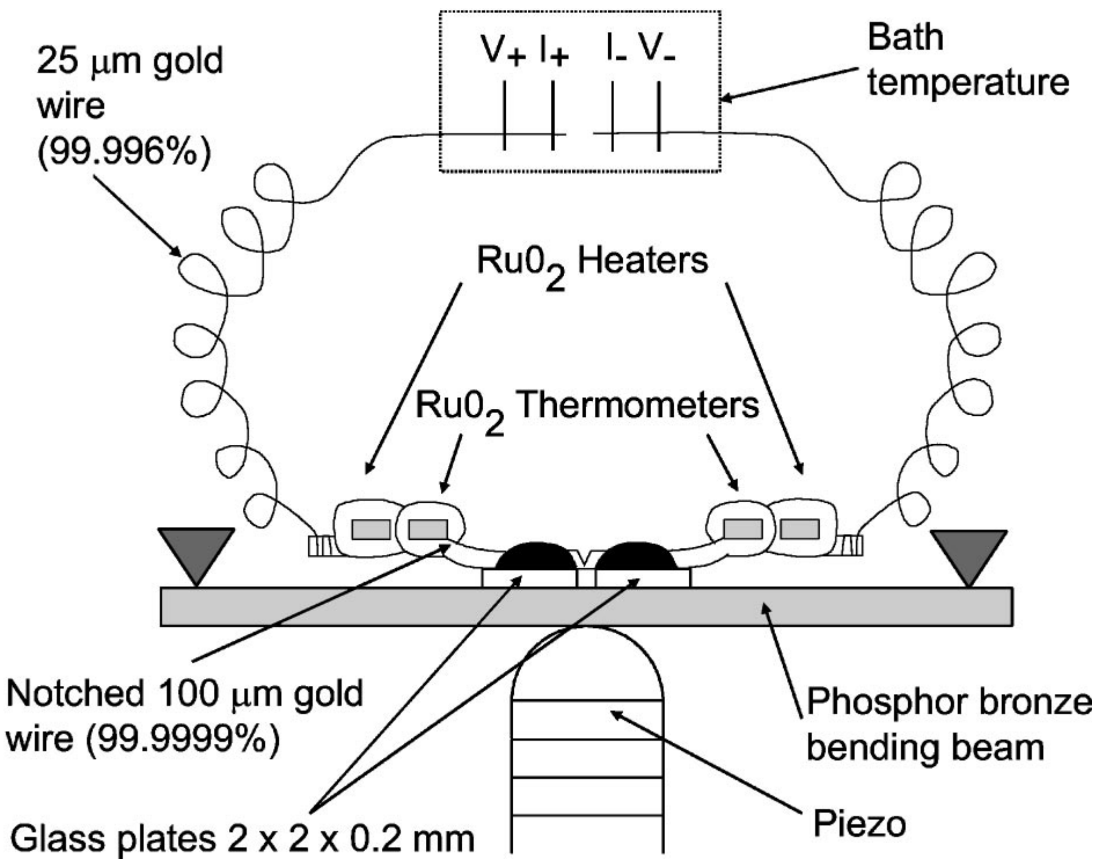
-> For molecules in junctions: Theory required!

Example HDT/Au

11.4 Measuring thermopower of metallic contacts (with MCBJ)

Thermopower (Seebeck coefficient) $S = V_{th}/\Delta T$

$T_{base} = 4.2$ K



Typical values

$\Delta T \sim 0.1\text{-}1$K

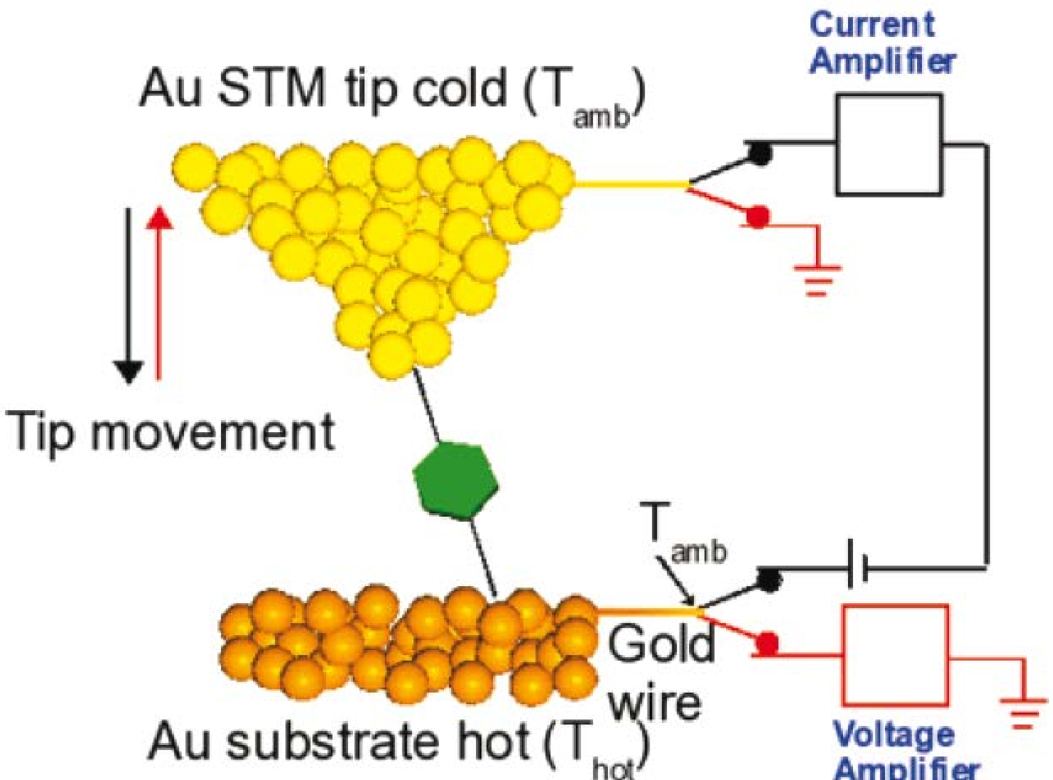
$V_{th} \sim$ nV - µV

-> requires 2 well-calibrated (resistive) thermometers and high-resolution voltage measurement (e.g. SQUID) or switching to same DVM at low T (avoiding thermopower contributions from wiring)

11.4 Measuring thermopower of molecular contacts with STM

Thermopower (Seebeck coefficient) $S = V_{th}/\Delta T$

$T_{base} = 300$ K



Step 0
Stabilize $\Delta T = T_{hot} - T_{amb}$
Step 1: V bias, I-meas. while approaching until $G = 0.1\ G_0$
Step 2: V bias = 0, V_{th} –meas. while withdrawing

Typical values
$\Delta T \sim 10$ - 30 K
$V_{th} \sim$ µV - mV

K. Baheti, J.A. Malen, P. Doak, P. Reddy, S.-Y. Jang, T. Don Tilley, Arun Majumdar, and R. A. Segalman, Nano Lett. ***8****, 715 (2008).*

11.5 Shot noise in atomic contacts

R. Cron, M. Goffman, D. Esteve and C. Urbina, Phys. Rev. Lett. ***86,*** *4104 (2001)*

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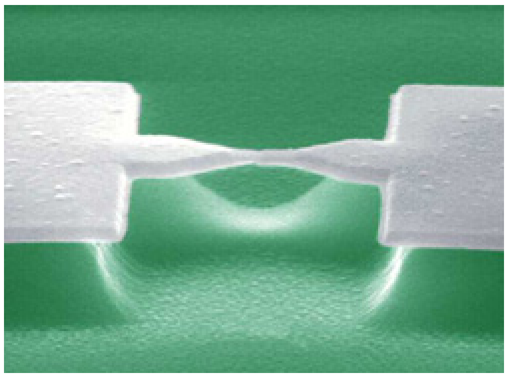
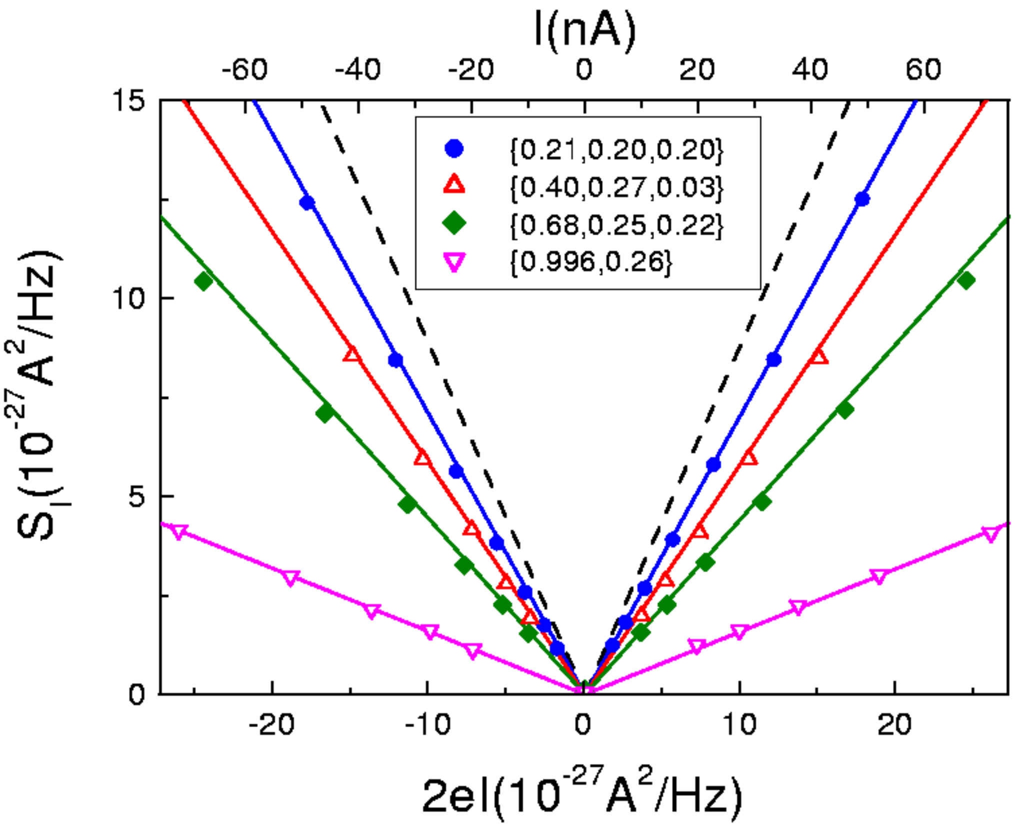
30 April 2001



$$S_I(V) = \int \langle I(t)I(0)\rangle - \langle I\rangle^2 dt = 2eV\frac{2e^2}{h}\sum_n \tau_n(1-\tau_n)$$

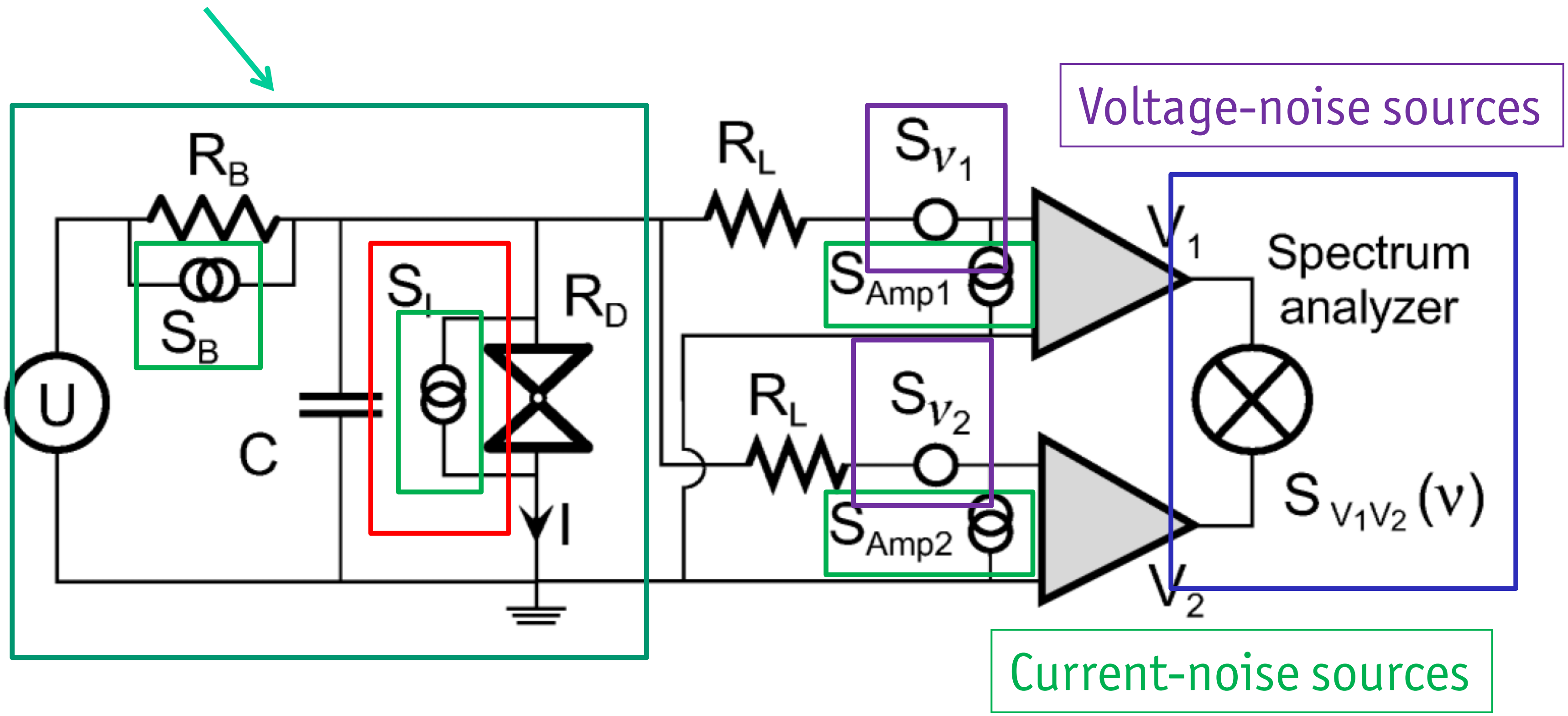


11.5 Correlation set-up for noise measurements

Problem: Signal of interest S_I is smaller than noise of measurement circuit
Solution: Correlation measurement

Cross-correlation eliminates all uncorrelated voltage noise (from leads and amplifiers)

Current-biasing sample R_D via R_B



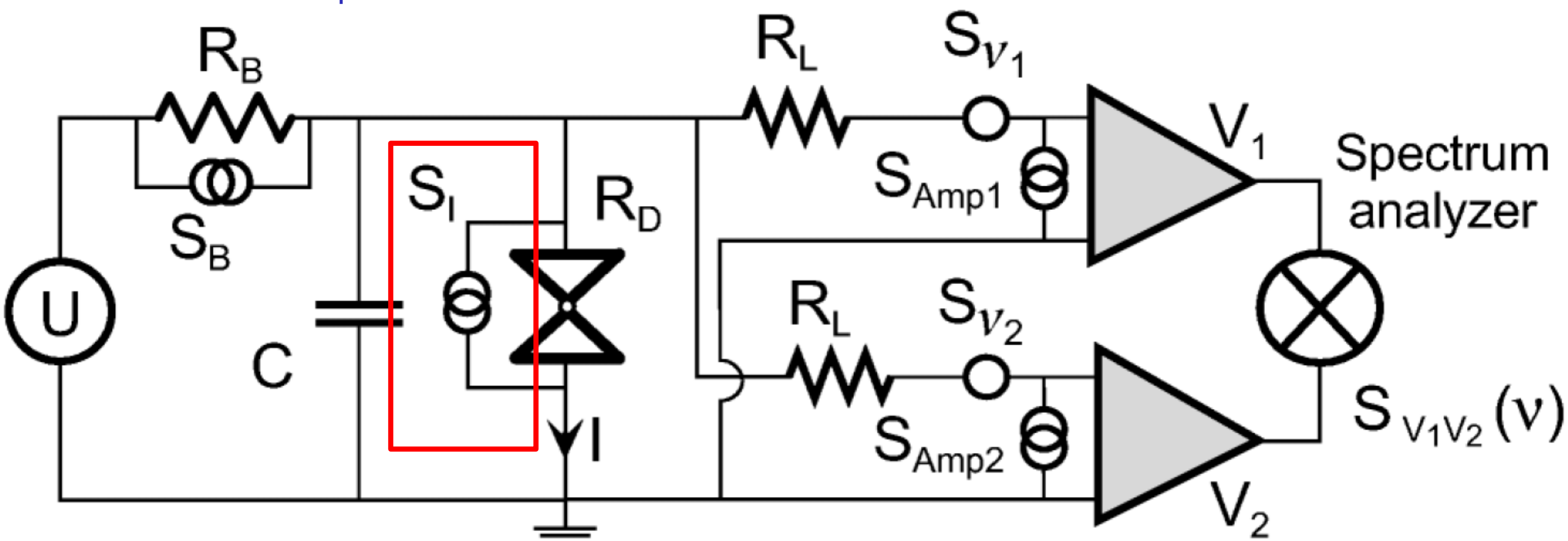
11.5 Correlation set-up for noise measurements

$$S_{V_1V_2}(\nu) = \frac{R_{\parallel}^2}{1 + (2\pi\nu R_{\parallel}C)^2} \times \left[S_I + S_B + 2\left(1 + \frac{R_L}{R_{\parallel}}\right) S_{\mathrm{Amp}} \right]$$

where

$$R_{\parallel} = R_B R_D/(R_B + R_D)$$

S_B and S_{amp} have to be measured independently



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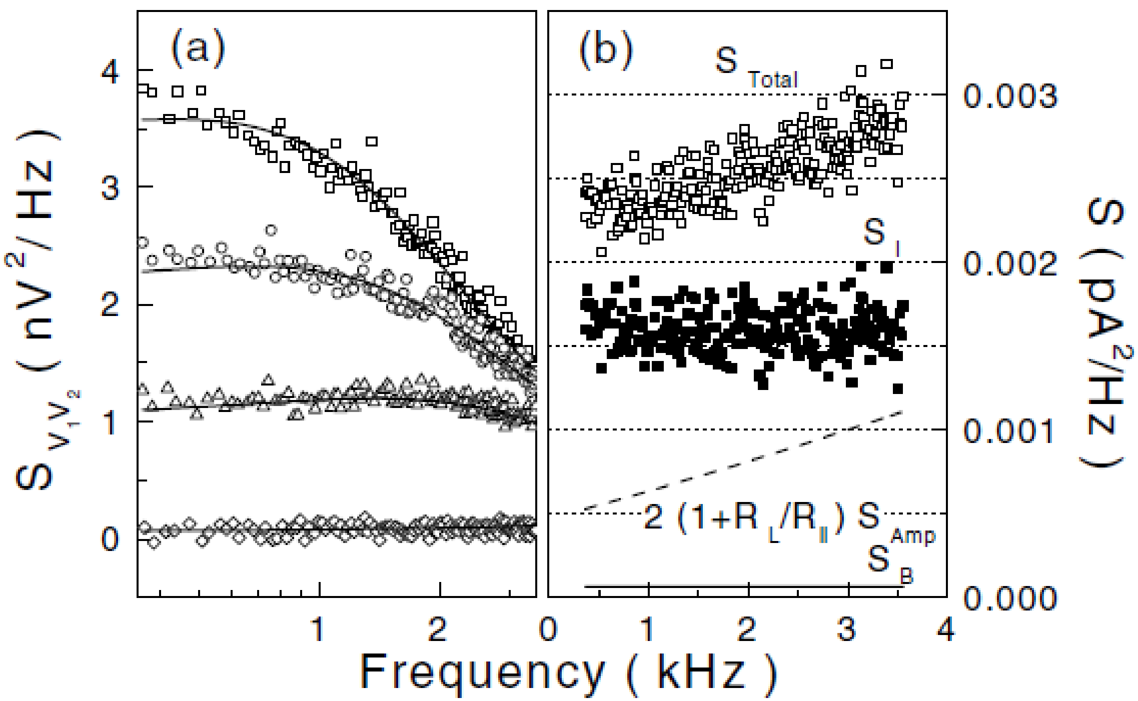
$$S_{V_1V_2}(\nu) = \frac{R_\parallel^2}{1 + (2\pi\nu R_\parallel C)^2} \times \left[S_I + S_B + 2\left(1 + \frac{R_L}{R_\parallel}\right)S_{\mathrm{Amp}}\right]$$

where

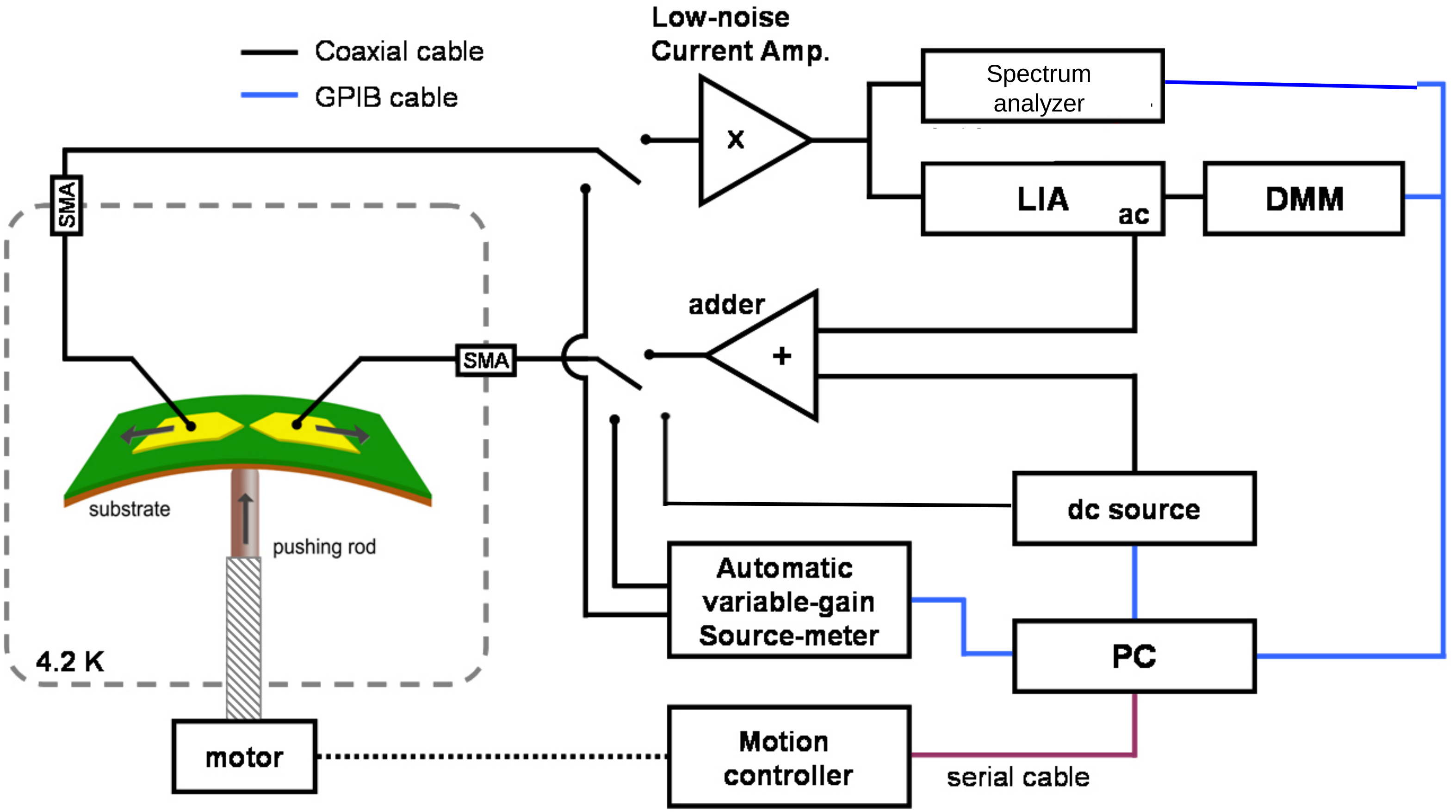
$$R_\parallel = R_B R_D/(R_B + R_D)$$

Examples of equilibrium
Noise spectra V = I = 0



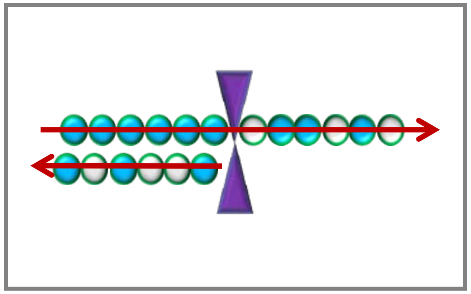
11.6 Lock-in Technique for measuring dI/dV and shot noise with simple wiring

- Measuring the conductance (opening and closing curves)
- First and second differential conductance
- Measurements of the noise in a rather broad range of conductance values from 0.01 G_0 to 1 G_0

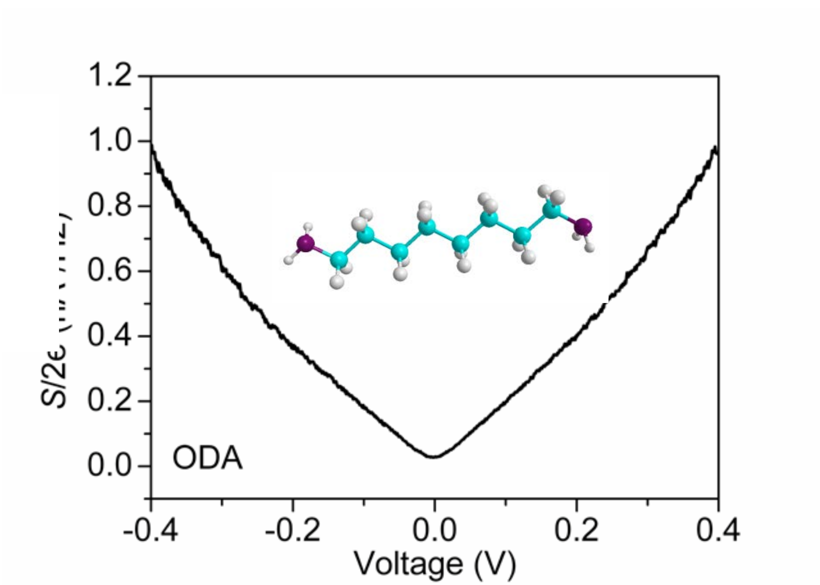


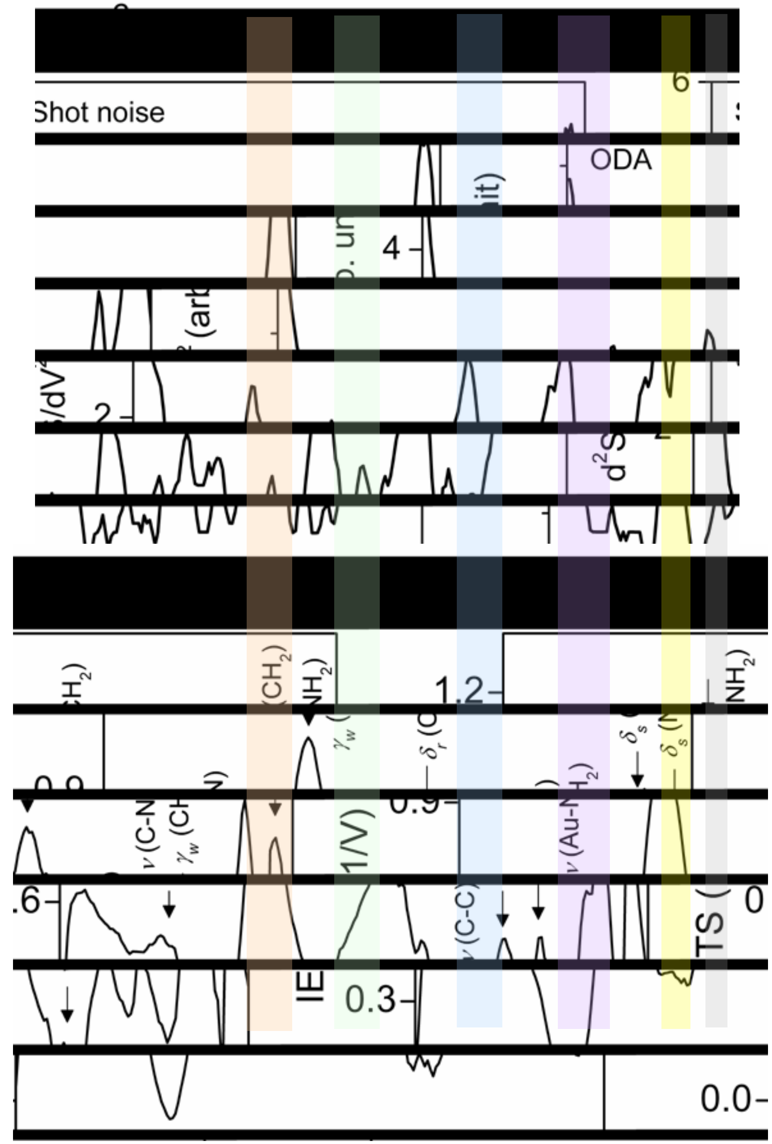
Source: Y. Kim, A. Karimi, D. Weber

Example: Inelastic shot noise



Quantum mechanical scattering & discreteness of charge generate shot noise.





Y. Kim, Univ Konstanz