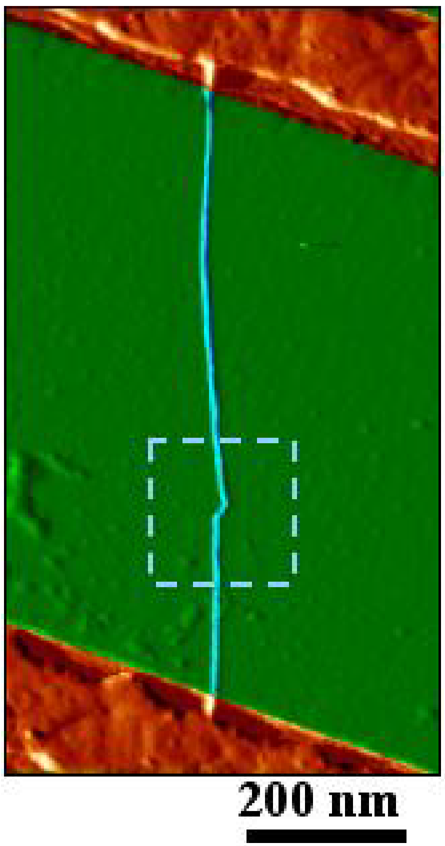
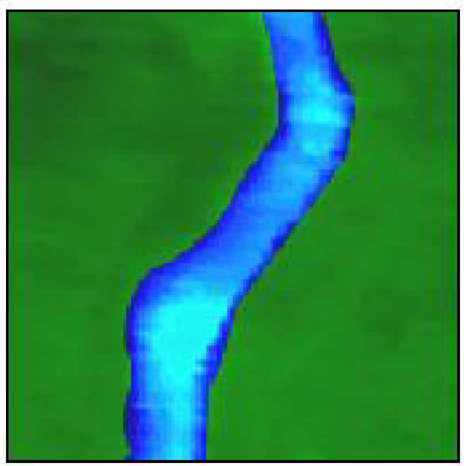
INTRAMOLECULAR NANOTUBE QUANTUM DOTS

MILENA GRIFONI





after Postma *et al.* Science (2001)

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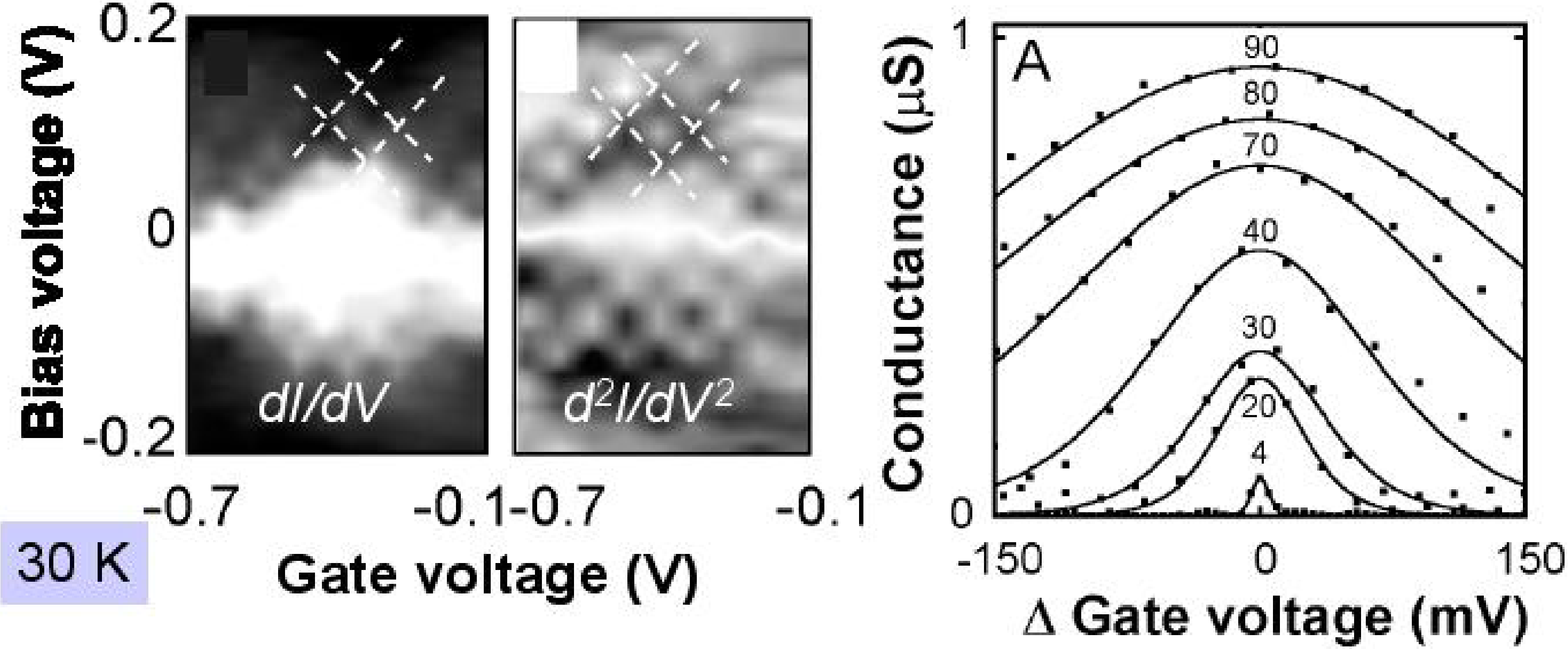
DIMES

NANOTUBE DOT IS A SET

Postma , Teepen , Yao, Grifoni ,Dekker, Science **293** (2001)



E_c = 41 meV, ΔE = 38 meV > $k_B T$ up to 440 K

→ **Coulomb blockade in quantum regime**

CONVENTIONAL SET´s

- quantum dot (Fermi leads)

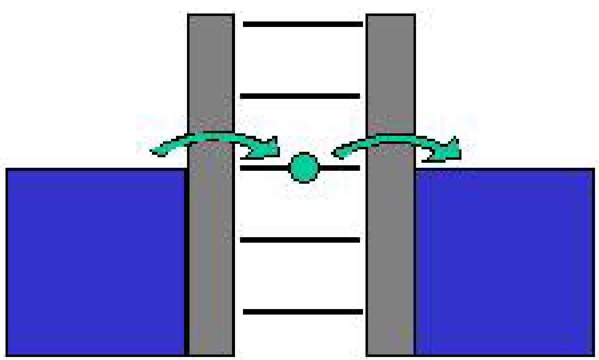
  $G_{\mathrm{MAX}} \propto T^{-1}$ Beenakker PRB (1993)

- Luttinger SET´s

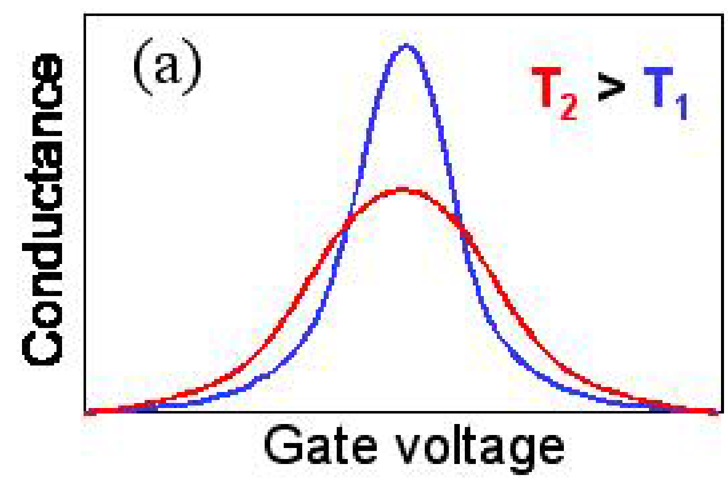
  $G_{\mathrm{MAX}} \propto T^{\alpha_{\mathrm{end}}-1}$ Furusaki , Nagaosa PRB (1993)

  α_{end} ∪0.8 for SWNT ? (a) woud hold

SET´s in quantum Coulomb blockade regime



Sequential tunneling

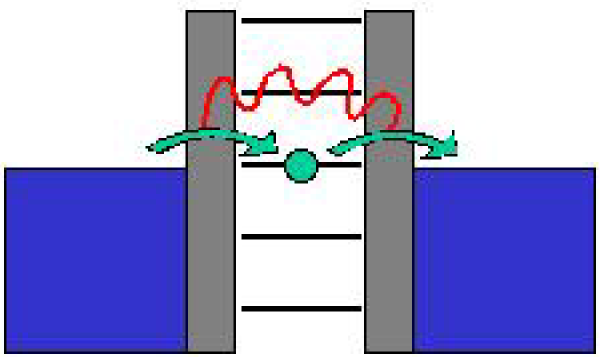


PUZZLE

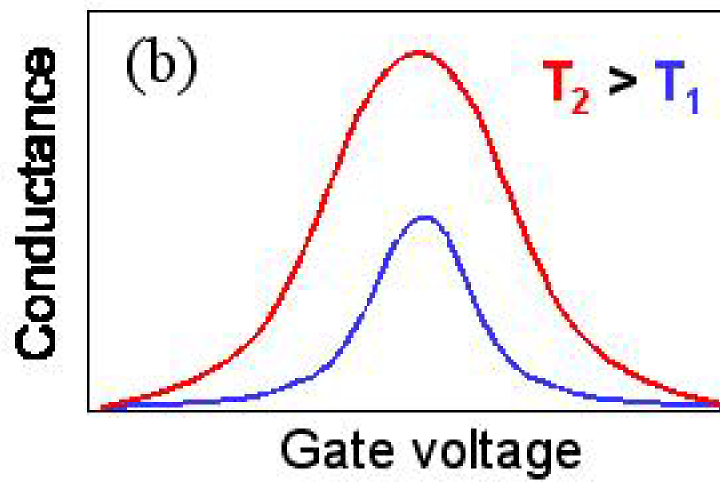
- nanotube SET

Not $G_{\mathrm{MAX}} \propto T^{\alpha_{\mathrm{end}}-1}$ but $G_{\mathrm{MAX}} \propto T^{\alpha_{\mathrm{end\text{-}end}}-1}$ $\alpha_{\mathrm{end\text{-}end}} = 2\alpha_{\mathrm{end}}$

novel tunneling mechanism !!!



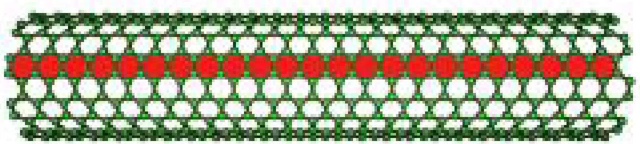
Correlated sequential tunneling

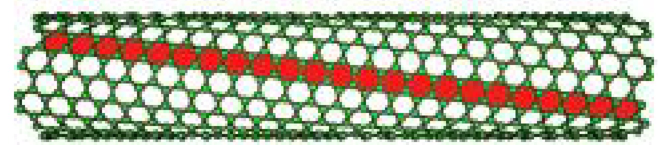


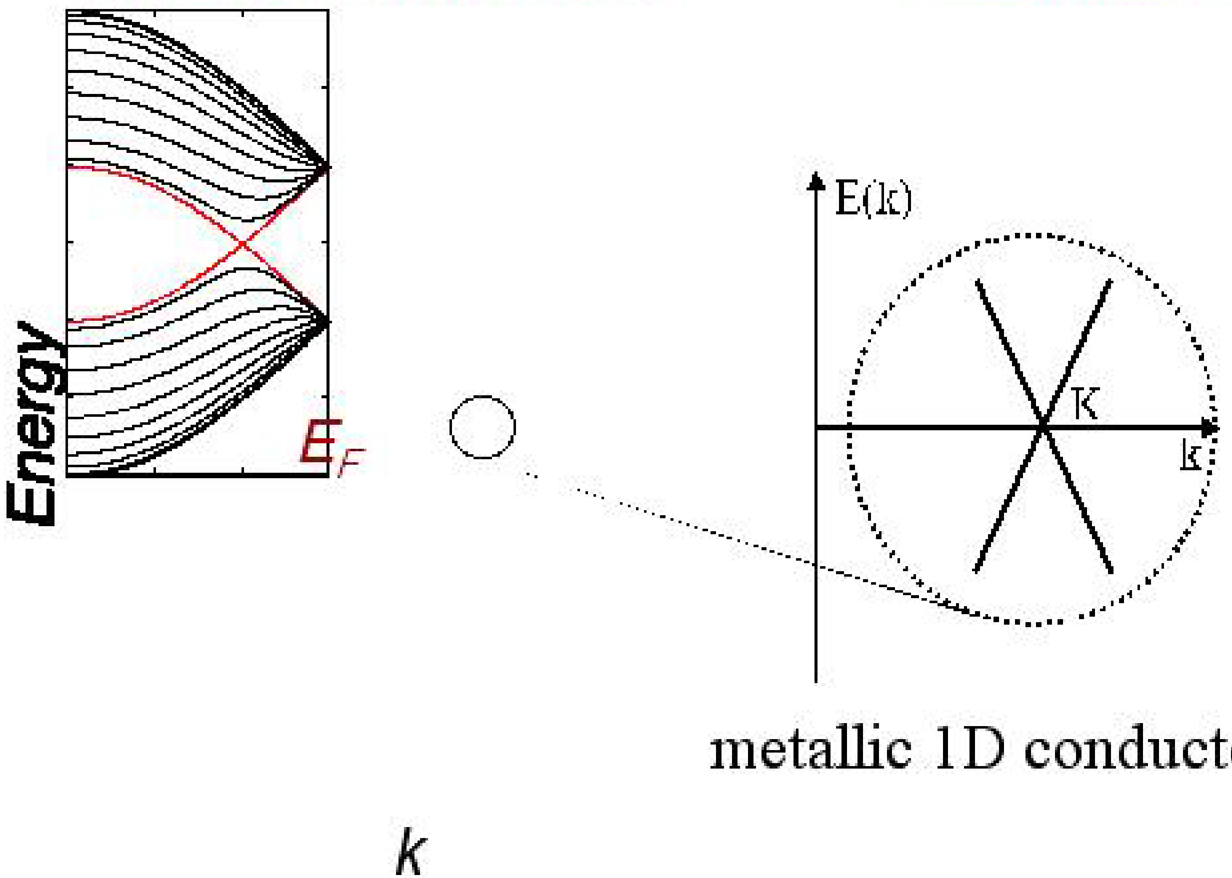
OVERVIEW

- METALLIC SINGLE-WALL NANOTUBES (SWNT)
- SWNT ⟷ LUTTINGER LIQUIDS
- SWNT WITH TWO BUCKLES
  - ↳ 1D DOT WITH LUTTINGER LEADS
- UNCOVENTIONAL RESONANT TUNNELING EXPONENT
  - ↳ NOVEL TUNNELING MECHANISM
- CONNECTION WITH ORTHODOX COULOMB BOCKADE

METALLIC SWNT MOLECULES

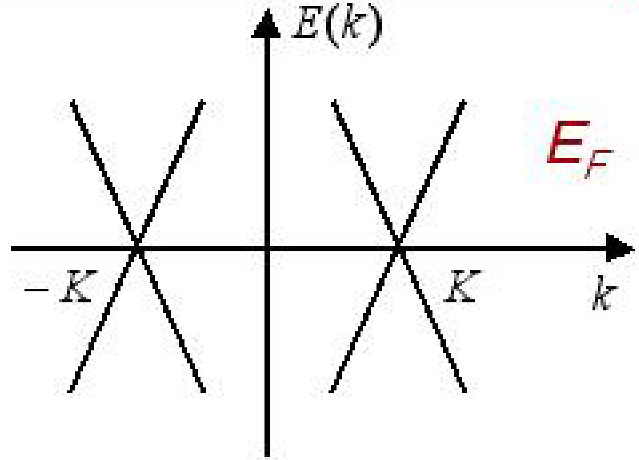






metallic 1D conductor with 2 linear bands

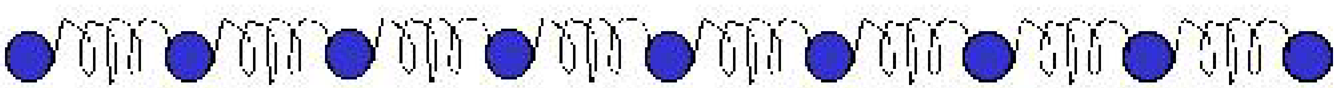
METALLIC SWNT AS LUTTINGER LIQUIDS



FERMI SURFACE HAS ONLY FOUR POINTS

failure of Fermi-Landau picture

Coulomb + Pauli interaction

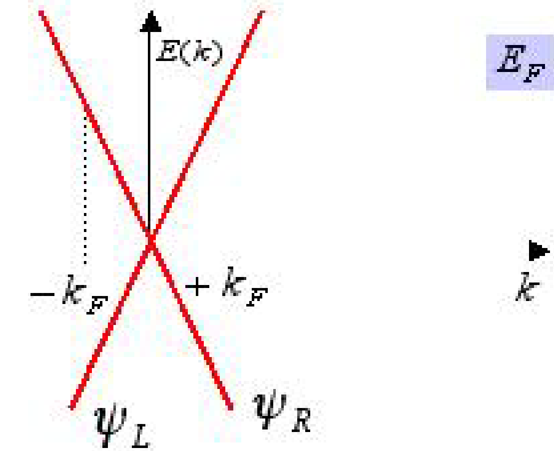


ELECTRONS FORM A HARMONIC CHAIN AT LOW ENERGIES (LUTTINGER LIQUID)

COLLECTIVE EXCITATIONS ARE VIBRATIONAL MODES

WHAT IS A LUTTINGER LIQUID ?

example: spinless electrons in 1D

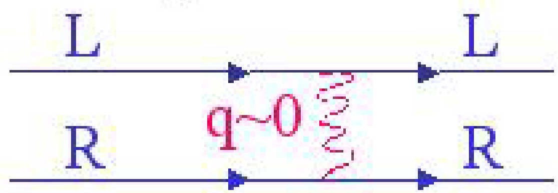


- linear spectrum

bosonization identity $\psi_{R/L} \sim e^{i(\varphi \pm \theta)}$

charge density $\rho(x) \cup \frac{k_F}{\pi} + \frac{f_x \theta}{\sqrt{\pi}}$

- + forward scattering

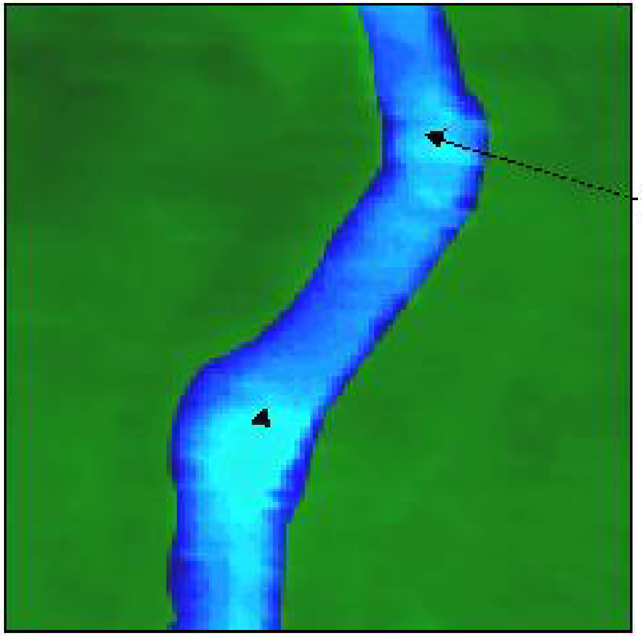


g Luttinger parameter

$$H_0 = \frac{v_\rho}{2} \square dx\,[g\Pi^2(x) + \frac{1}{g} f_x \theta(x)^2],$$

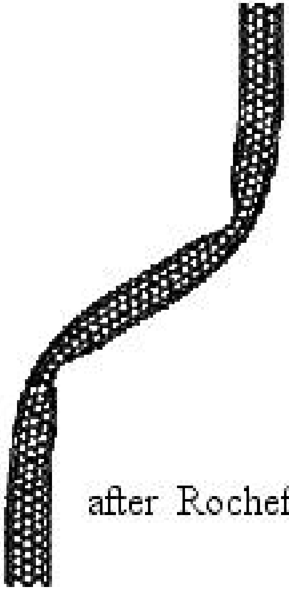
$v_\rho = \frac{v_F}{g}$ plasmon velocity

DOUBLE-BUCKLED SWNT´s



50 x 50 nm²

buckles act as tunneling barriers

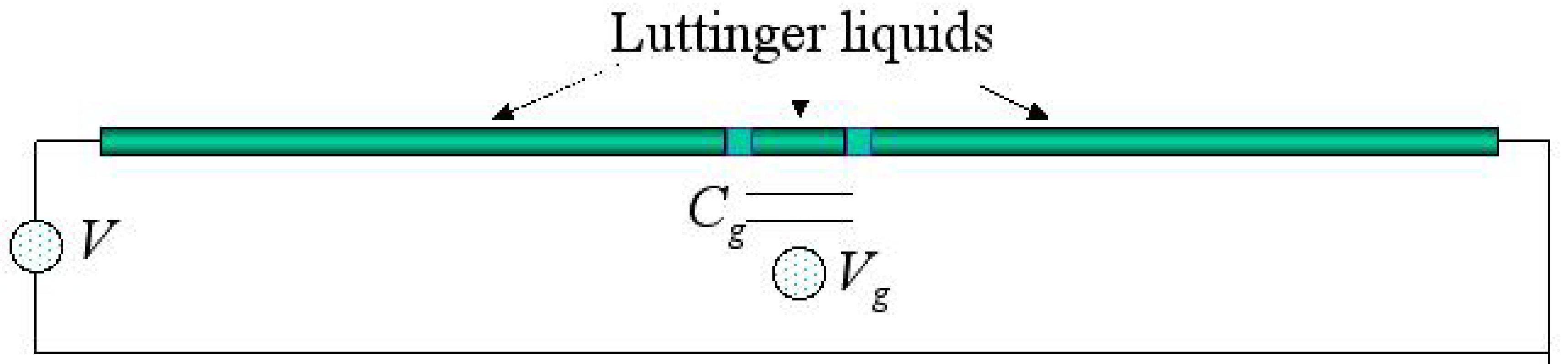


after Rochefort *et al.* 1998

➡ Luttinger liquid with two impurities

Let us focus on spinless LL case, generalization to SWNT case later

TRANSPORT



$$H_{tot} = H_0 + H_{imp} + H_{ext}$$

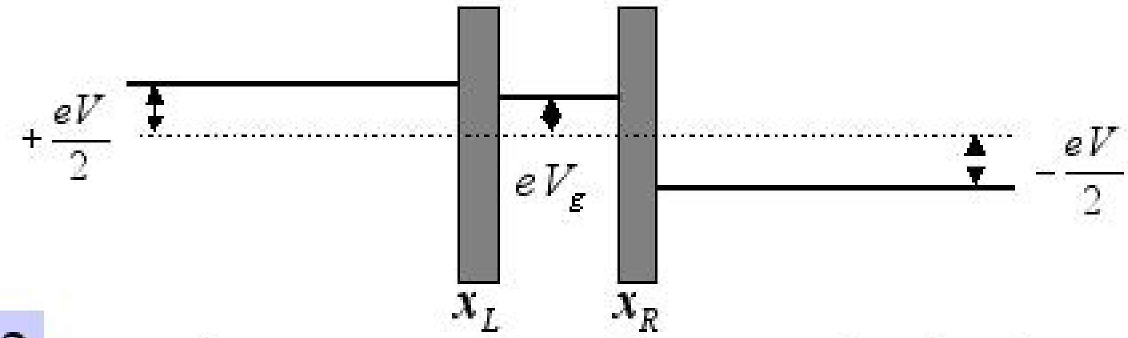
localized impurities

voltage sources

- $H_{imp} = \int dx\, \rho(x) U_{imp}(x), \quad \hat{U}_{imp}(k) \propto const$ ⟹ backscattering
- $H_{ext} = -e \int dx\, \rho(x) V_{ext}(x), \quad V_{ext}(k) \propto const$ ⟹ forward scattering

continuity equation ⟹ $I(x_R, t) = -e \left\langle \dot{\theta}(x_R, t) \right\rangle$

TRANSPORT II



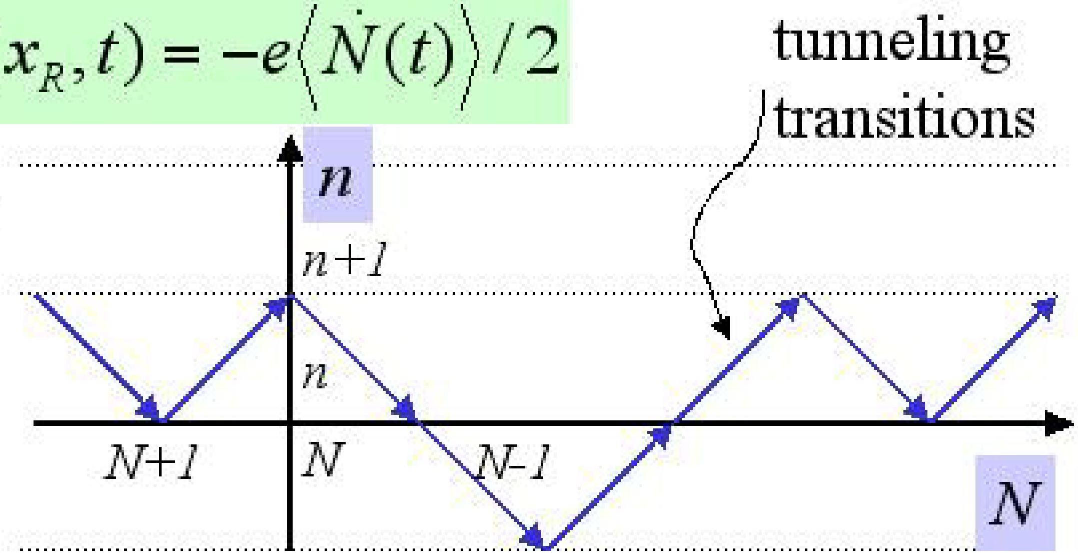
$$N(t)/2 \cup \left[\theta(x_R)+\theta(x_L)\right]/2$$ charge transferred across the leads

$$n(t) \cup \left[\theta(x_R)-\theta(x_L)\right]$$ electrons on the island

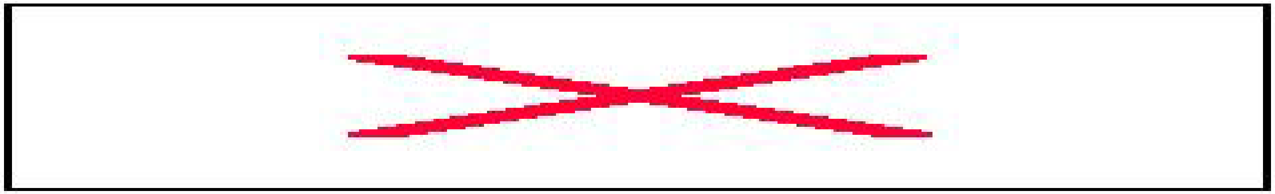
$$\Rightarrow \quad I(x_R,t) = -e\left\langle \dot{N}(t)\right\rangle/2$$

`particles´ n, N in washboard potential

- $H_{imp} = U_{imp}\cos(\pi N)\cos(\pi n)$
- $H_{ext} = -\frac{eV}{2} - \frac{eV_g C_g}{C_{tot}} C$



CURRENT



- Exact trace over bosonic modes $\theta(x)$, x ? x_R, x_L $\qquad \vec{N} = (N, n)$

$$P(N_f, n_f, t; N_i, n_i, 0) = \int \mathcal{D}\vec{N} \int \mathcal{D}\vec{N}' A[\vec{N}] A^*[\vec{N}'] F_N[N, N'] F_n[n, n']$$

bare action

$H_{imp} + H_{ext}$

bulk modes

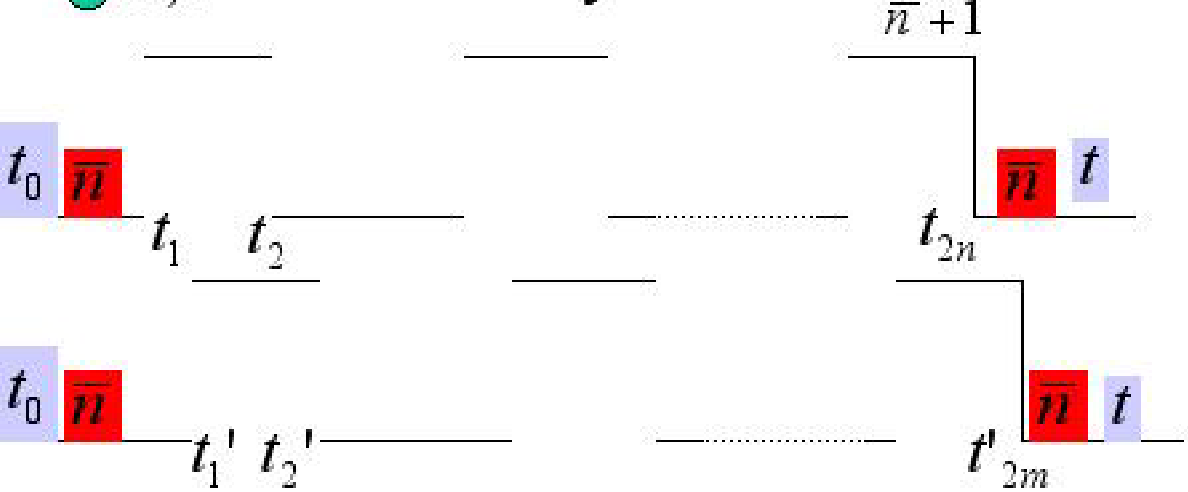
- nonlocal in time coupling

$\vec{N}(t), \vec{N}'(t')$

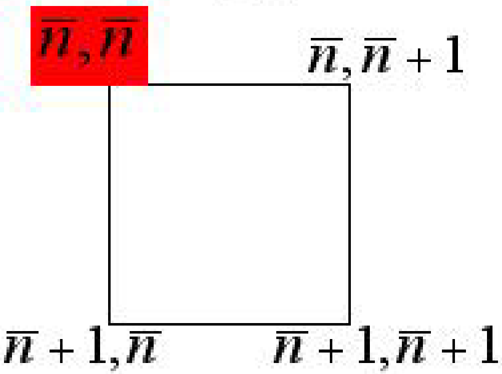
- F_n ♦ mass gap for n

LINEAR TRANSPORT

- n, n' two state dynamics

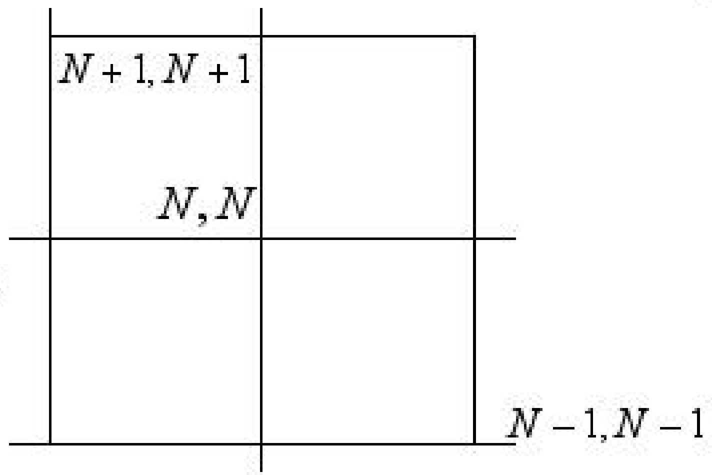


single path
over four states
in (n,n') plane



- N, N' ∞ – states dynamics

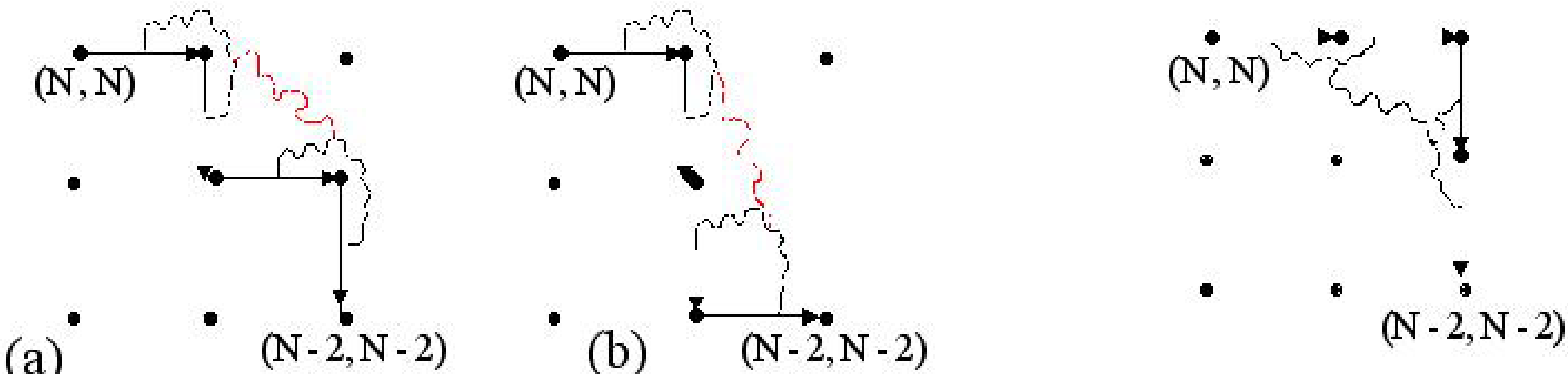
single path
over infinite states
in (N,N') plane



CORRELATIONS

- Minimal paths for transfer of charge through the dot

N ♦ N ± 2

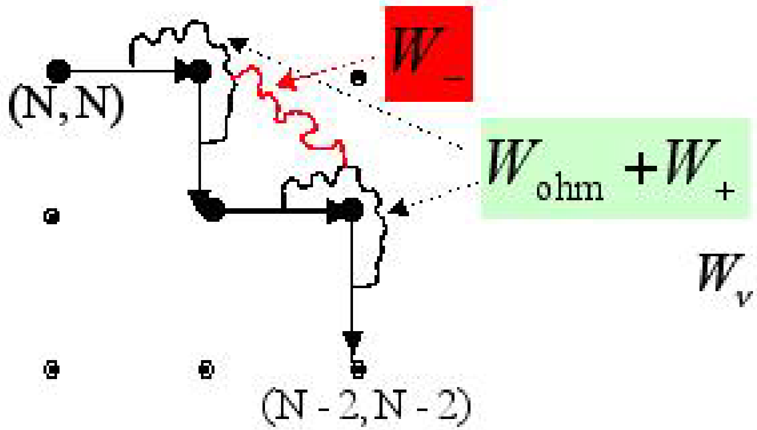


real dot occupation
(sequential tunneling)

virtual occupation
(cotunneling)

ONLY if can be neglected → conventional uncorrelated ST

CORRELATIONS II



$\nu = \text{ohm}, \pm$

$$W_\nu(t) = \int_0^\infty d\omega \, \frac{J_\nu(\omega)}{\omega^2} \left[(1 - \cos\omega t) \coth\frac{\hbar\omega}{k_B T} + i \sin\omega t \right]$$

$$J_{\text{ohm}}(\omega) = \frac{\omega}{g} e^{-\omega/\omega_c}$$

plasmon continuous in the leads

plasmon modes quantized in the dot

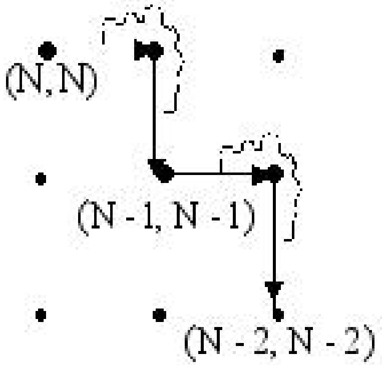
$$\Delta E = \frac{\pi \hbar v_F}{x_0 g}$$

$$J_\pm(\omega) = \omega \, e^{-\omega/\omega_c} \frac{\Delta E}{\hbar g} \sum_{m=1}^{\infty} \left[\delta(\omega - \omega_{2m-1}) \pm \delta(\omega - \omega_{2m}) \right]$$

$$W_\pm(t + \tau_{\Delta E}) = W_\pm(t)$$

RATES

a) High temperaures $k_B T > \Delta E$ $W_+ \blacklozenge W_{ohm}, \quad W_- \blacklozenge 0$



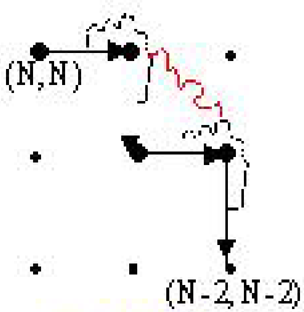
uncorrelated ST

$$\Gamma_{UST} = \frac{\Delta_R^2 \Delta_L^2}{\Delta_R^2 + \Delta_L^2} f_{UST}(T, g)$$

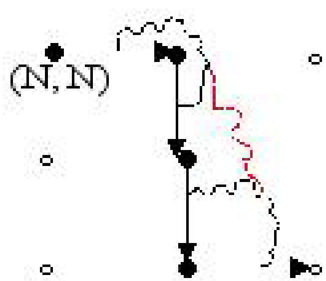
2nd order

b) Low temperatures $k_B T < \Delta E$ $W_- ? 0$

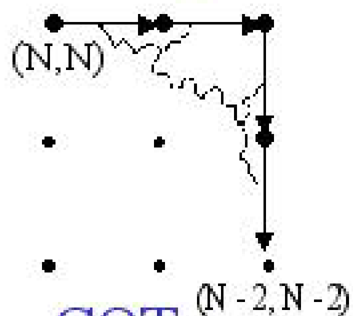
correlated ST + cotunneling



CSTa



CSTb



COT

$$\Gamma = \Gamma_{CSTa} + \Gamma_{CSTb} + \Gamma_{COT}$$

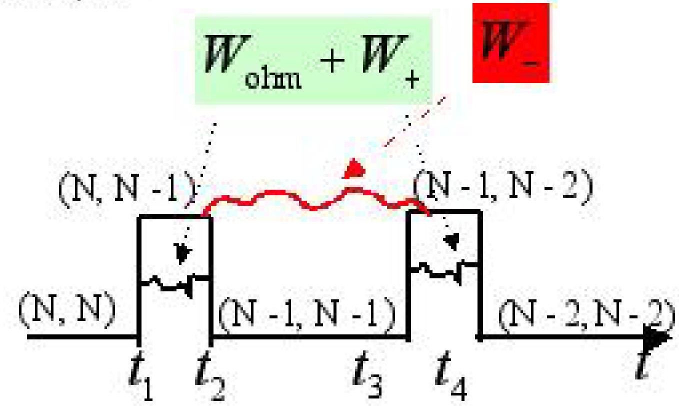
$$\Gamma = \Delta_R^2 \Delta_L^2 f_{COR}(T, g)$$

4th order !!!

CORRELATED RATES

$k_B T < \Delta E$

$$\Gamma_\nu^f = \Delta_R^2 \Delta_L \,\mathrm{Re} \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3\, e^{-i\varepsilon_\nu(\tau_1,\tau_2,\tau_3)} e^{\Phi_\nu(\tau_1,\tau_2,\tau_3)}$$



- Expansion in Fourier series
  → exact analytical evaluation

- At resonance $\varepsilon(n,V_G) \approx 0$

$$\Gamma_\nu^f \propto \Delta_R^2 \Delta_L^2 f_g f_g = \Delta_R^2 \Delta_L^2 T^{\alpha_{end}} T^{\alpha_{end}}$$

$$\Phi_{CSTa} = -W^*(\tau_1) - W(\tau_3) + W_\Delta(\tau_2) - W_\Delta^*(\tau_1+\tau_2) - W_\Delta(\tau_2+\tau_3) + W_\Delta(\tau_1+\tau_2+\tau_3)$$

LUTTINGER MODES IN SWNT

SWNT : 2 ↔ band + 2 ↔ spin

→ 4 bosonic fields

Kane, Balents , Fisher PRL (1997)
Egger, Gogolin , PRL (1997)

$$\theta_{\rho}(x) = \theta_{1-}(x) + \theta_{1/}(x) + \theta_{2-}(x) + \theta_{2/}(x)$$

$$\theta_{\sigma}(x) = \theta_{1-}(x) - \theta_{1/}(x) + \theta_{2-}(x) - \theta_{2/}(x)$$

$$\theta_{\Delta\rho}(x) = \theta_{1-}(x) + \theta_{1/}(x) - (\theta_{2-}(x) + \theta_{2/}(x))$$

$$\theta_{\Delta\sigma}(x) = \theta_{1-}(x) - \theta_{1/}(x) + (\theta_{2-}(x) - \theta_{2/}(x))$$

$$g_{\rho} = \frac{1}{\sqrt{1 + \frac{8e^2}{\pi\hbar v_F} \ln \frac{R_S}{R}}} \cup 0.2$$

$$g_{\sigma} = g_{\Delta\rho} = g_{\Delta\sigma} = 1$$

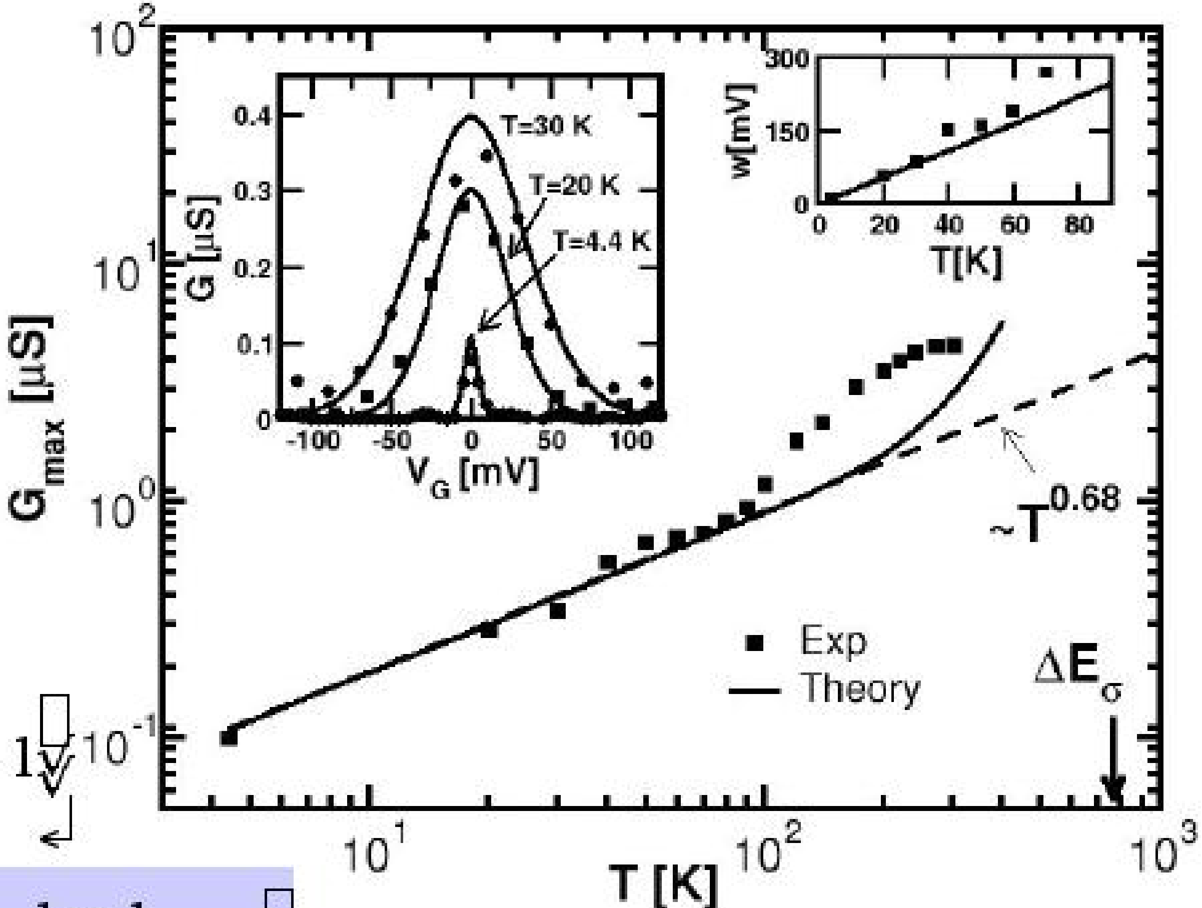
$$H_0 = \sum_{a=\rho,\sigma,\Delta\rho,\Delta\sigma} \frac{v_{\rho}}{2} \int dx \, [g_a \Pi_a{}^2(x) + \frac{1}{g} f_x \theta_a(x)^2],$$

RESULTS

At resonance

$$\Gamma_{CSTa} = \Gamma_{CSTb} = \Gamma_{COT}$$

$$? \quad G \propto \Gamma / T \cup T^{\alpha_{\text{end-end}}-1}$$



spinless LL $\alpha_{\text{end-end}} = 2\left(\frac{1}{g} - 1\right)$

nanotubes g ♦ $g_{\text{eff}}, \quad g_{\text{eff}}^{-1} = \frac{1}{4}\left(\frac{1}{g_\rho} + 3\right)$

CONCLUSIONS & REMARKS

ò LOW TEMPERATURES $L_T = \hbar v_F / k_B T > x_0$

BREAKDOWN OF UST IN LINEAR REGIME

BUT UST O.K. IN NONLINEAR REGIME $eV > k_B T$

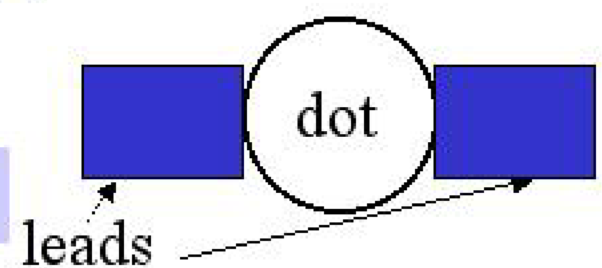
ò NONINTERACTING ELECTRONS $g = 1 \longrightarrow \alpha_{\text{end-end}} = \alpha_{\text{end}} = 0$

$G_M(g = 1) \propto T^{-1}$

WHAT IS THE TUNNELING MECHANISM IN CONVENTIONAL DOTS ?

ò CONVENTIONAL DOTS:

THREE SEPARATE PIECES UST ok!?!



Part 2: NANOTRIBOLOGY by ATOMIC FORCE MICROSCOPY on CARBON NANOTUBES

S. Decossaset al., Europhys. Lett. 53 (6), 742 (2001)

- Interaction between an AFM tip and ananotube carpet :

  Experiments and numerical simulations

Nanotube carpets grown by HFCVD (A.M. Bonnot)

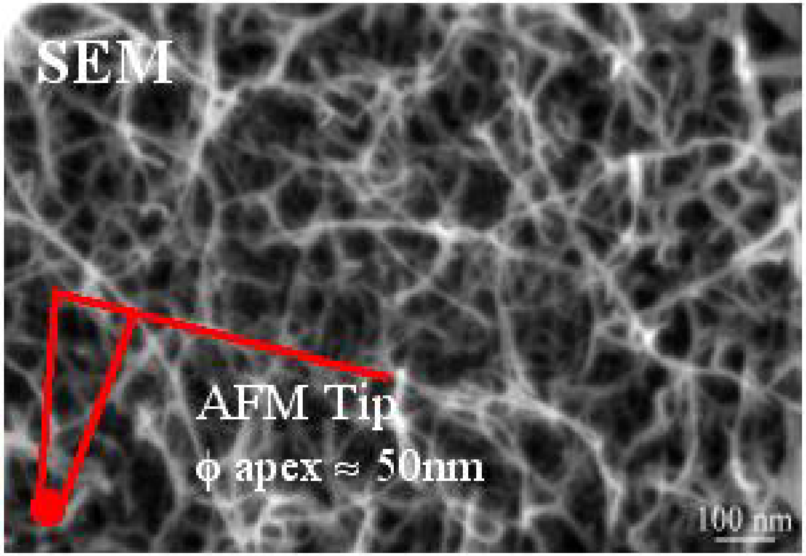
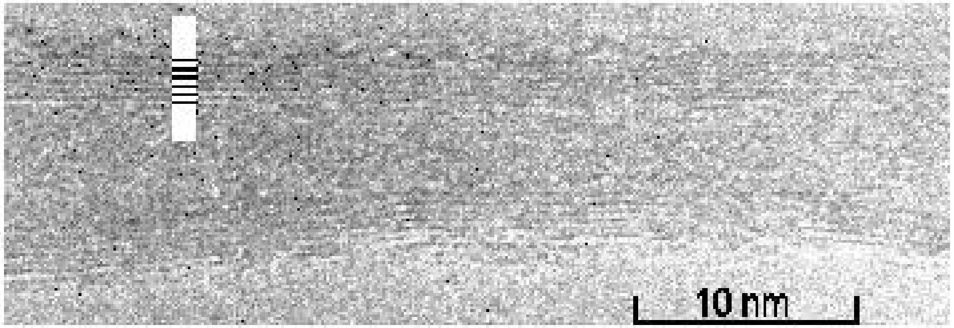
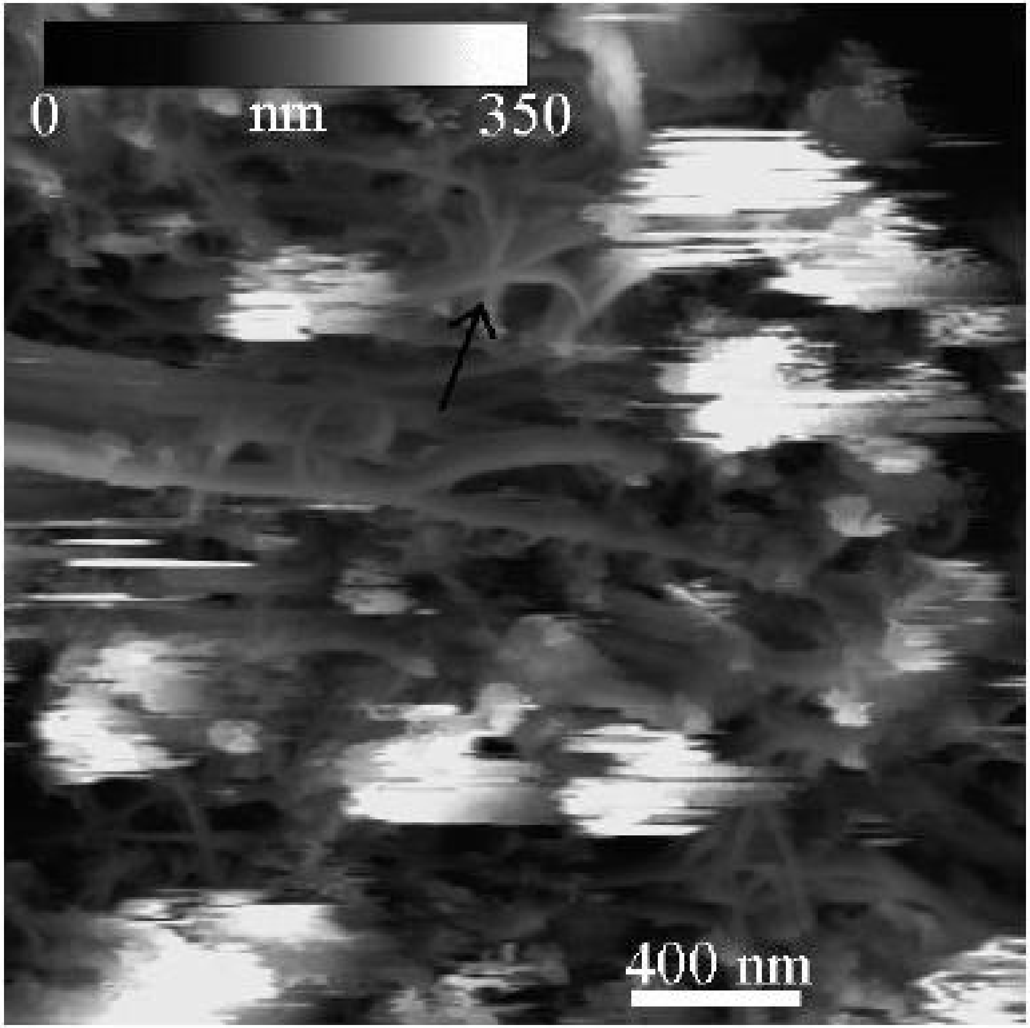
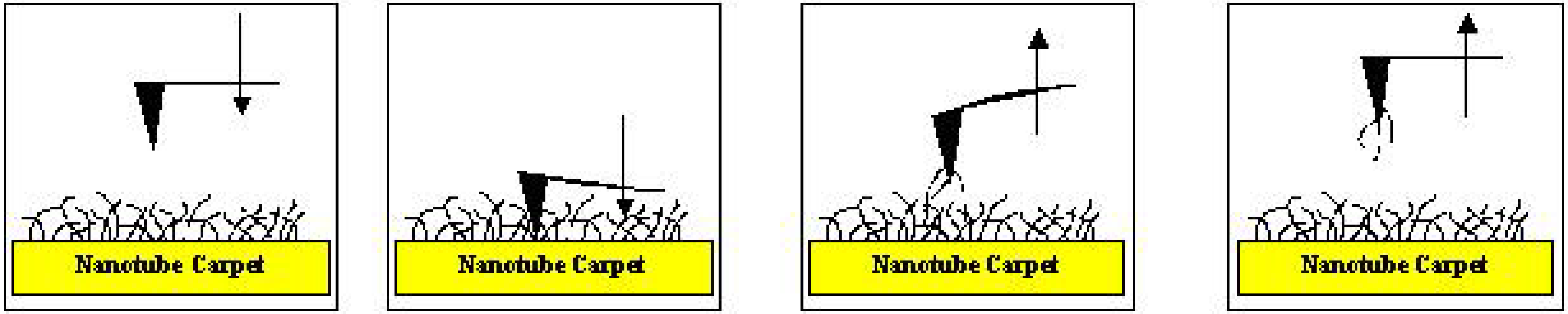


Image L. PONTONNIER, Cristallographie, CNRS Grenoble

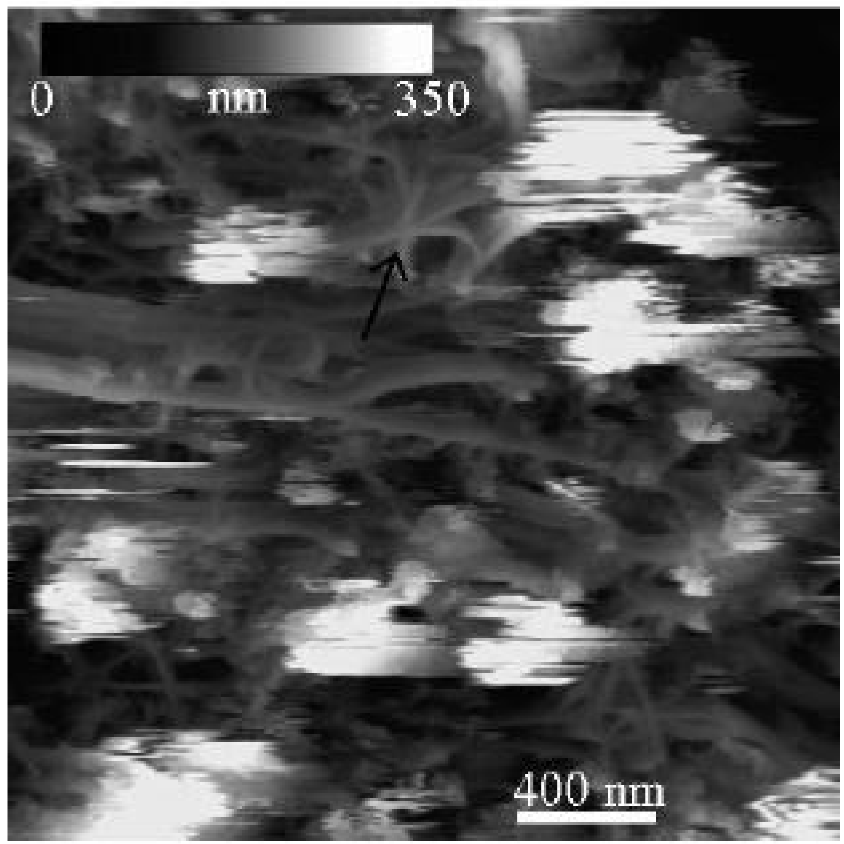


- Entangled nanotubes (length : 100nm to few μm)
- Multi-walled carbon nanotubes (φ ≈25nm) with well defined atomic structure
- Discrete medium for an AFM tip

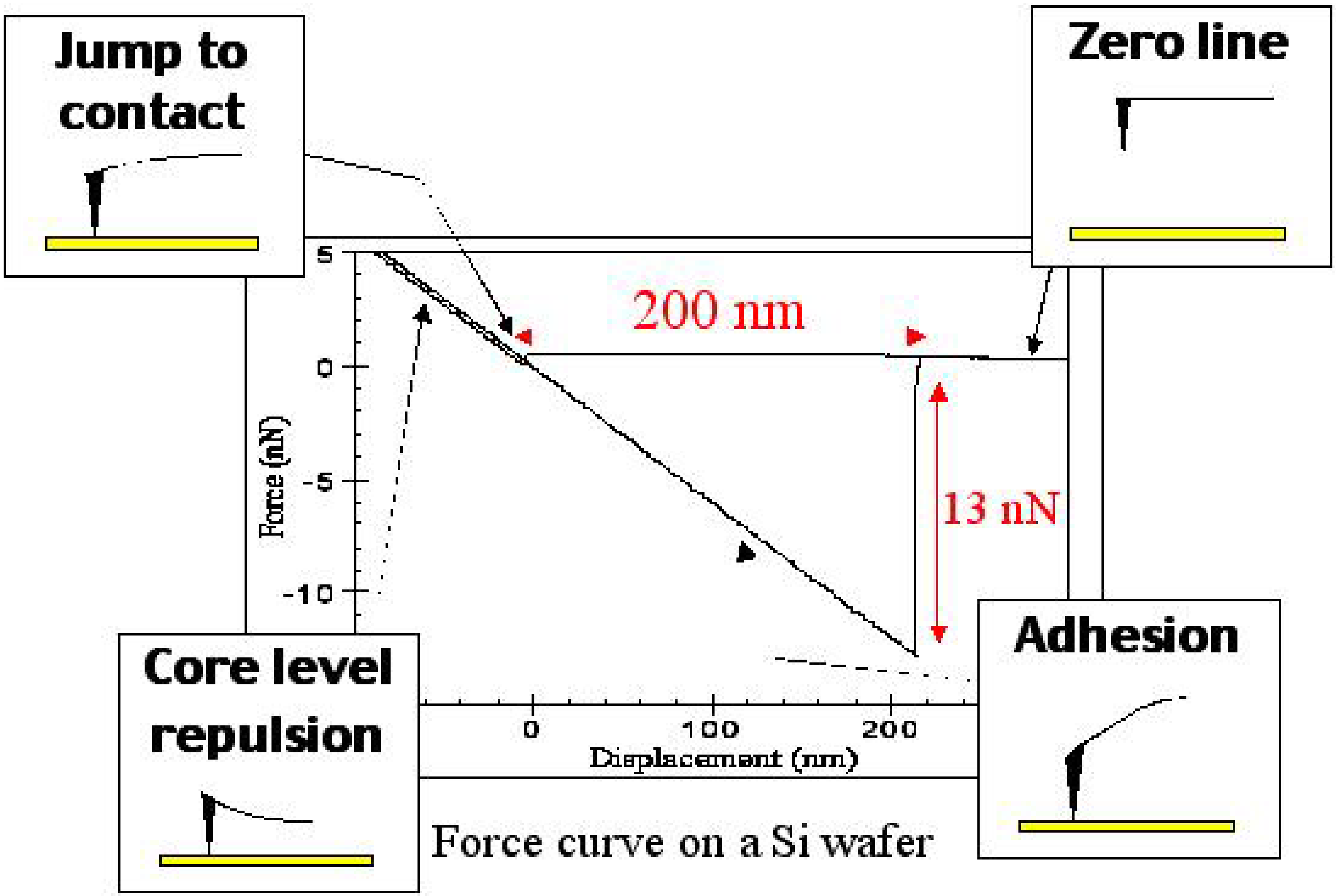

0
nm
350
400 nm



AFM: Force curve measurements

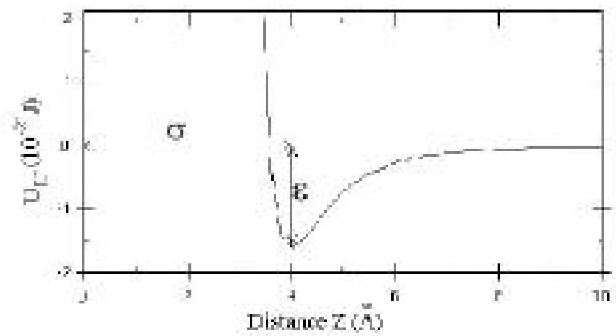


Force curve measurement by AFM



Force curve on a Si wafer

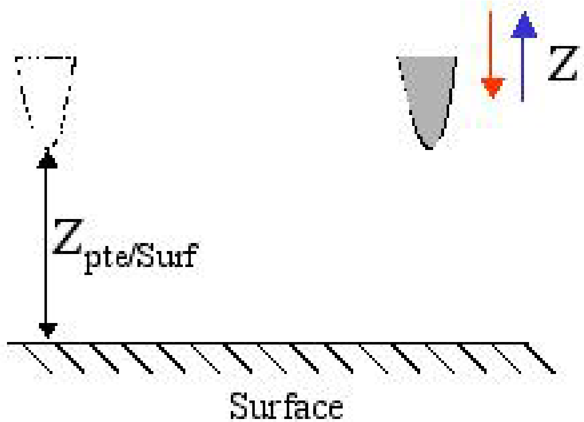
(air, tip spring constant = 0.06 $N.m^{-1}$)

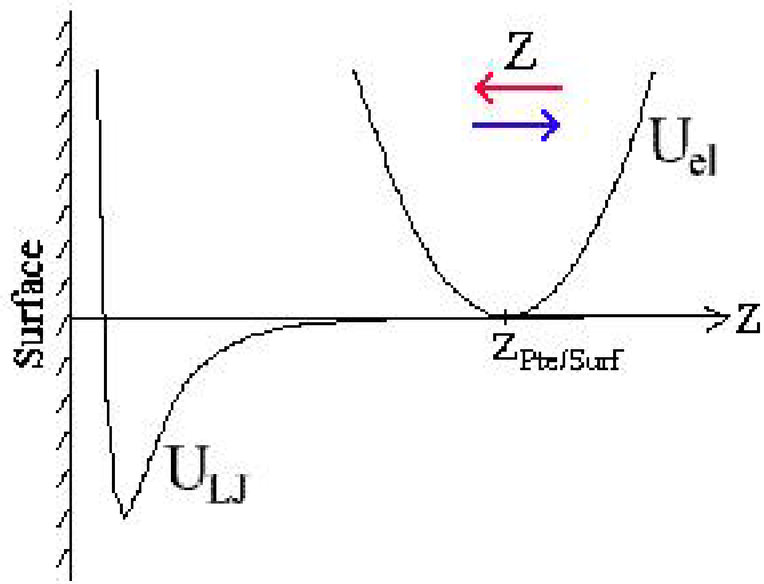


$$U_{LJ} = \frac{\epsilon}{(n-m)}\left(\frac{m\sigma^n}{Z^n} - \frac{n\sigma^m}{Z^m}\right)$$

Fig. 1.7 – Potentiel de Lennard Jones ($\sigma = 4$ Å, $\epsilon = 10$ meV, n = 12, m = 6).

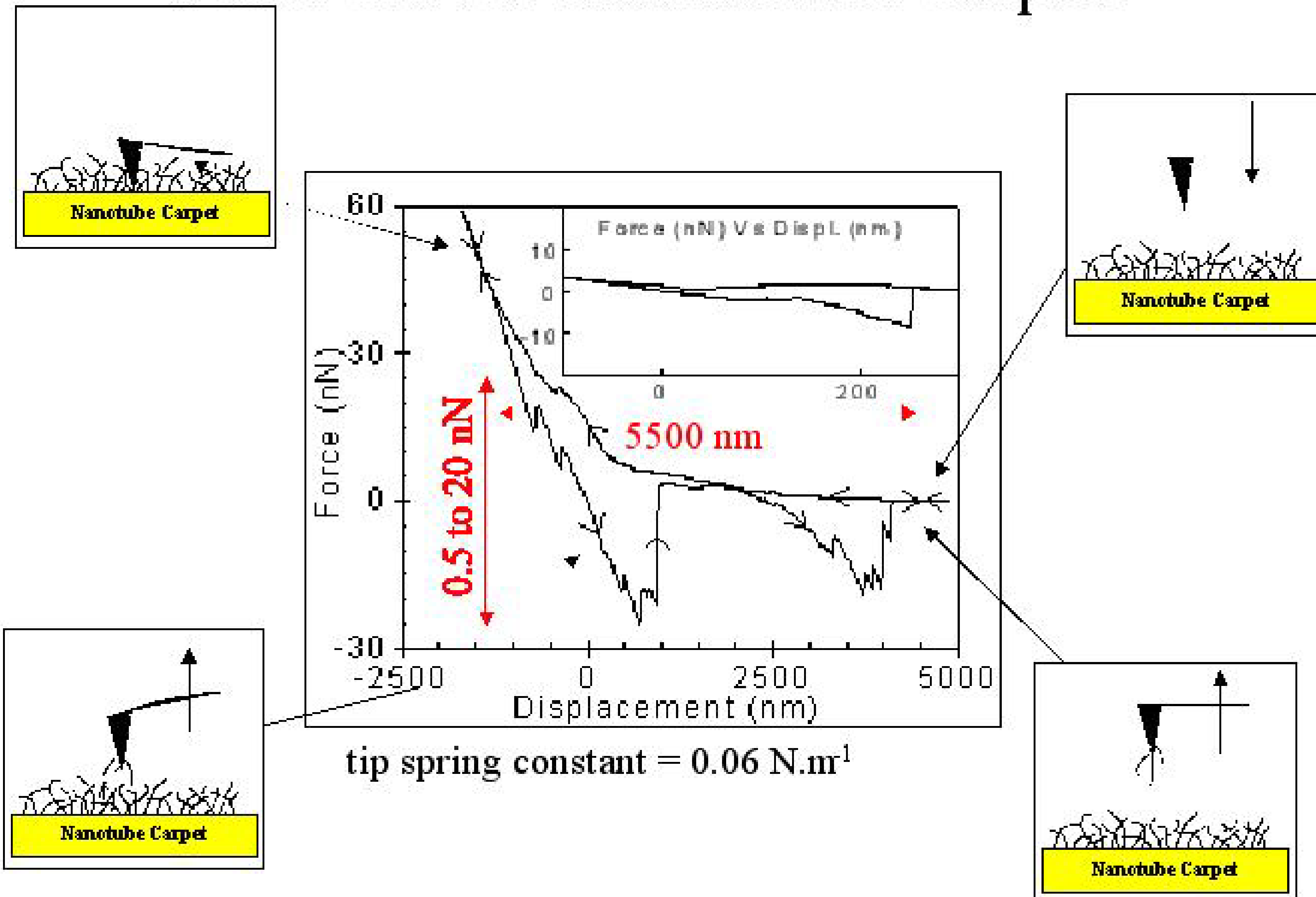
AFM tip stable position





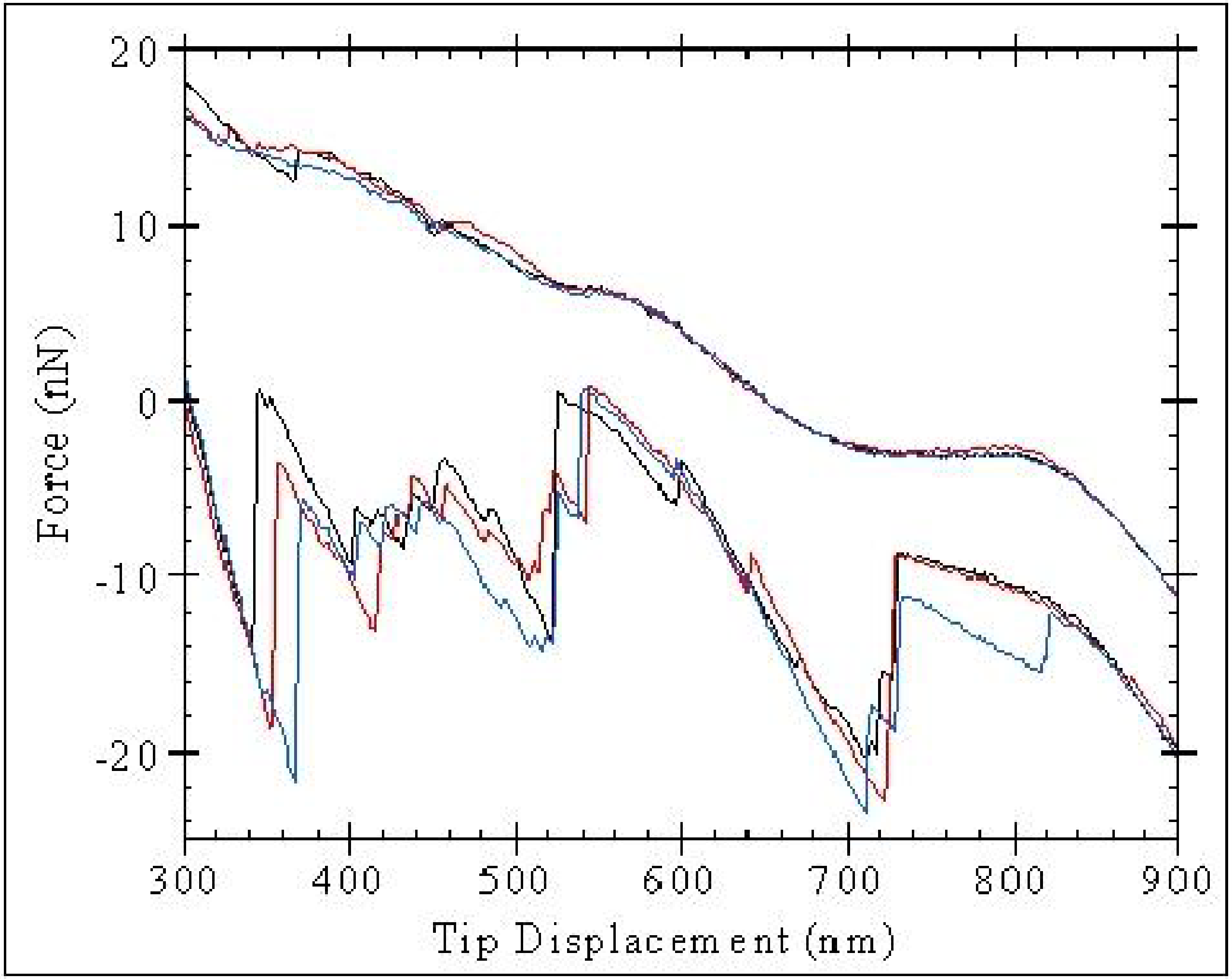
(1-4)

Force curves on nanotube carpets



tip spring constant = 0.06 N.m^{1}

Reproducibility of force curves on nanotube carpets



Tip spring constant = 0.58 N.m[1]

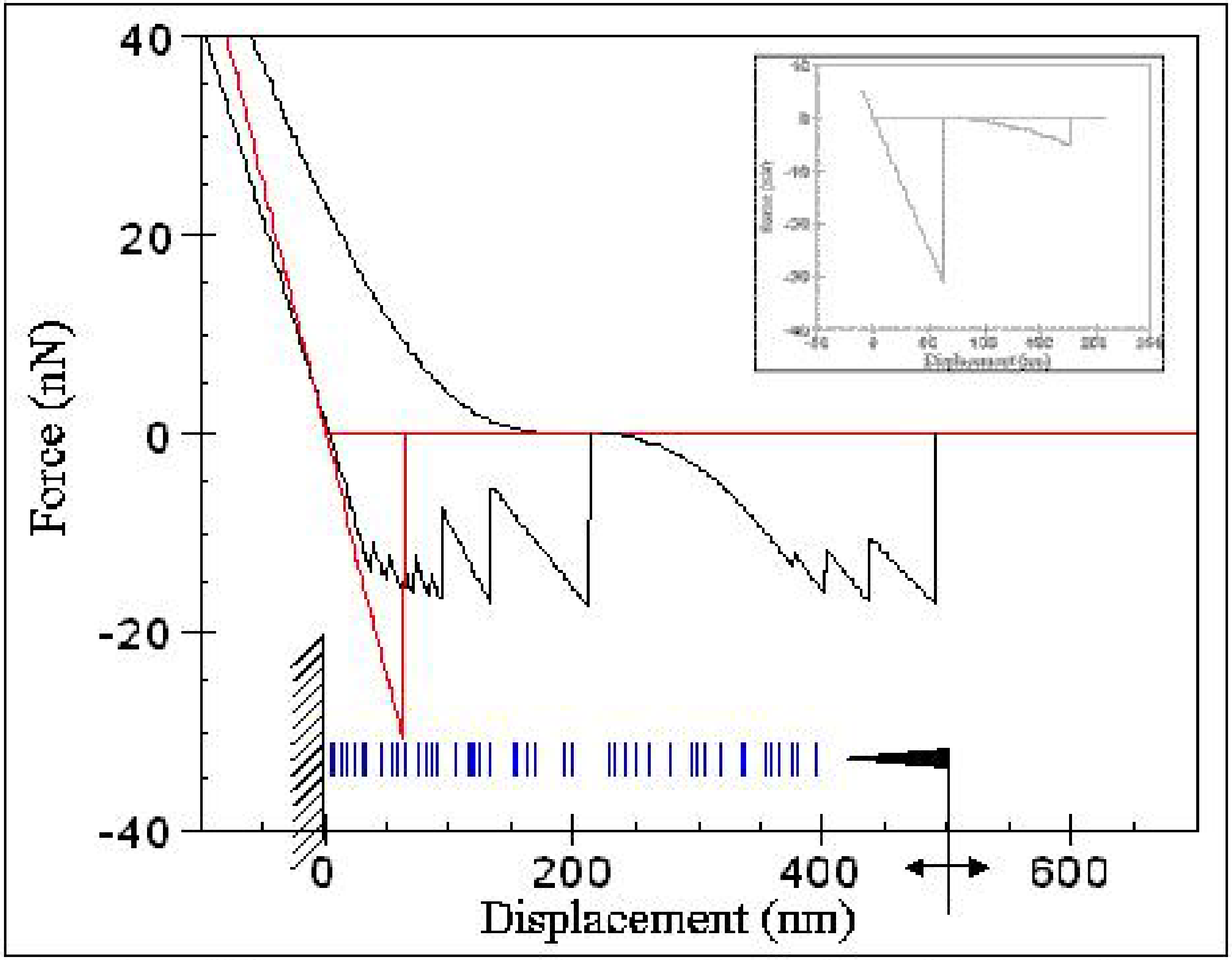
Force curves on nanotube carpet: numerical simulation

Analysis of the mechanical stability: CNT carpet indented by the tip

- Point like tip of spring constant k_{tip}
- Tip wafer interaction : 12-3 Lennard Jones potential
- Tip nanotube interaction = Long range attractive (Vander Waals like)
- Schematic description of the nanotube carpet
  - Nanotube wafer distance randomly chosen
  - Nanotube length and diameter randomly chosen
  - Nanotube can be elongated (Young modulus E)
  - Nanotube can be bent (spring constant k_{CNT})
- No nanotube-nanotube nor nanotube-wafer interaction

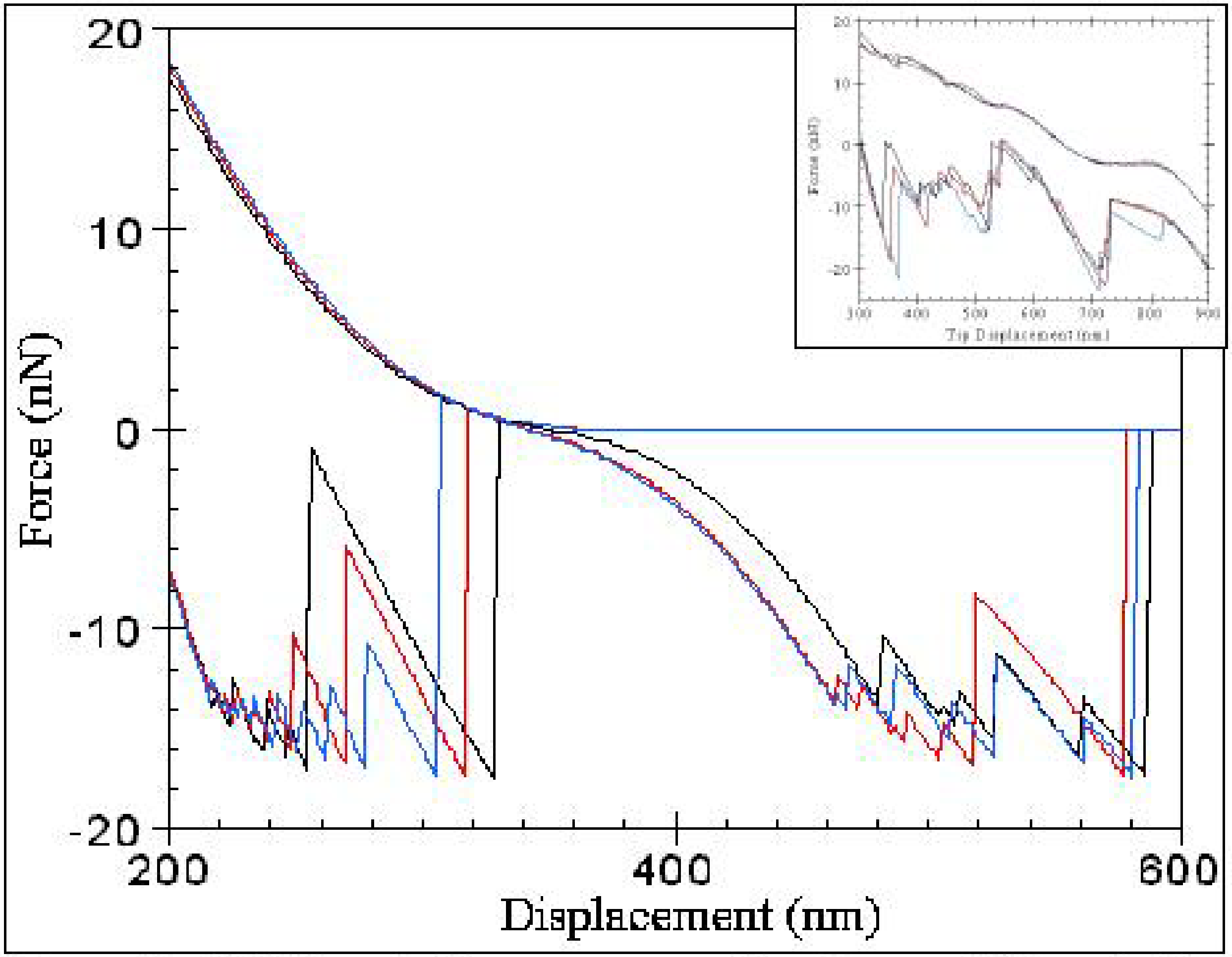
S. Decossas et al., Europhys. Lett. **53** (6), 742 (2001)

Force curves on nanotube carpet: numerical simulation



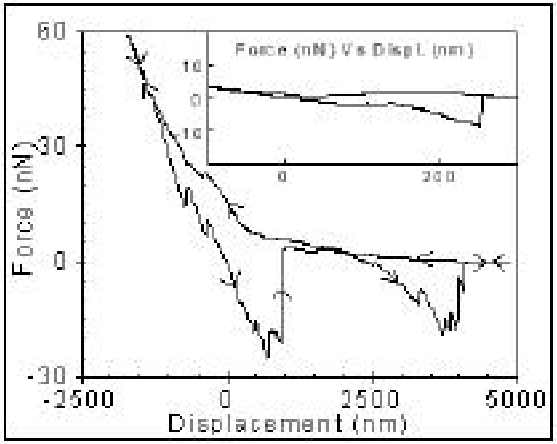
Simulated force curves with a 50nanotube distribution

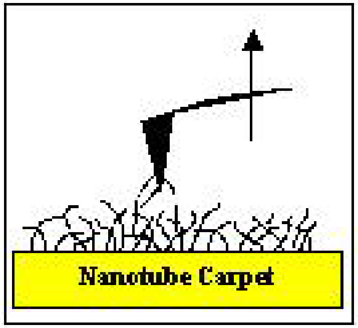
Force curves on nanotube carpet: numerical simulation

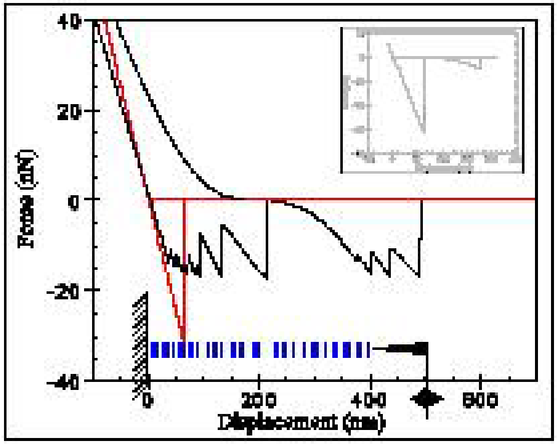


Reproducibility of force curves for three different 50 nanotube distributions

Conclusion on the second part: tribology on a nanotube carpet





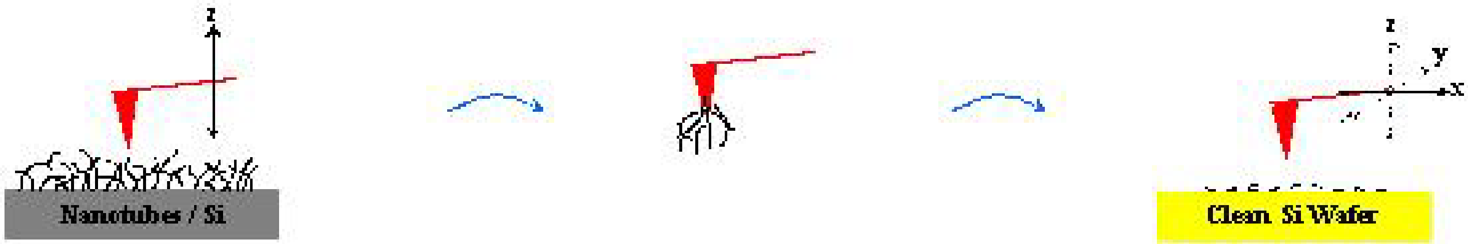


Here tip moved at constant speed! ⟶ Force variation

Constant force ⟶ carpet rheology:

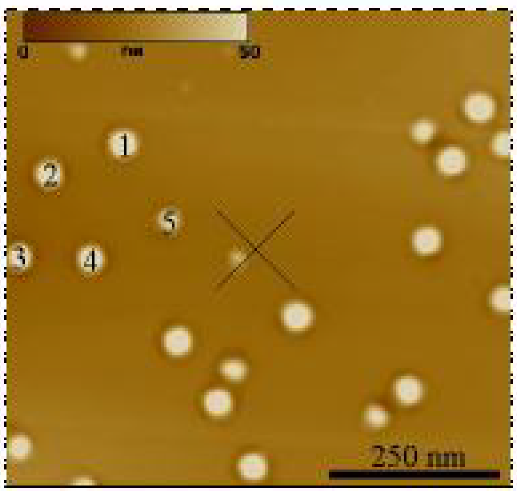
time variation of deformation

Transport and absolute positioning of nanotubes

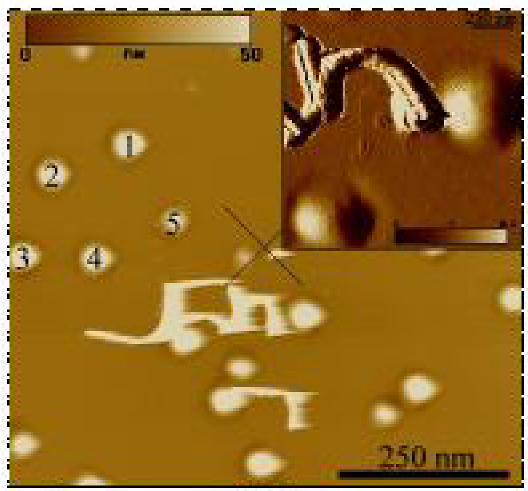


1 / Force curves on nanotube carpet

2 / Images on Si wafer after nanotube deposition



Before Deposition
Contact mode



After Deposition
Tapping mode

Deposition method

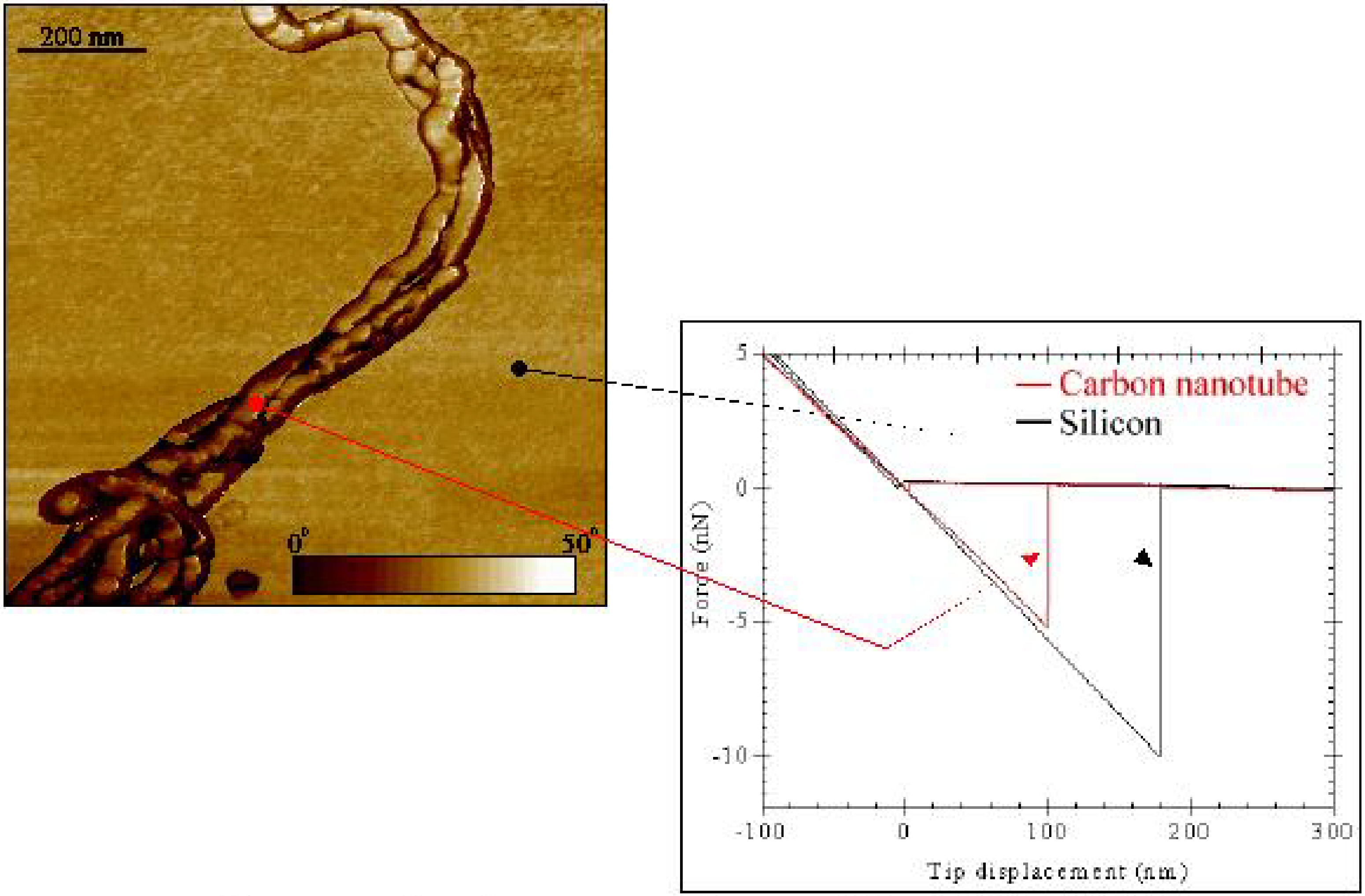
- precision ≈ 500 nm
- No wet chemistry
- limited number ofnanotube

Change of the Tip possible

- All the AFM modes
- Work with a clean tip

S. Decossas et al., In preparation

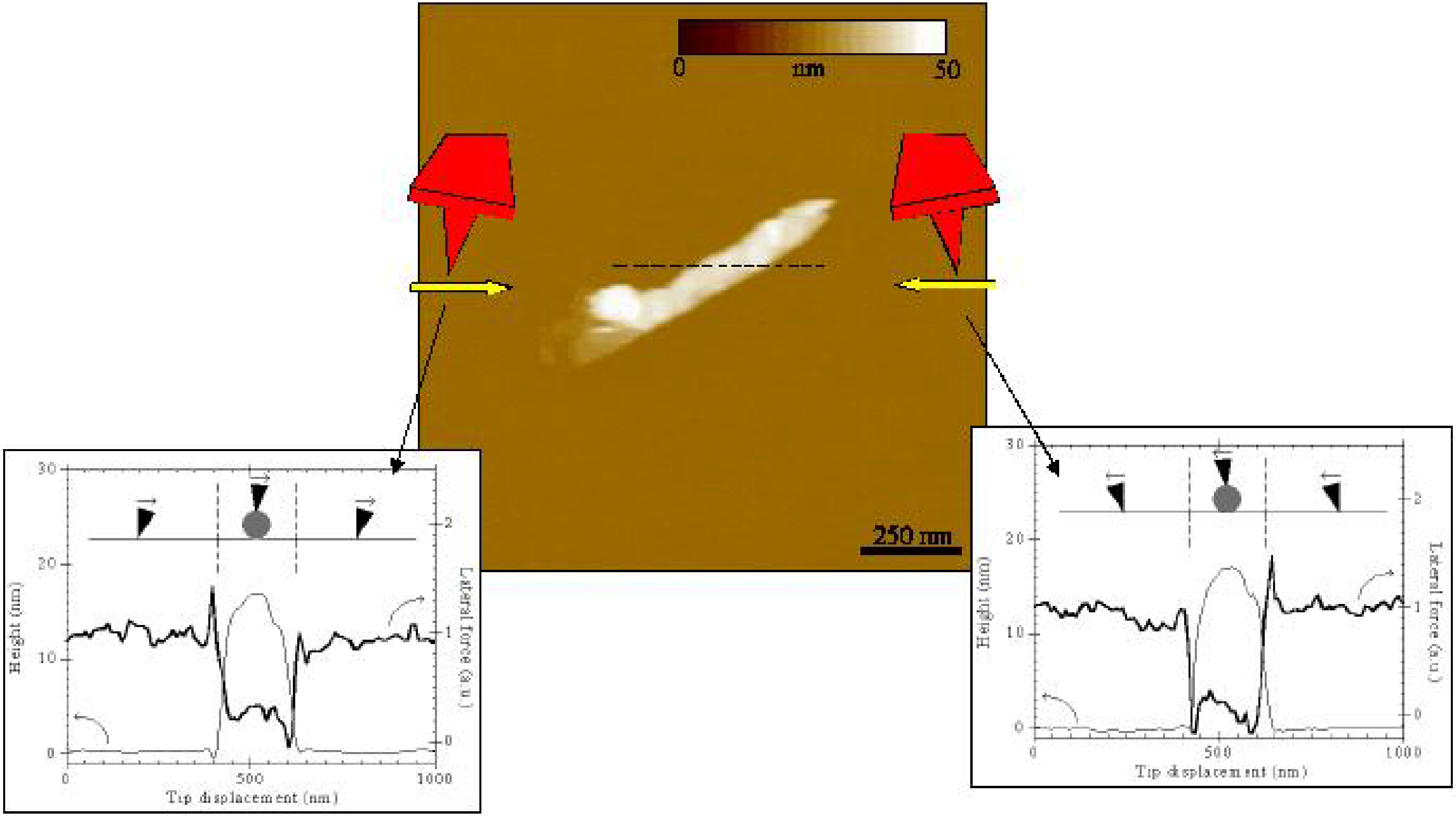
Adhesion measurement on isolated nanotubes



Whatever is the atmosphere (air or dry N_2):

$$\textbf{Adhesion}_{\textbf{Silicon}} \approx 2 \times \textbf{Adhesion}_{\textbf{nanotube}}$$

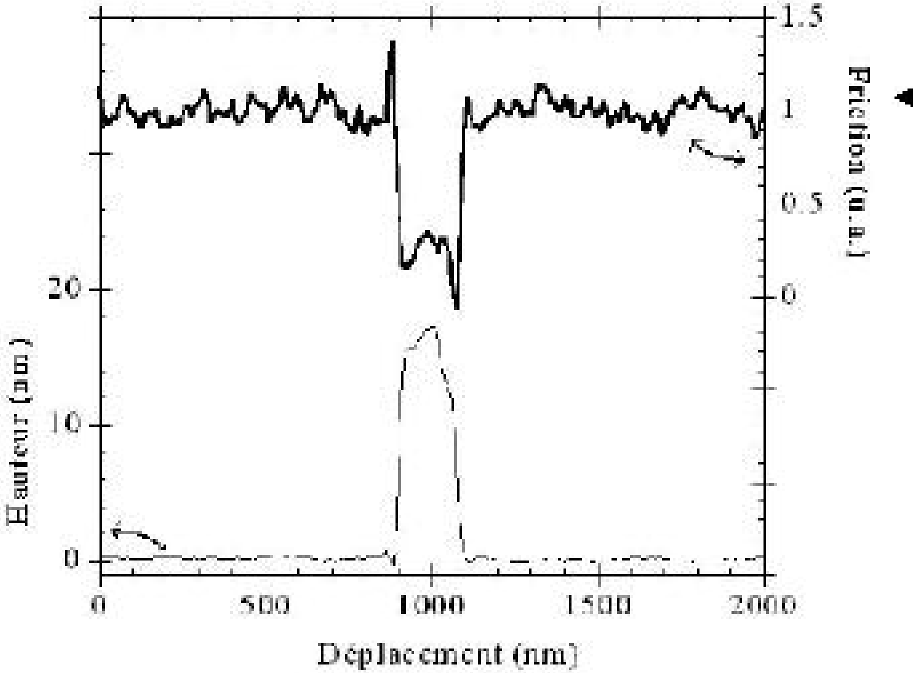
Friction measurement on isolated nanotube



Whatever is the atmosphere (air or dry N_2):

$$\textbf{Friction}_{\textbf{Silicon}} \approx 5 \times \textbf{Friction}_{\textbf{nanotube}}$$

Friction
on carbon nanotubes



Friction on tuniciers