

Longitudinal vibrations of suspended carbon nanotubes - Franck-Condon effect, cotunneling, and nonequilibrium

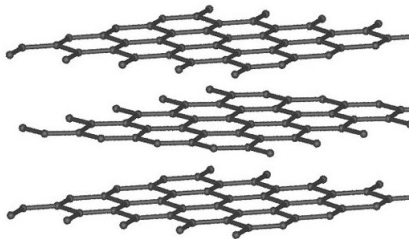
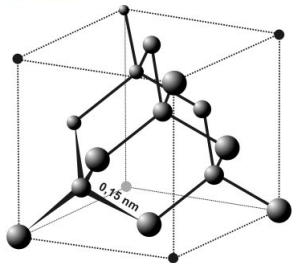
Andreas K. Hüttel

Kavli Institute for Nanoscience,
Technische Universiteit Delft, Netherlands

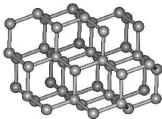
Current address:
Institute for Experimental and Applied Physics,
Universität Regensburg, Germany

Workshop on Nano-Opto-Electro-Mechanical Systems Approaching the Quantum Regime,
Abdus Salam International Centre for Theoretical Physics, Trieste 2010

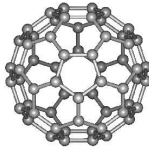
Carbon, as we know it



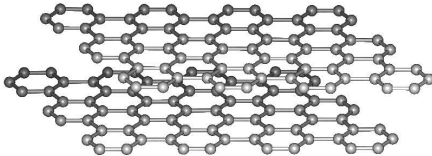
Carbon nanotubes: a more exciting form of carbon



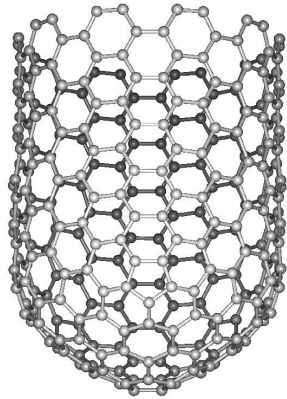
diamond



fullerene (C₆₀)



graphite / graphene



nanotube

Carbon nanotubes

- different production methods; often:
 - use small catalyst particles
 - hot gas, with carbon feed (e.g. CH_4)
 - nucleation of tube structure
- many different structures
 - single-wall, double-wall, multi-wall
 - zigzag, armchair, chiral (how the sheet is “wrapped together”)



Mechanical properties of carbon nanotubes

- stiffer than steel
- resistant to damage from physical forces
- very light

- Young's modulus $E = \frac{F/A}{\Delta L/L}$:
 $E_{\text{CNT}} \simeq 1.2 \text{ TPa}$, $E_{\text{steel}} \simeq 0.2 \text{ TPa}$

- Density:

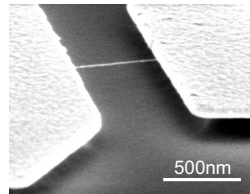
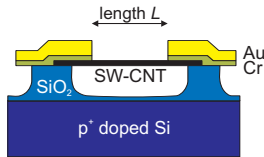
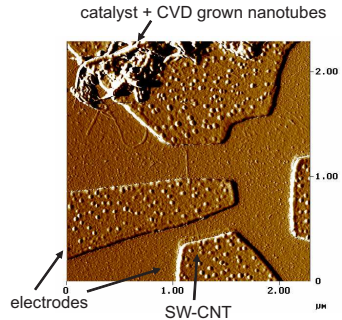
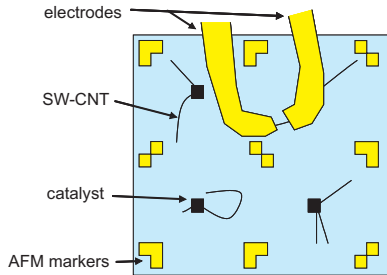
$$\rho_{\text{CNT}} \simeq 1.3 \frac{\text{g}}{\text{cm}^3}, \quad \rho_{\text{Al}} \simeq 2.7 \frac{\text{g}}{\text{cm}^3}$$

- (still) “material of dreams”



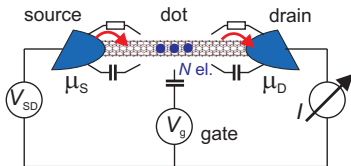
Suspended carbon nanotube sample fabrication

“the old way of doing things”

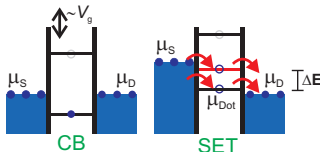
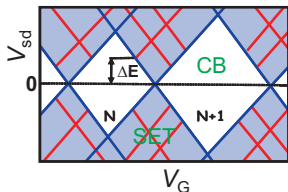


Low-temperature transport measurements

- Tunnel barriers between leads and nanotube
- Low temperature $k_B T \ll e^2/C$: formation of a **quantum dot**



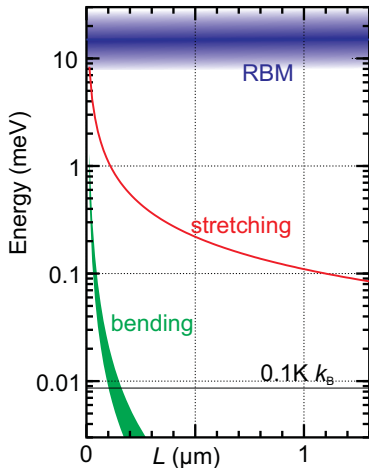
stability diagram: $\frac{dI}{dV_{SD}}(V_g, V_{SD})$



CB - Coulomb blockade “diamonds”
SET - single electron tunneling

Excited states visible at finite bias!
Spectroscopy of the electronic system

Vibration modes of carbon nanotubes



- radial breathing mode(s)

- **stretching** (longitudinal) mode:

$$h\nu \propto L^{-1}$$

$$h\nu = 1100 \dots 110 \mu\text{eV},$$

$$\nu = 270 \dots 27 \text{GHz}$$

(for 100 nm ... 1 μm)

- **bending** (transversal) mode:

$$h\nu \propto L^{-2}$$

$$h\nu = 10 \dots 0.1 \mu\text{eV},$$

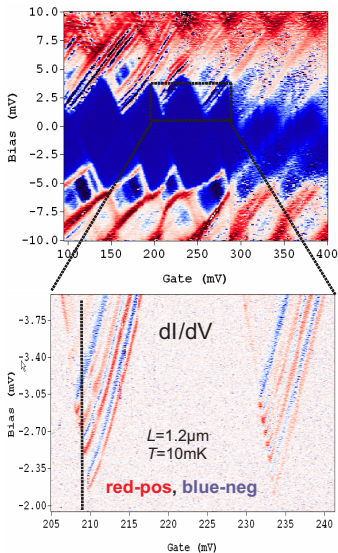
$$\nu = 2.4 \text{GHz} \dots 24 \text{MHz}$$

(for 100 nm ... 1 μm)

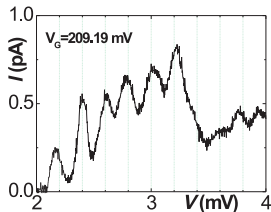
$h\nu \propto d$, also tension-dependent

purely electronic excitations have different energy scale

The stretching mode – visible in electronic transport



- Low-energy excitations
- Equally spaced, $\hbar\omega = 140\mu\text{eV}$
- Identical for different charge states
- **Stretching mode** as harmonic oscillator

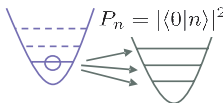
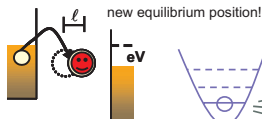


Electron-vibron coupling, Franck-Condon physics

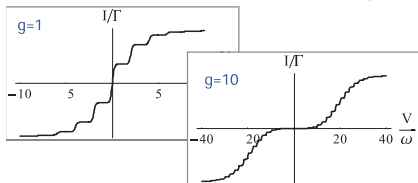
$$\hat{H} = \frac{\hat{p}^2}{2m} + \frac{1}{2}m\omega_0^2\hat{x}^2 + \lambda\hat{x}$$

$$g = \frac{\lambda^2}{2} = \frac{1}{2} \left(\frac{\ell}{\ell_0} \right)^2$$

$$\ell_0 = \sqrt{\frac{\hbar}{m\omega_0}}$$



$$\Gamma \rightarrow \Gamma_{el} \underbrace{|\langle \Psi_{\text{after}} | \Psi_{\text{before}} \rangle|^2}_{P_{nm}}$$



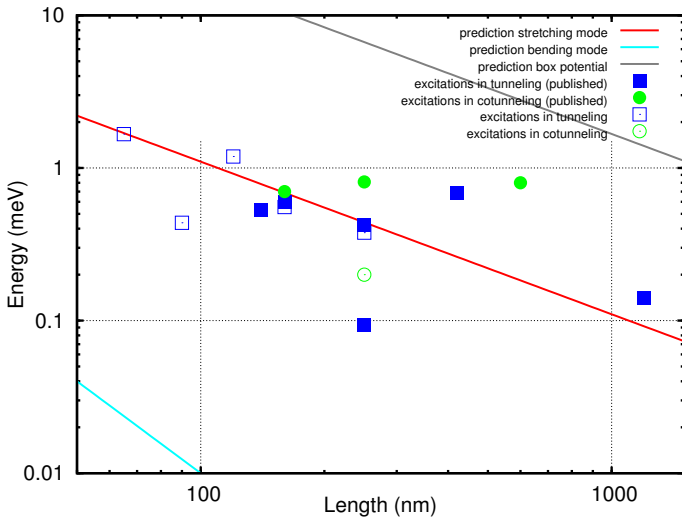
$$P_{n0} = |\langle \Psi(x - \ell) | \Psi(x) \rangle|^2 = \frac{e^{-g} g^n}{n!}$$

no effect for $g < 0.1$

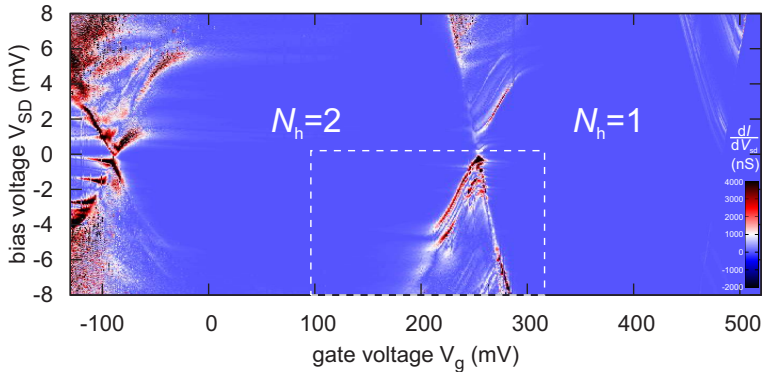
additional steps in $I(V_{sd})$ for $g > 0.1$

phonon blockade of transport for $g > 1$, $V_{sd} < g\hbar\omega_0$

Vibrational excitations observed so far



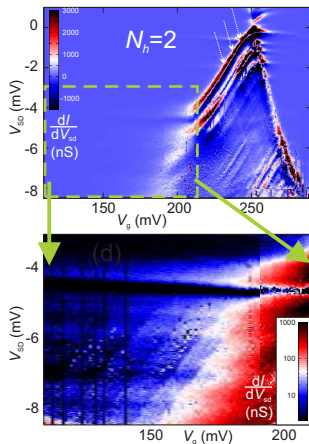
$L = 250$ nm SC nanotube, few-hole regime



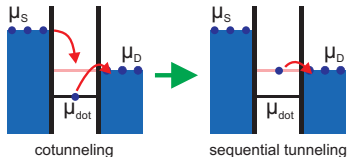
$E_{\text{gap}} = 0.2 \text{ eV} \rightarrow d = 3.7 \text{ nm}$, $\hbar\omega_{\text{RBM}} \simeq 7.8 \text{ meV}$, maybe (0,46), $\varepsilon \simeq 6.2 \text{ meV}$
 length $L = 250 \text{ nm} \rightarrow \text{expected } \hbar\omega_{\text{bend}} \simeq 0.002 \text{ meV}$, $\hbar\omega_{\text{stretch}} \simeq 0.44 \text{ meV}$

bending lines $\rightarrow$ shifting potential minima, DQD-like properties

Stretching mode in SET and cotunneling ($1 \leq N_{h+} \leq 2$)

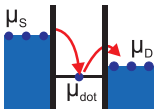


- excitations in SET, positive slope:
harmonic, $\Delta\epsilon = 0.42 \text{ meV} \simeq \hbar\omega_{\text{stretch}}$
- harmonic excitations in **cotunneling**!
- Cotunnel-assisted sequential tunneling, “CO-SET”

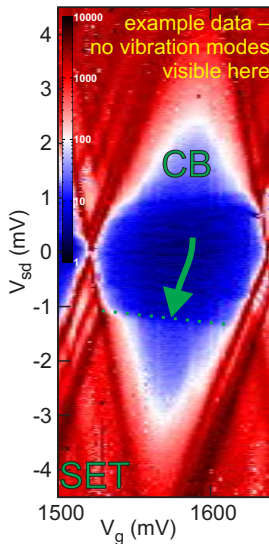
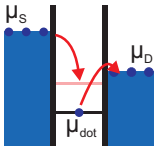


Reminder: cotunneling – second-order process

- current in Coulomb blockade:
“several electrons tunneling at once”
- two-electron processes:
 - elastic:

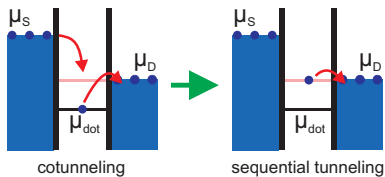


- inelastic (green arrow):

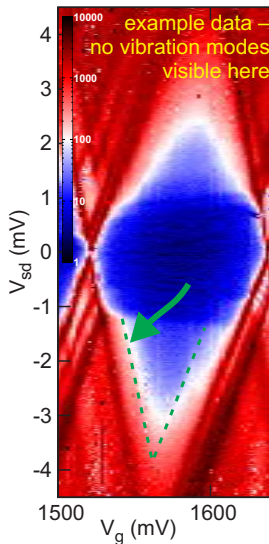


Cotunnel-assisted sequential tunneling (CO-SET)

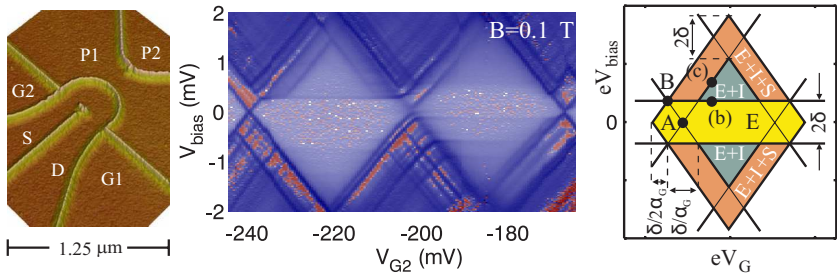
- inelastic cotunneling, followed by a tunnel-out process



- requires energy storage
- this is the process we've seen
- requires energy storage:
tunnel-out must be faster than relaxation

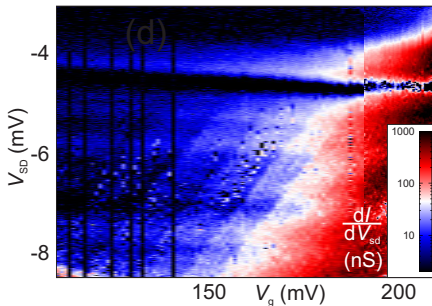


Cotunnel-assisted sequential tunneling (CO-SET)



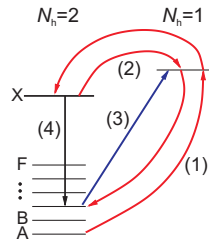
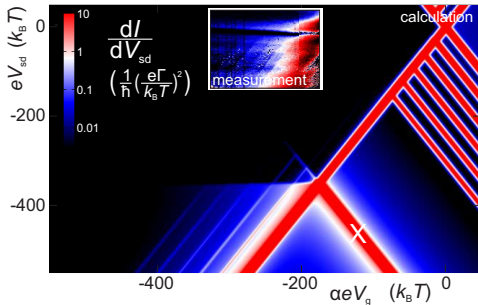
- first observed and explained by Schleser *et al.* ~ 2005
electronic excitations in GaAs/AlGaAs quantum dots

Measurement detail



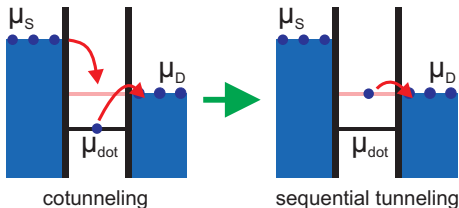
- CO-SET current sets in at additional (electronic) excited state X
- Tunnel rates coupling an 2h state to 1h ground state:
 small for 2h ground state, **large** for 2h excited state X
- Real-time transport theory calculations, M. Leijnse & M. Wegewijs
- Vibration mode is **pumped** by multiple inelastic cotunnel processes involving X (e.g. sequence $(1) \rightarrow (2) \rightarrow (3)$)

Numerical calculation



- CO-SET current sets in at additional (electronic) excited state X
- Tunnel rates coupling an 2h state to 1h ground state:
small for 2h ground state, large for 2h excited state X
- Real-time transport theory calculations, M. Leijnse & M. Wegewijs
- Vibration mode is pumped by multiple inelastic cotunnel processes involving X (e.g. sequence $(1) \rightarrow (2) \rightarrow (3)$)

CO-SET process requires energy storage, nonequilibrium

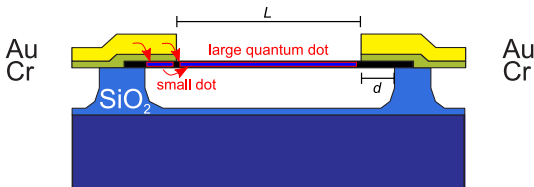


- Vibration mode must remain excited until tunnel-out
- Vibrons are pumped as in a three-level laser!
- Comparison of timescales & tunnelling rates
 - first (weak) lower boundary for mechanical quality factor
 - $Q_{\text{stretch}} \gtrsim 30$
- Known values for transversal CNT mode:

$$Q_{\text{bend,RT}} \lesssim 2000, \quad Q_{\text{bend,20mK}} \lesssim 150000$$

Open question #1: Nature of the excited state X

- simplest possibility: orbital excited state of the nanotube quantum dot
 - different orbital wavefunction
 - different tunnel couplings
 - our model idea should work fine
- alternative explanation: potential side minimum / double quantum dot
 - possible since the suspended nanotube is partially covered by the contacts
 - bending resonance lines in Coulomb diamonds: shifting potential minima
 - our model idea should still work fine!

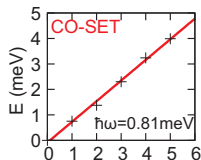
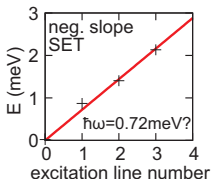
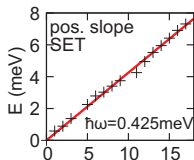


Some speculations about a “phaser”



- idea: use analogy with the 3-level laser
- current pumps vibration via the electronic excited state
- use a double quantum dot to generate this level structure
- feed a mechanical mode faster than it can decay, population inversion
- ?

Open question #2: Frequency doubling



- pos. slope SET excitations: $\hbar\omega = 0.42 \text{ meV}$
- CO-SET and neg. slope SET excitations: $\hbar\omega \simeq 0.8 \text{ meV}$
- **few-carrier system**
- adding a hole redistributes the entire charge on the nanotube
- up to now, all measurements of the Franck-Condon sidebands were in the metallic limit

Some speculations about 1D Wigner crystals

- few charge carrier limit
- one-dimensional chain of electrons or holes
- different charge number $\longrightarrow$ completely different charge distribution
- influence of charge distribution on the electrostatic forces inducing the vibrations
- **dynamic interaction?**
- electronic system much faster than vibration
 $\longrightarrow$ can regard mechanical oscillator fixed for each point in time
- ?

The team at Molecular Electronics & Devices, Delft and theory friends

Thanks!



Herre van der Zant



Benoit Witkamp



Hari Pathangi



Menno Poot



Martin Leijnse



Maarten Wegewijs

& Samir Etaki, Yaroslav Blanter, Fabio Pistolessi, Ivar Martin, Sami Sapsmaz, Pablo Jarillo-Herrero, Raymond Schouten, Hidde Westra, ...

Meanwhile, things have moved on a bit...

... and me as well, back to Bavaria

- new research group at Universität Regensburg
- spin injection and spin transport in carbon nanotubes
- carbon nanotubes with superconducting contacts
- and now (since funding has finally come):
carbon nanotubes as nano-electromechanical hybrid systems
- **postdoc position available**

