Designing Carbon-Based Nanotechnology on a Supercomputer

David Tománek

Michigan State University

tomanek@msu.edu
http://www.pa.msu.edu/~tomanek

Acknowledgements

Gianaurelio Cuniberti, *University of Regensburg*
Yoshiyuki Miyamoto, *N.E.C. Tsukuba, Japan*
Norbert Nemec, *University of Regensburg*
Angel Rubio, *University of Pais Vasco, Spain*

Financial Support:

NSF-NSEC
NSF-NIRT

JAMSTEC-ESC (Japan)
RIST (Japan)

Outline

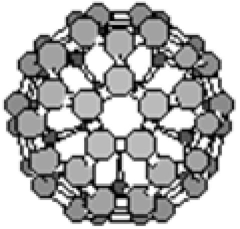
- Introduction
  - **Carbon nanotubes: Ideal building blocks for nanotechnology?**
  - **Computational tools**
- Can computation guide nanomanufacturing?
  - **What limits the frequency response of nanotube electronics?**
  - **How to best contact a carbon nanotube?**
  - **How to cure atomic-scale defects?**
- Summary and Conclusions

- Printed Review:

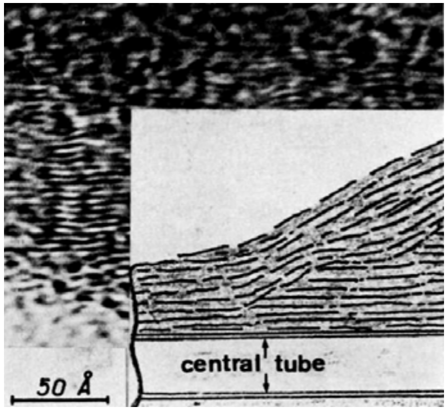
  David Tománek, Carbon-based nanotechnology on a supercomputer, Topical Review in
  J. Phys.: Condens. Matter **17**, R413-R459 (2005).

Nanocarbon pioneers

- The C_{60} 'buckyball' and other fullerenes:
  - successful synthesis
  - potential applications:
    - lubrication
    - superconductivity



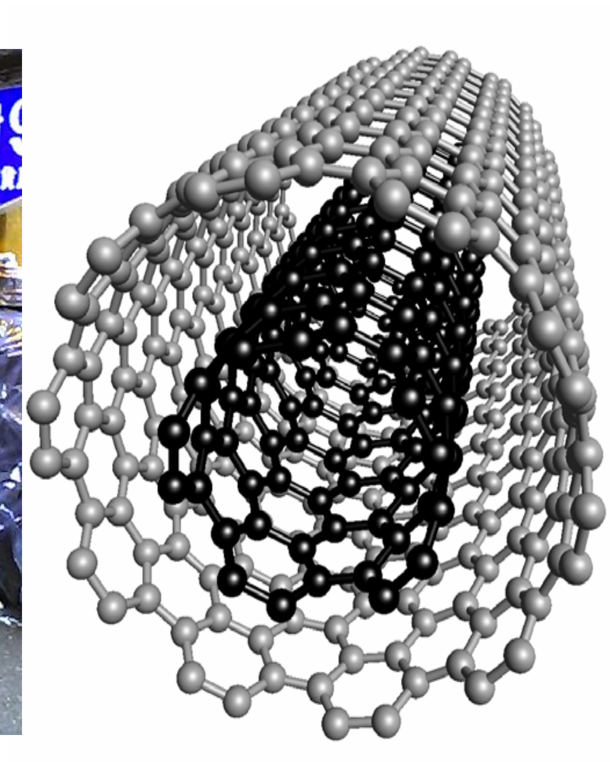
- Nanotubes:
  - successful synthesis
  - potential applications:



Nanotubes in the core of carbon fibers: A. Oberlin, M. Endo, and T. Koyama, J. Cryst. Grow. 32 (1976) 335-349

composites	flat-panel displays
Li-ion batteries	super-capacitors
medication delivery	fuel cells
EMI shielding	hydrogen storage

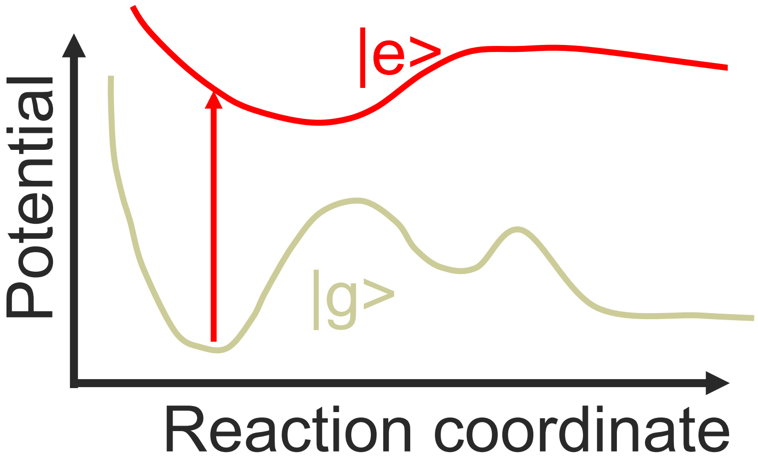
Carbon nanotubes:
Ideal building blocks for nanotechnology?



- 1-20 nm diameter
- Atomically perfect
- Chemically inert
- 100 times stronger than steel
- Extremely high melting temperature
- Ideal (ballistic) conductors of electrons, or insulators
- Ideal heat conductors
- Non-toxic

Computational Approach to Nanostructures

What approach to use?



Ground state dynamics:
Solve the eigenvalue problem:

$$H\psi_n = \varepsilon_n \psi_n$$

Density Functional Theory (codes including SIESTA, VASP, CASTEP, GAUSSIAN, etc.)

Excited state dynamics:
Solve the time-dependent problem:

$$i\hbar \frac{d\psi_n}{dt} = H\psi_n$$

FPSEID (éf-psái-dí:)

First-**P**rinciples
Simulation tool for
Electron-**I**on **D**ynamics

- Based on time-dependent density functional theory (TDDFT):
  E. Runge and E. K. U. Gross, Phys. Rev. Lett. ***52****, 997 (1984).*
- Computational details for real-time MD simulations:
  Sugino & Miyamoto PRB 59, 2579 (1999) ; ibid, B 66, 89901(E) (2002),
  using the Suzuki-Trotter split operator method to compute the time-propagator

Need massively parallel computer architectures and suitable algorithms distribute load over processors for speed-up

Computational Nanotechnology Laboratory:

Earth Simulator, Tokyo

www.nytimes.com

The New York Times ON THE WEB

April 20, 2002

Japanese Computer Is World's Fastest, as U.S. Falls Back

By JOHN MARKOFF

SAN FRANCISCO, April 19 — A Japanese laboratory has built the world's fastest computer, a machine so powerful that it matches the raw processing power of the 20 fastest American computers combined and far outstrips the previous leader, an I.B.M.-built machine.

Cost: $500,000,000

Maintenance: $50,000,000/year

<70% used for nano-carbons

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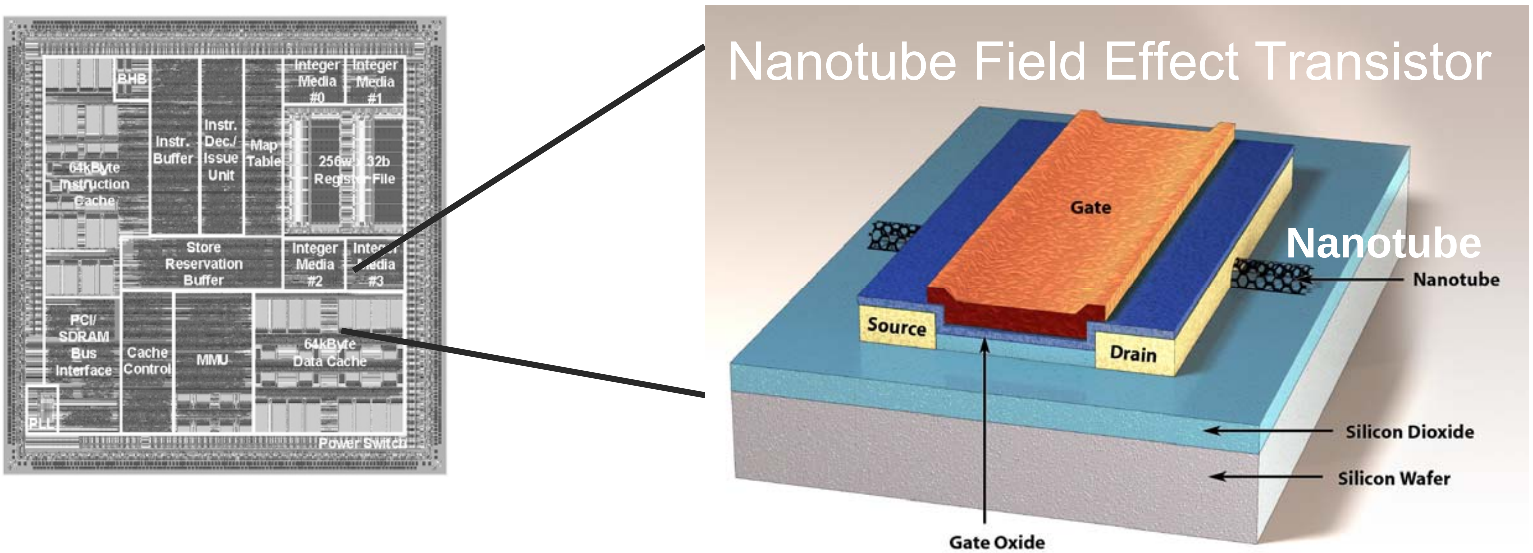
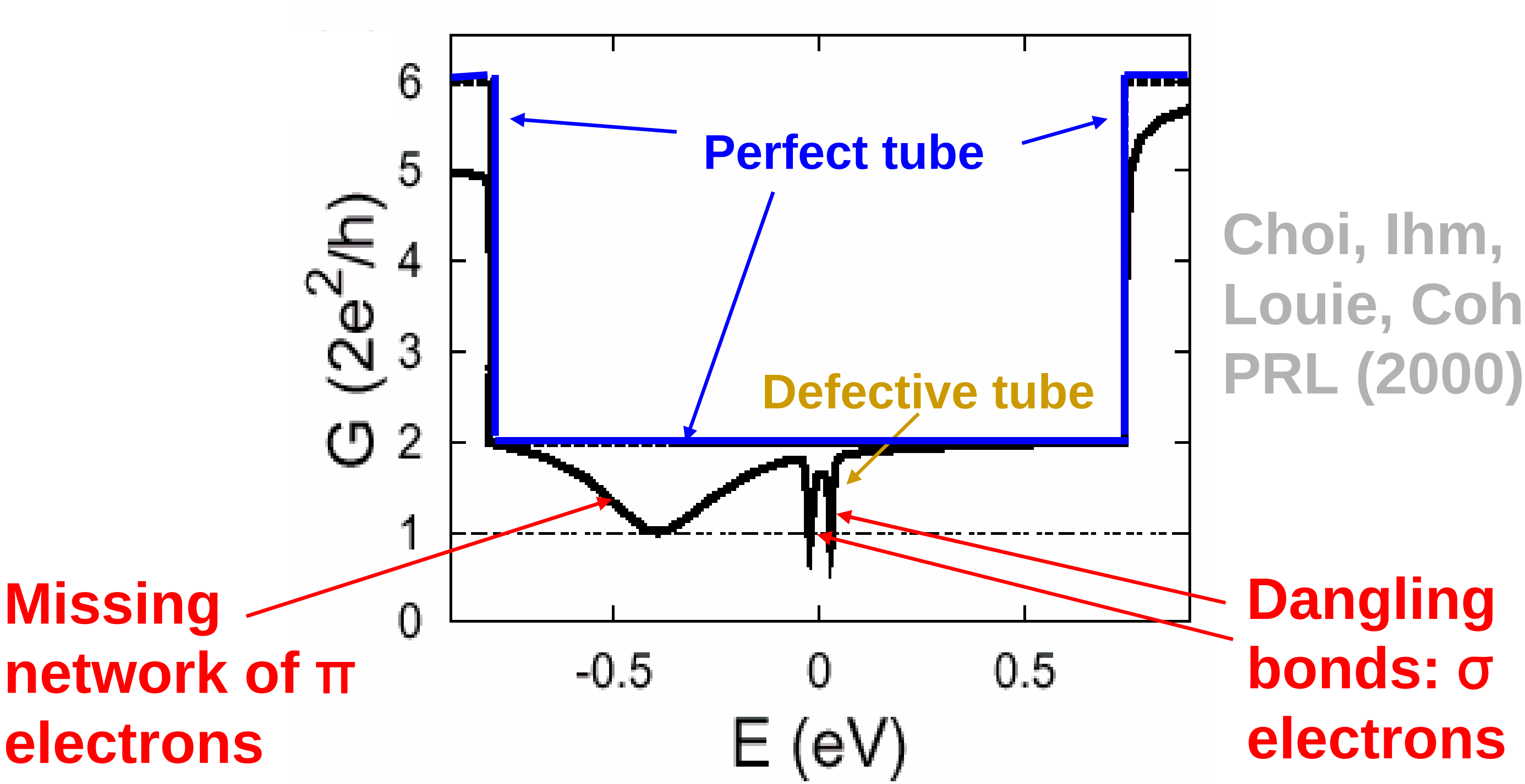
Can computation guide nanomanufacturing?



- What limits the speed of nanotube-based electronics?
- How to best contact a carbon nanotube?
- Are nanotube devices as sensitive to defects as Si-LSI circuits?
- Are there ways to selectively remove defects?

Quantum conductance of a (10,10) nanotube with a single vacancy

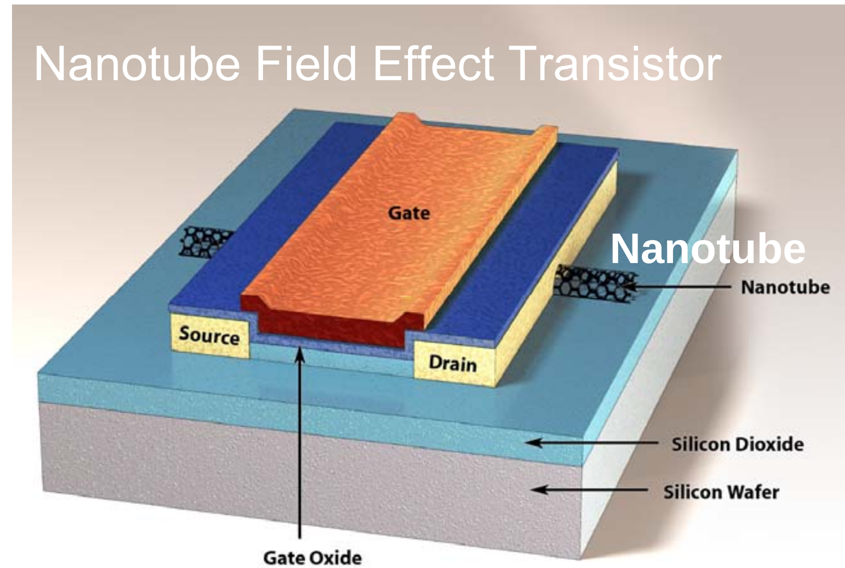


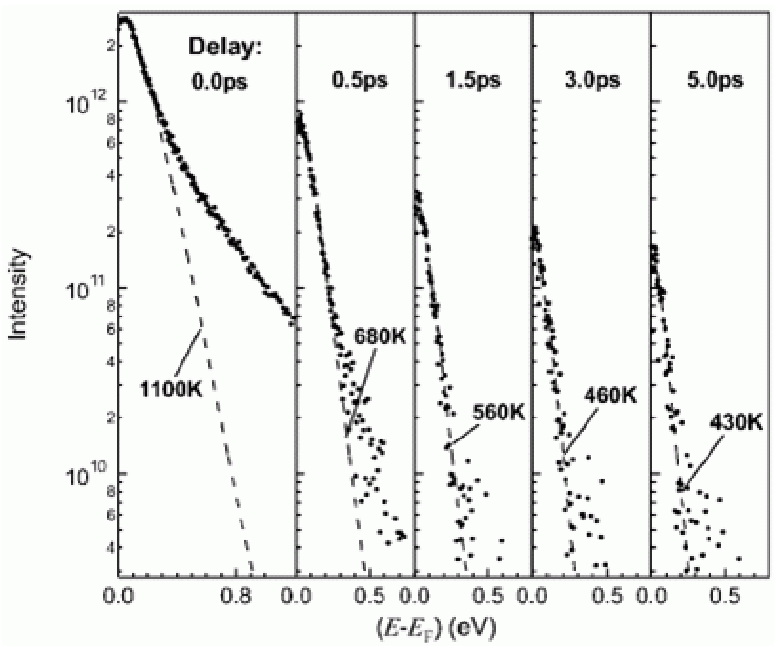
Choi, Ihm, Louie, Cohen, PRL (2000)

Individual defects significantly degrade conductance of a nanotube

What limits the frequency response of nanotube electronics?

- How useful are carbon nanotube devices (field-effect transistors, non-linear optical devices)?
- Maximum switching frequency:
  ➡ lifetime of excited carriers
- How long do electronic excitations last?
- What dampens electronic excitations:
  - Electron gas?
  - Phonons?



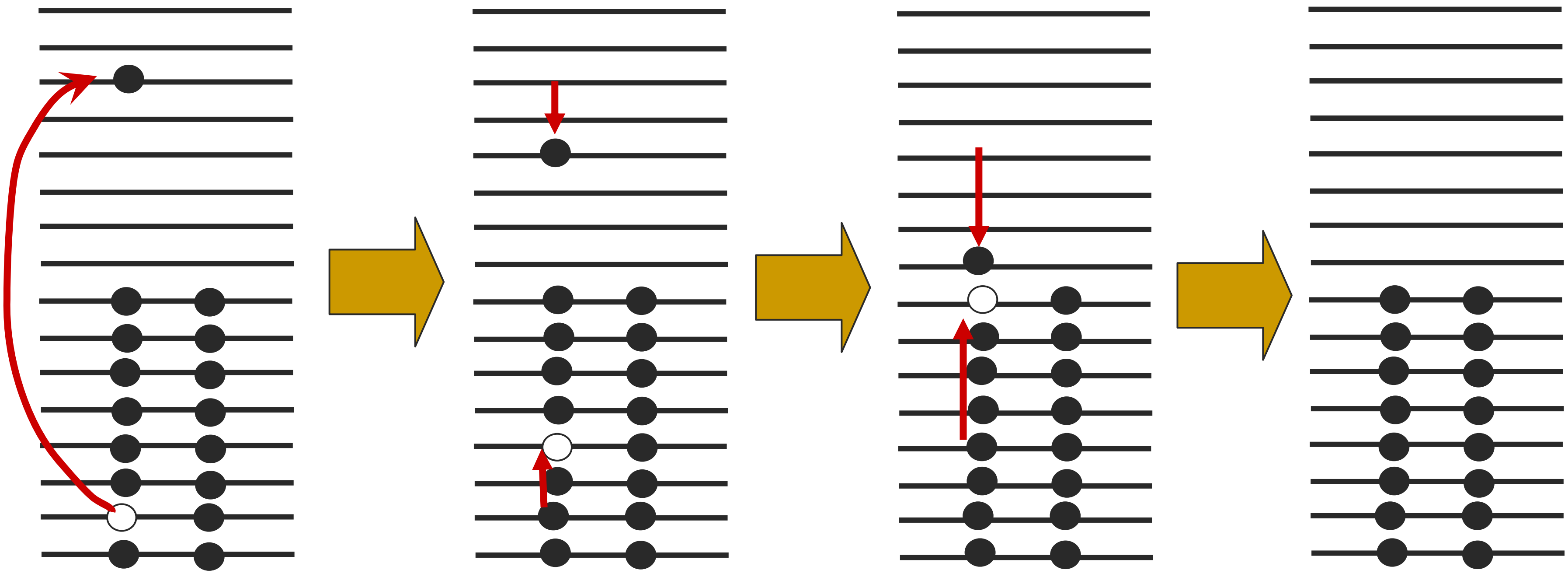


Evolution of photoelectron spectra as a function of pump-probe delay. At pump-probe delays of over 200 fs, the spectra can be well described by a Fermi-Dirac distribution (dashed lines).

Experiment: T. Hertel and G. Moos, PRL **84**, 5002 (2000)

Interpretation:
e-e comes before e-ph

Relaxation of hot carriers after a photo-excitation

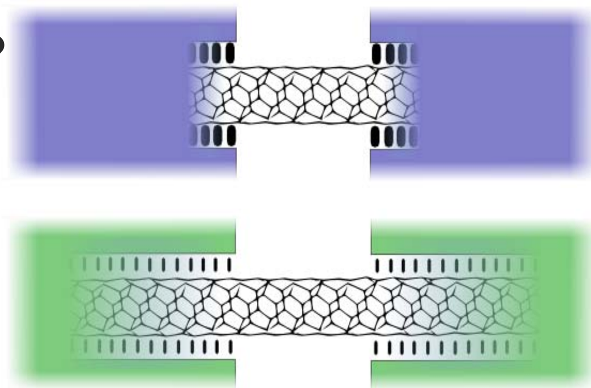


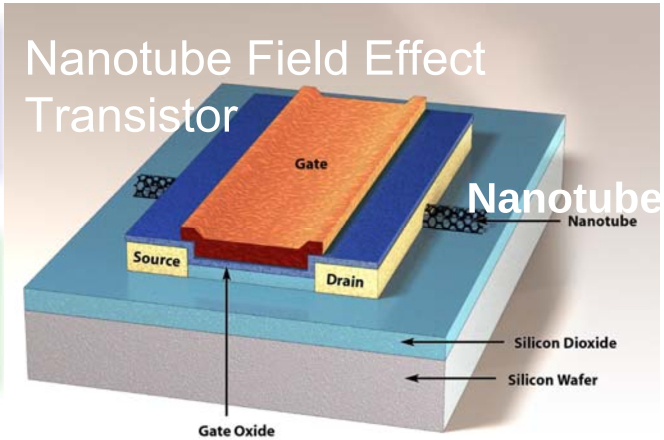
How to best contact a carbon nanotube?

- Which metal-nanotube contacts optimize charge injection?

• Short, strong contact?

• Long, weak contact?

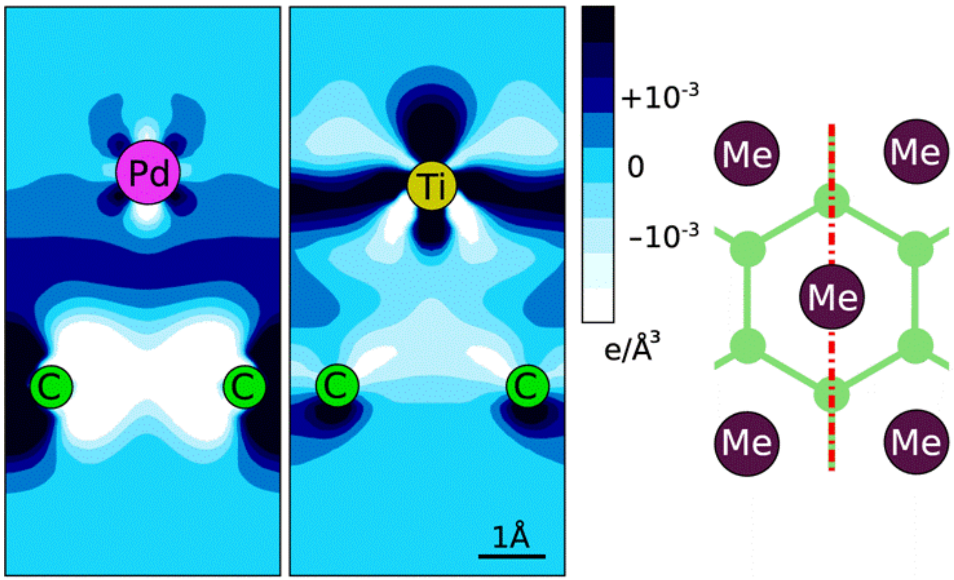




• Fermi momentum conservation?

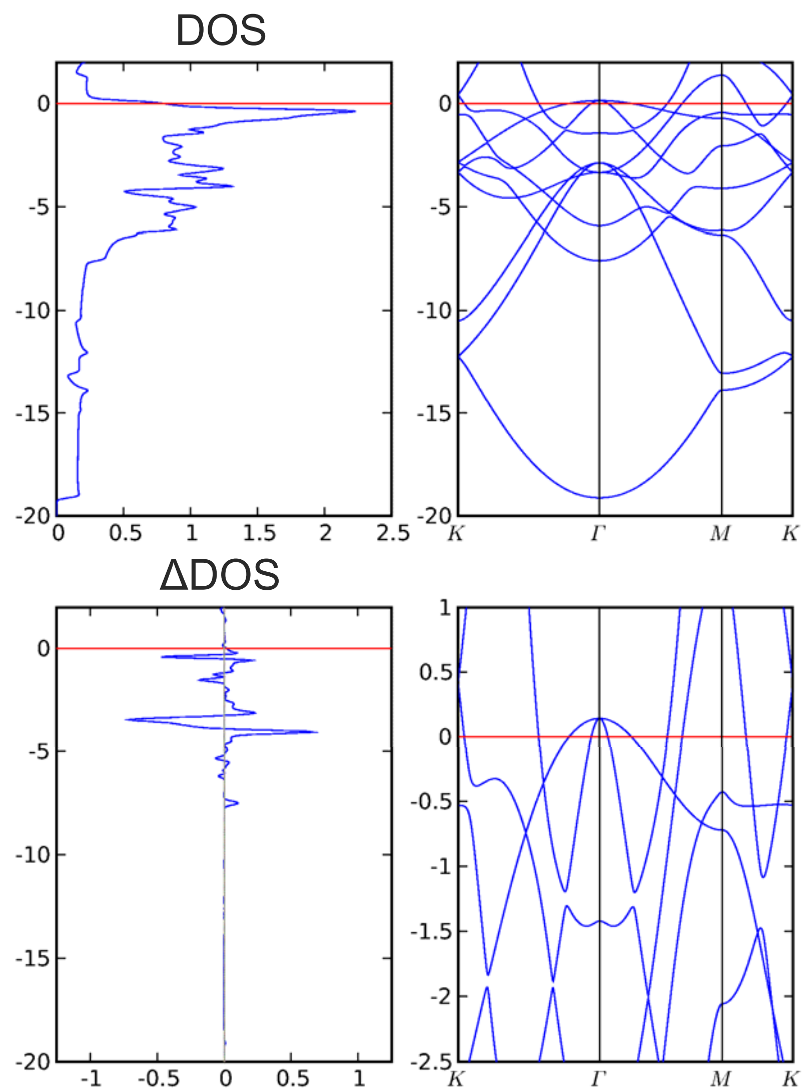
Norbert Nemec, David Tománek, and Gianaurelio Cuniberti, Phys. Rev. Lett. 96, 076802 (2006).

- Charge redistribution in Pd/graphite and Ti/graphite:



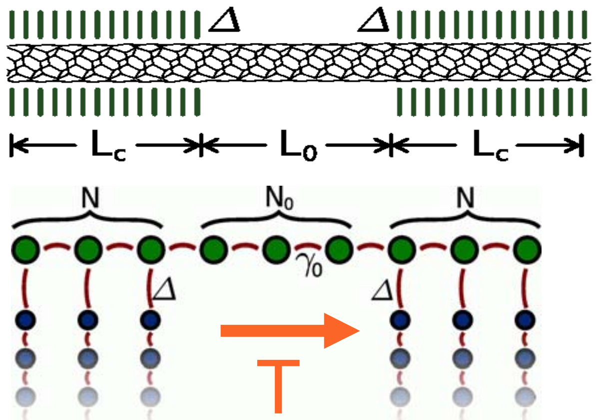
$$\Delta\rho(\mathbf{r}) = \rho_{C\&Pd}(\mathbf{r}) - \rho_{C}(\mathbf{r}) - \rho_{Pd}(\mathbf{r})$$
$$\Delta\rho(\mathbf{r}) = \rho_{C\&Ti}(\mathbf{r}) - \rho_{C}(\mathbf{r}) - \rho_{Ti}(\mathbf{r})$$

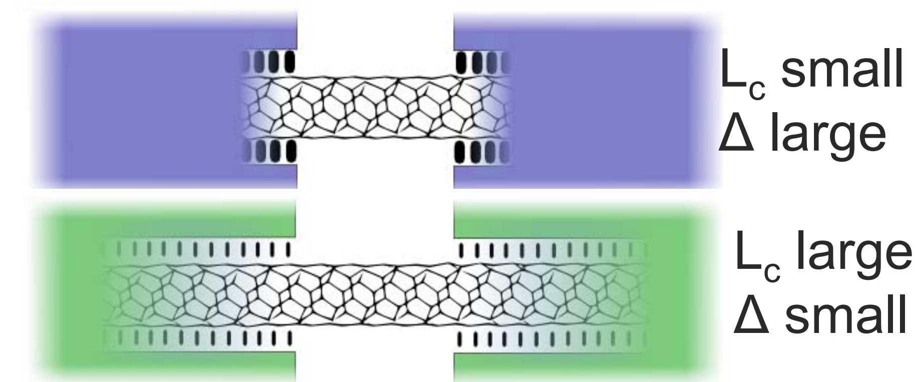
- Electronic structure of Pd/C:



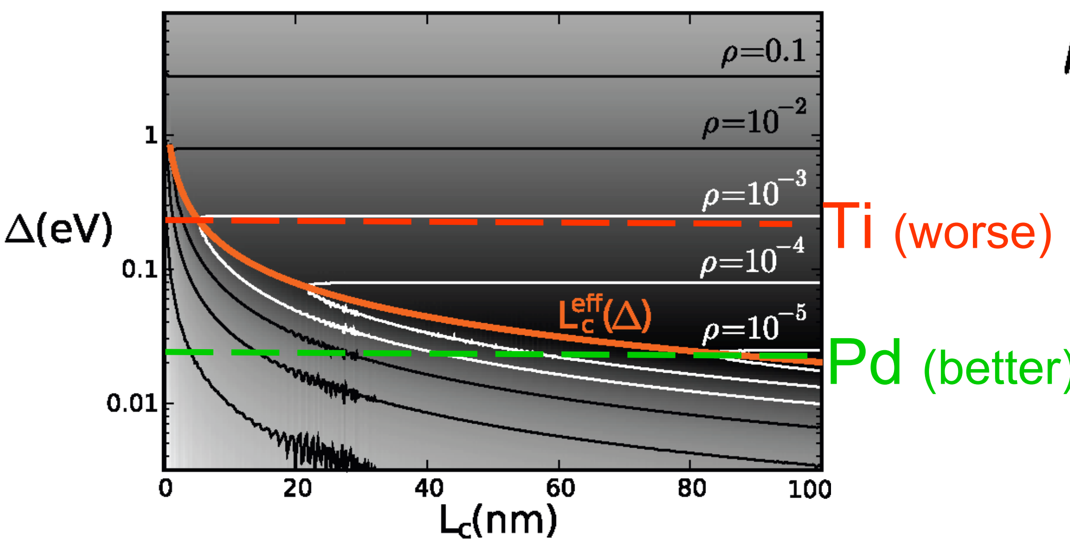
Pd/graphite:
Carrier injection with Fermi momentum of graphite seems possible

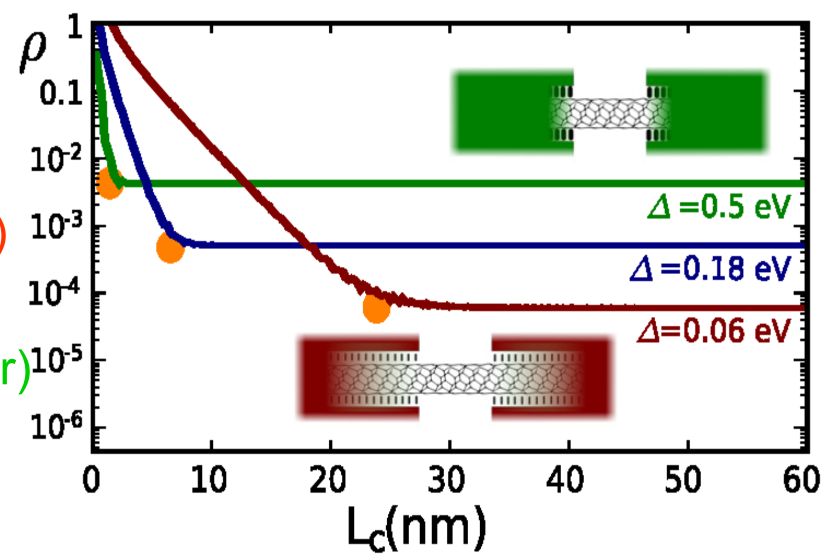
•Model of nanotube interacting with metal leads





•Transmission loss ΔT:



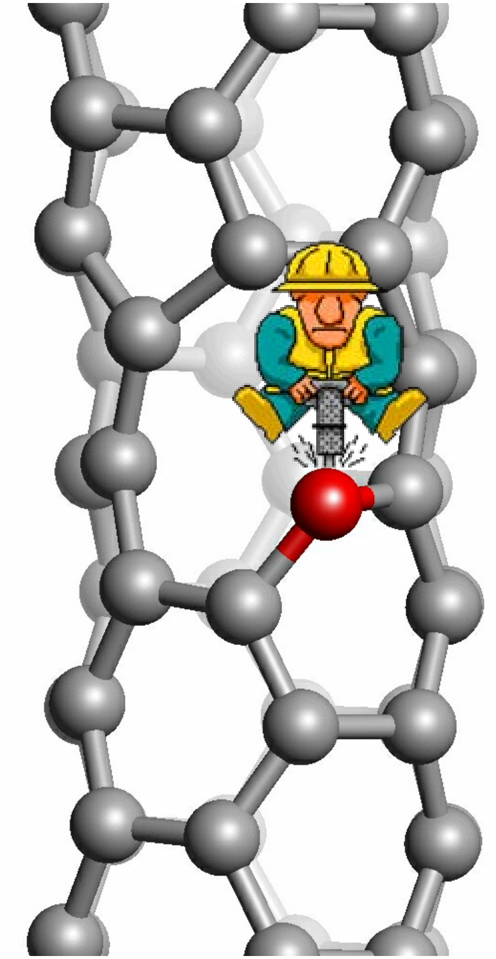


•Optimum contacts are *weak and long*

•Certain metals (Pd) are preferred over others (Ti, Au)

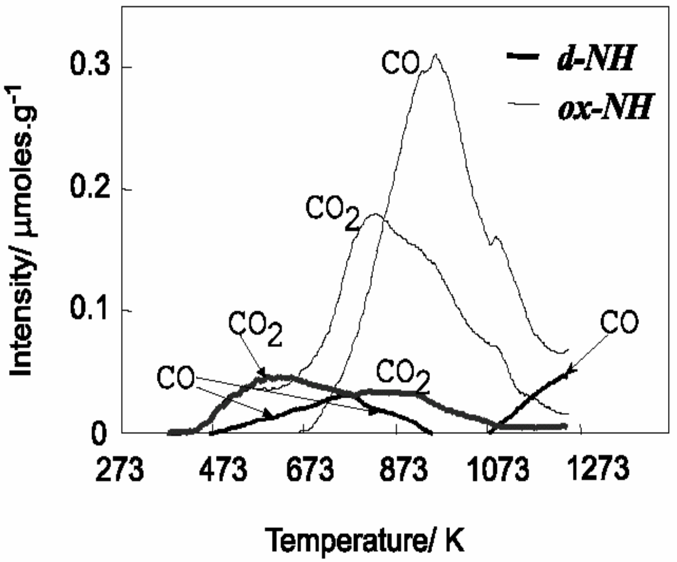
How to cure atomic-scale defects?

How to deoxidize?

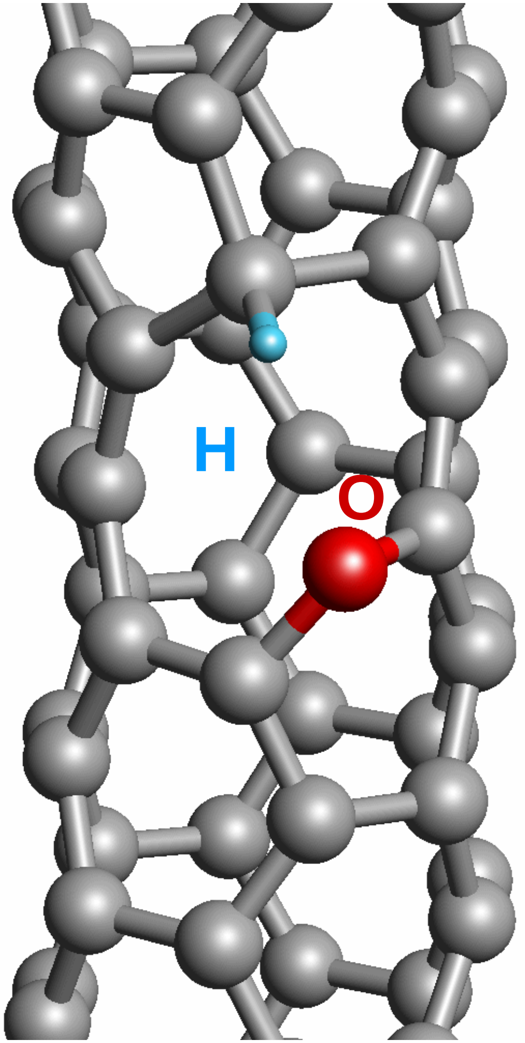


- **By heat treatment?**

⇒No: Larger damage to nanotube



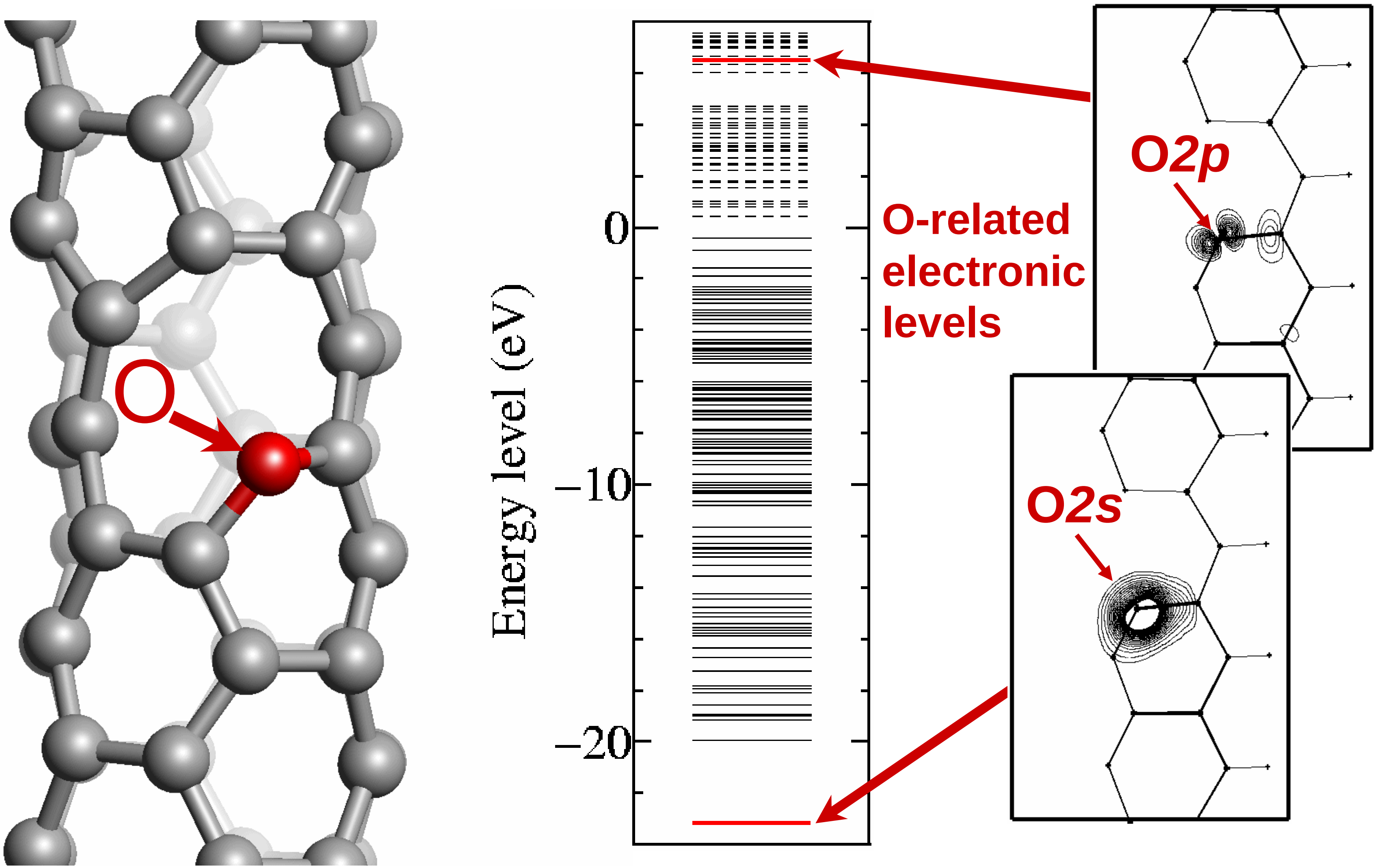
- **By chemical treatment with H?**



Y. Miyamoto, N. Jinbo, H. Nakamura, A. Rubio, and D. Tománek, Phys. Rev. B 70, 233408 (2004).

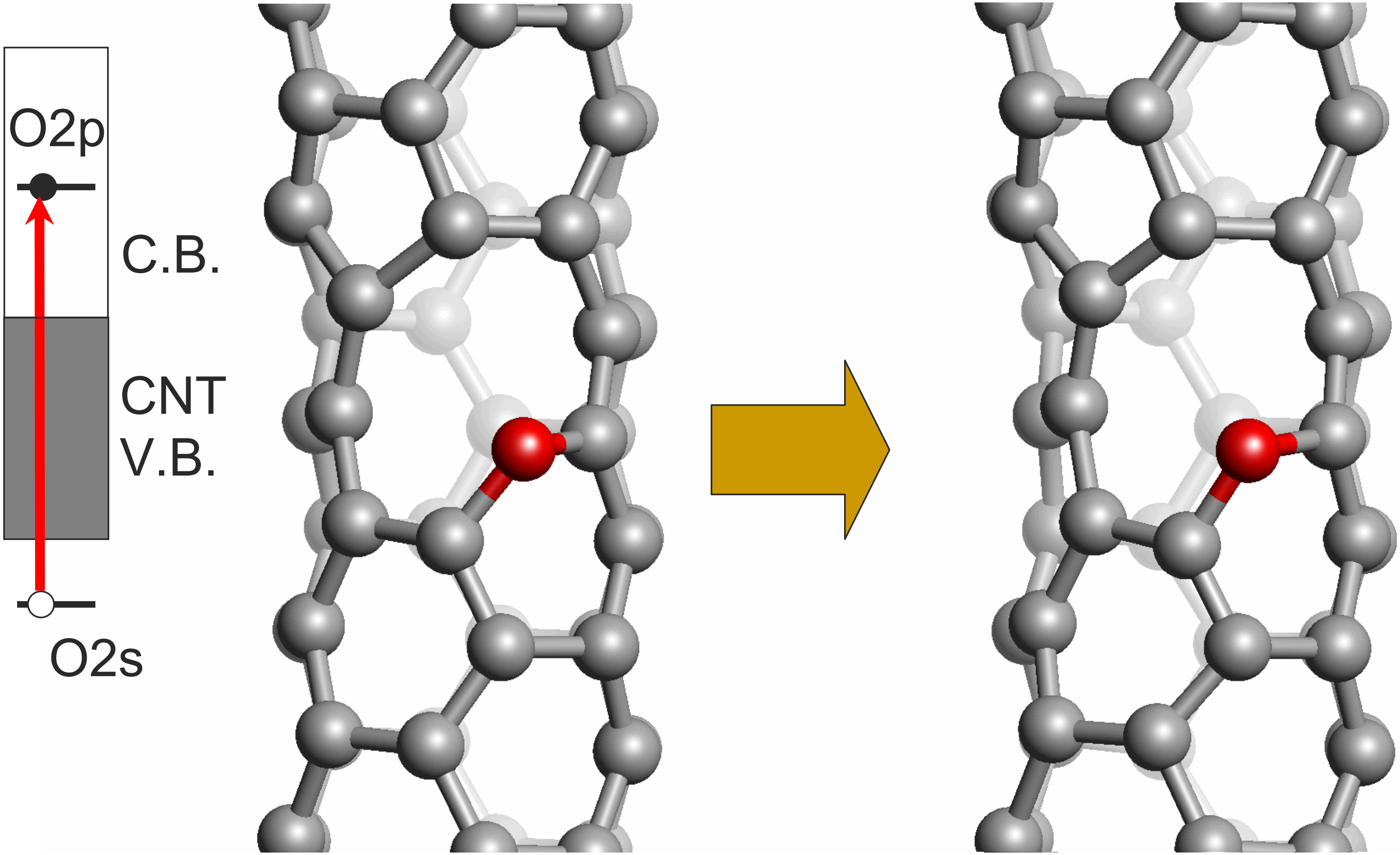
Alternative to thermal and chemical treatment

Electronic excitations!

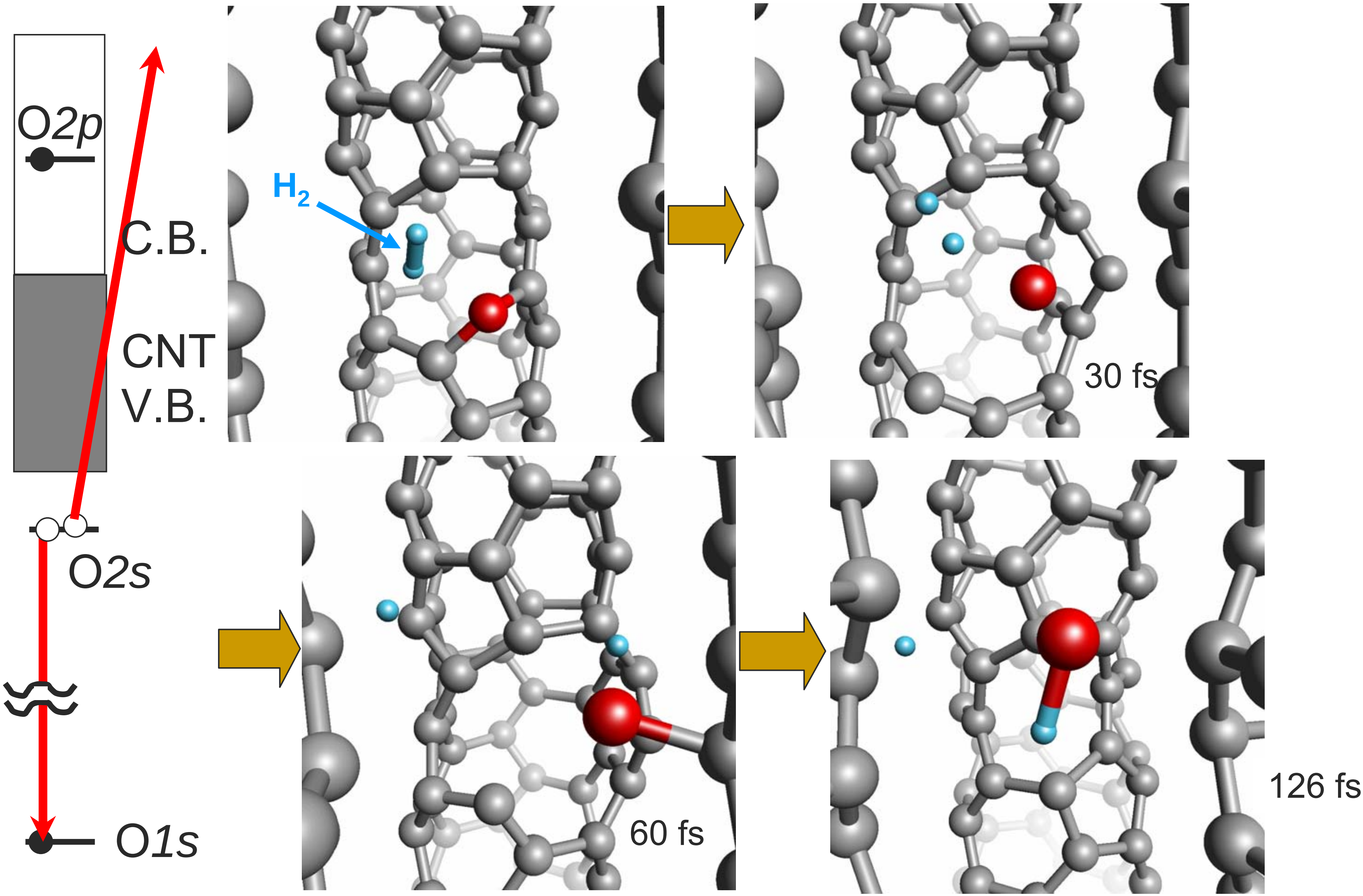


O2s → O2p excitation (33 eV)

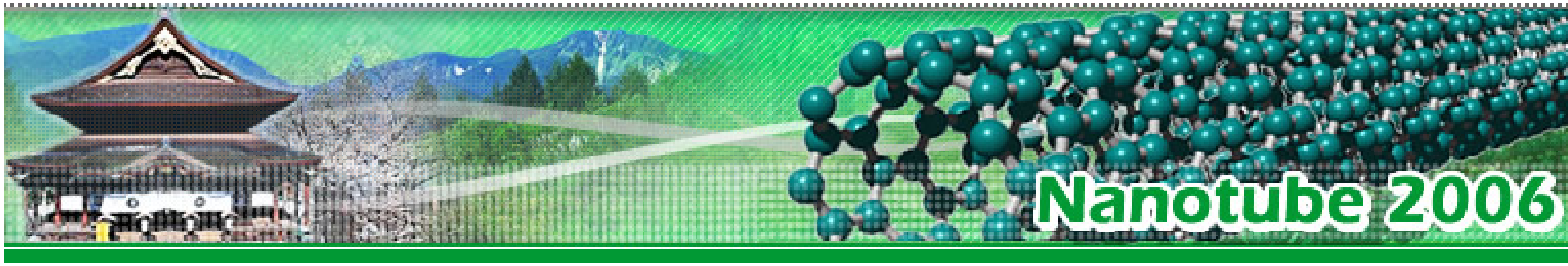
hopeless



Auger decay following the O1s → 2p excitation (~520 eV)



- Photoexcitations are long-lived
- Deoxidation by photo-surgery



Seventh International Conference on the Science and Application of Nanotubes

NT06

URL: **http://endomoribu.shinshu-u.ac.jp/nt06/**
or: **http://nanotube.msu.edu/nt06/**

Hotel Metropolitan
Nagano
Nagano, Japan
June 18-23, 2006

Summary and Conclusions

- Selected technological challenges in nanotube-based electronics can be best understood and solved by combining time-dependent DFT simulations with classical MD simulations.
- Electronic excitations in nanotubes exhibit ultrafast dynamics and decay by electronic and phonon channels.
- Optimum nanotube-metal contacts are long and strongly depend on the element, not the morphology.
- Photo-excitations are very long-lived and can be used to selectively remove oxygen impurities and heal other defects.

The End