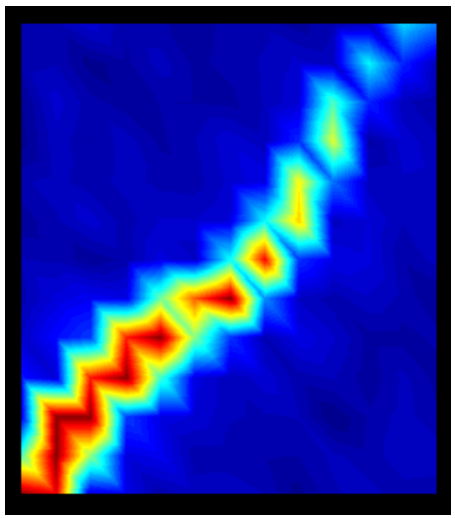
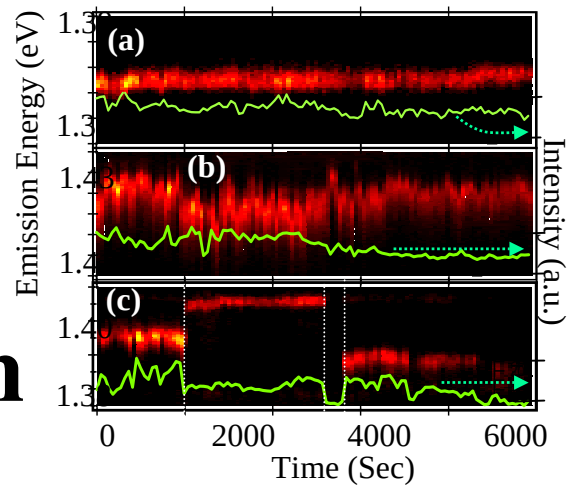
# Raman Imaging and Fluorescence Studies Of Single Isolated Carbon Nanotubes




## Stephen K. Doorn



## Chemistry Division, C-ACS
## Los Alamos National Laboratory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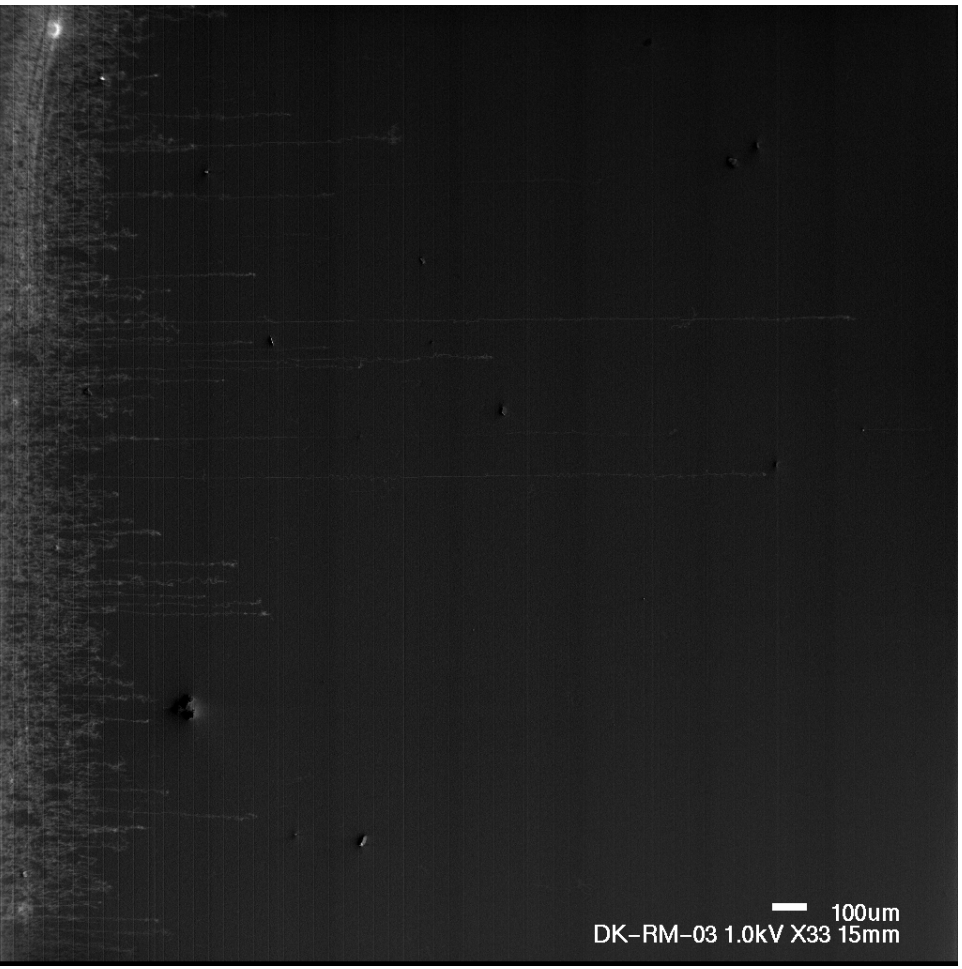
*Ideas That Change the World*

# Ultralong Nanotubes: Up to 4 cm Long.




Huang, Cai and Liu, JACS, **2003**, *125*, 5636.

Zheng *et. al.*, Nature Materials, **2004**, *3*, 673.

## Motivations

--Significant materials, electronics, and sensor applications.

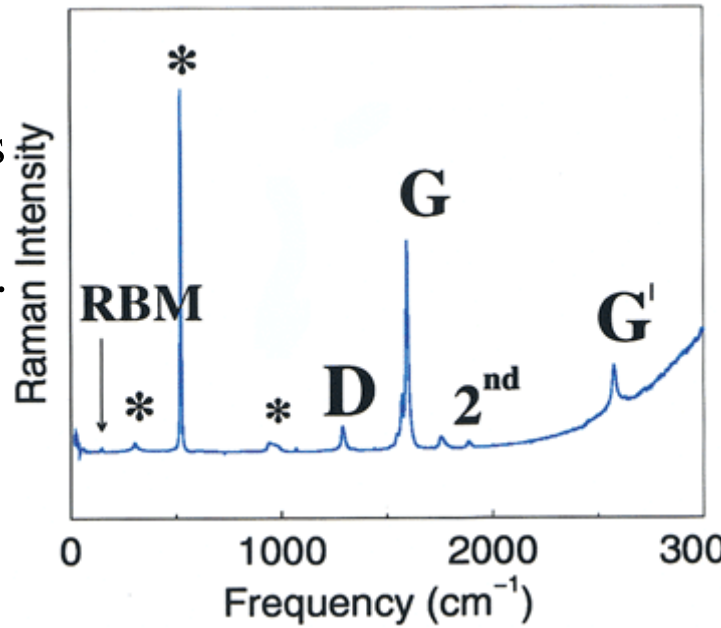
--What is defect density?

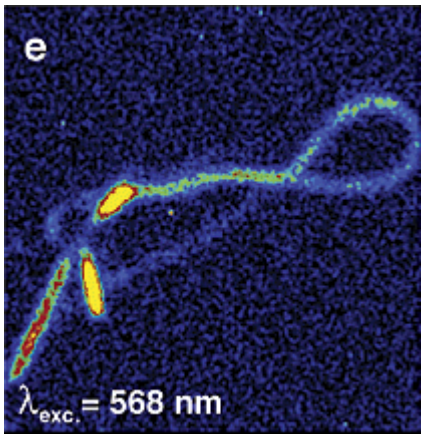
--What is structural/electronic uniformity?

--What are growth and termination mechanisms?

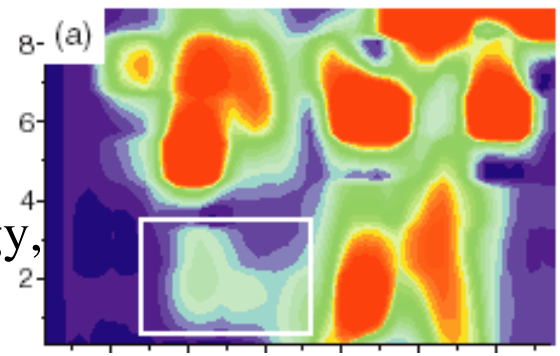
# Confocal Raman Studies of Single Nanotubes

Pioneered by Dresselhaus and coworkers.
--Carbon, **2002**, *40*, 2043.
--Accts. Chem. Res. **2002**, *35*, 1070.



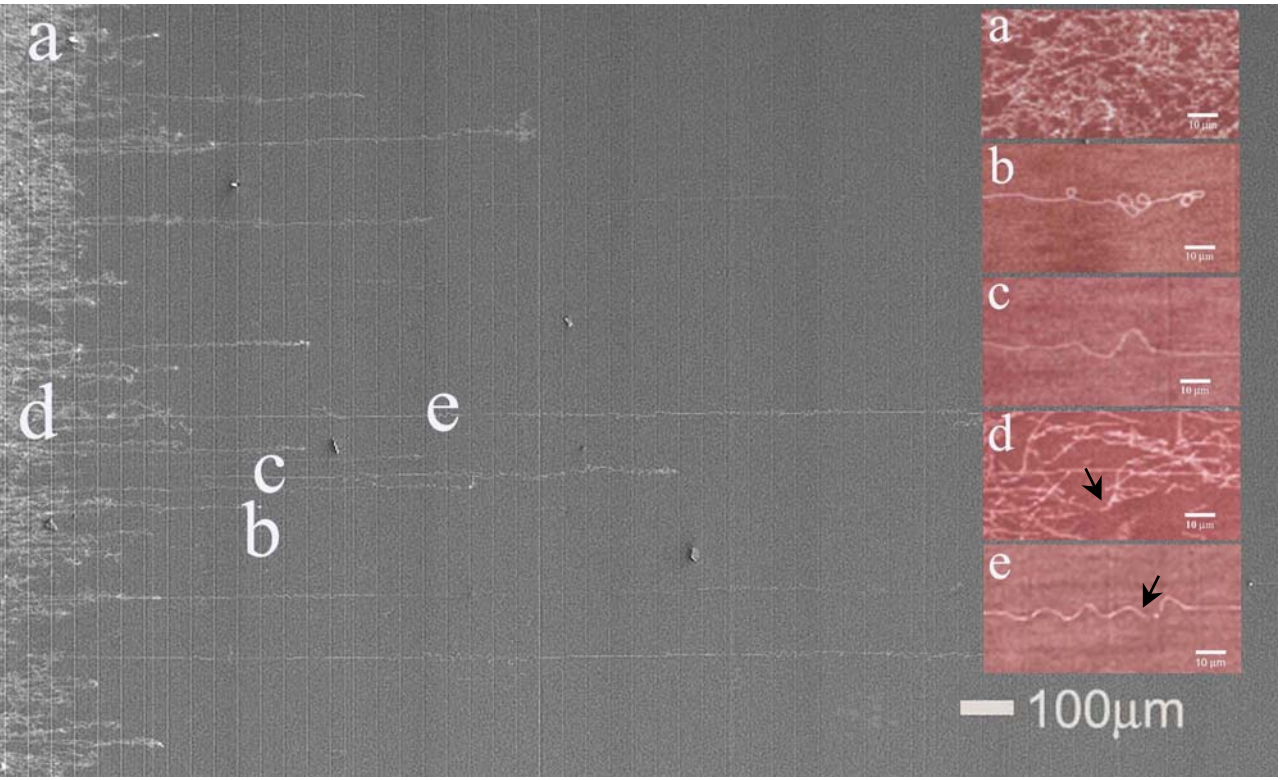




Jiang *et. al.*, J. Phys. Chem. B, **2003**, *107*, 8742.

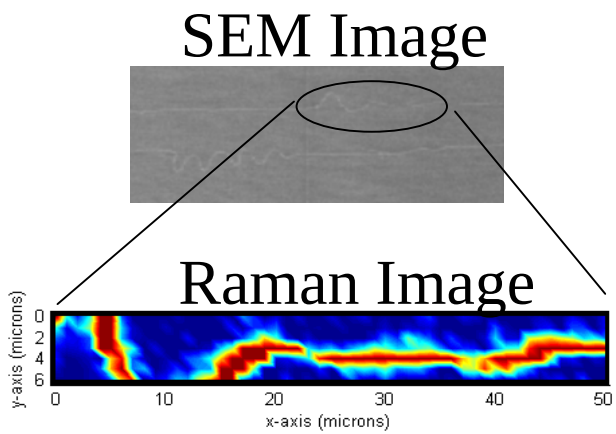



Hadjiev *et. al.*, Nanotechnology, **2004**, *15*, 562.

# Two Distinct Growth Regions

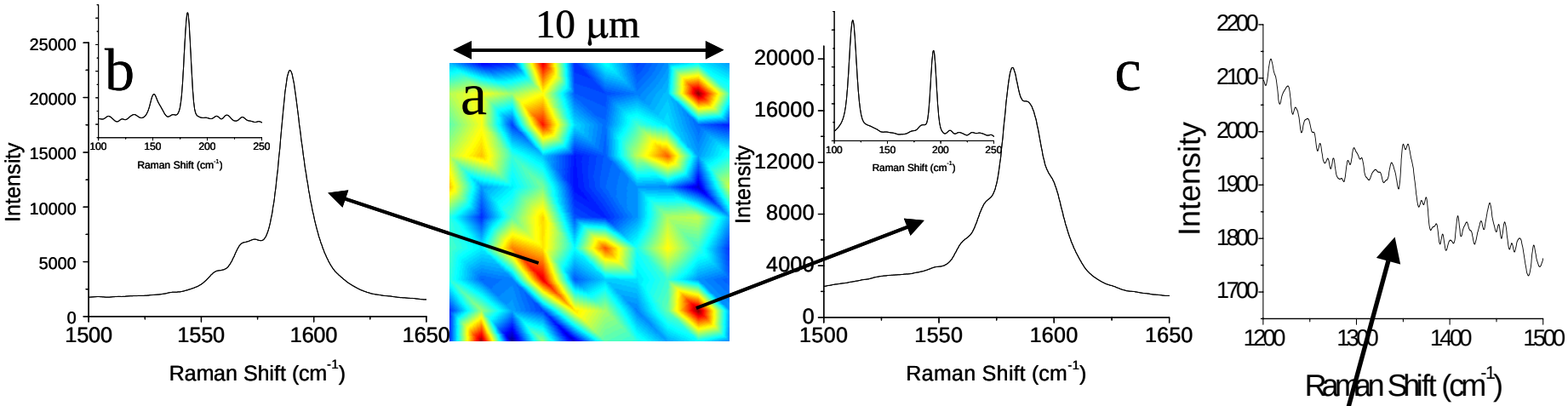



# Direct Image Correspondence

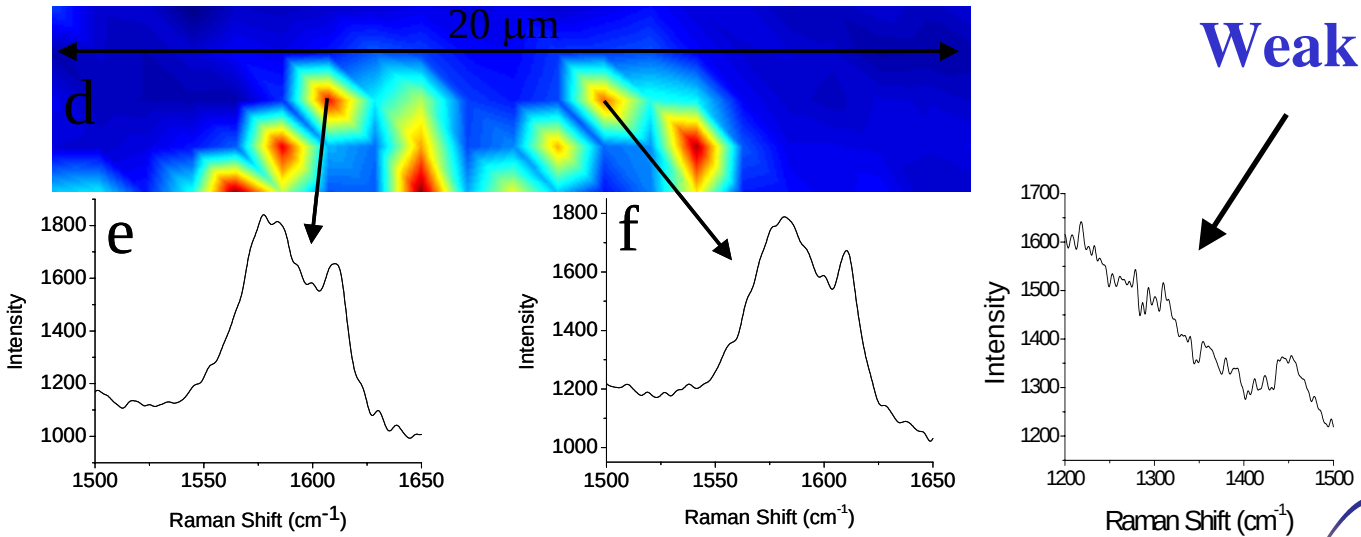



# Substrate Landmarks for Registry

# Catalyst Region: High Density, Short Tubes

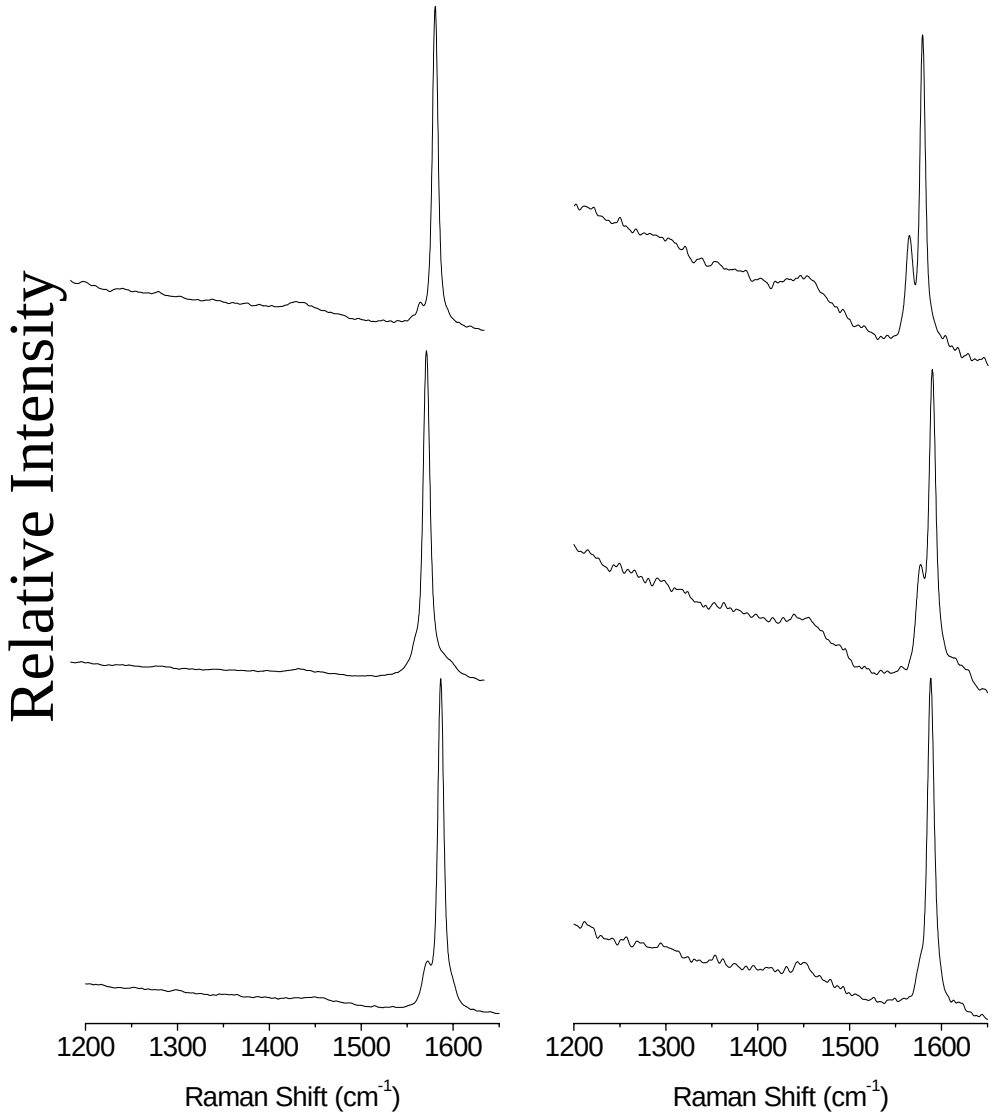



# Long Growth Region: Individual Isolated Tubes




## Weak D Bands

# G & D Band Spectra for 6 Individual Nanotubes



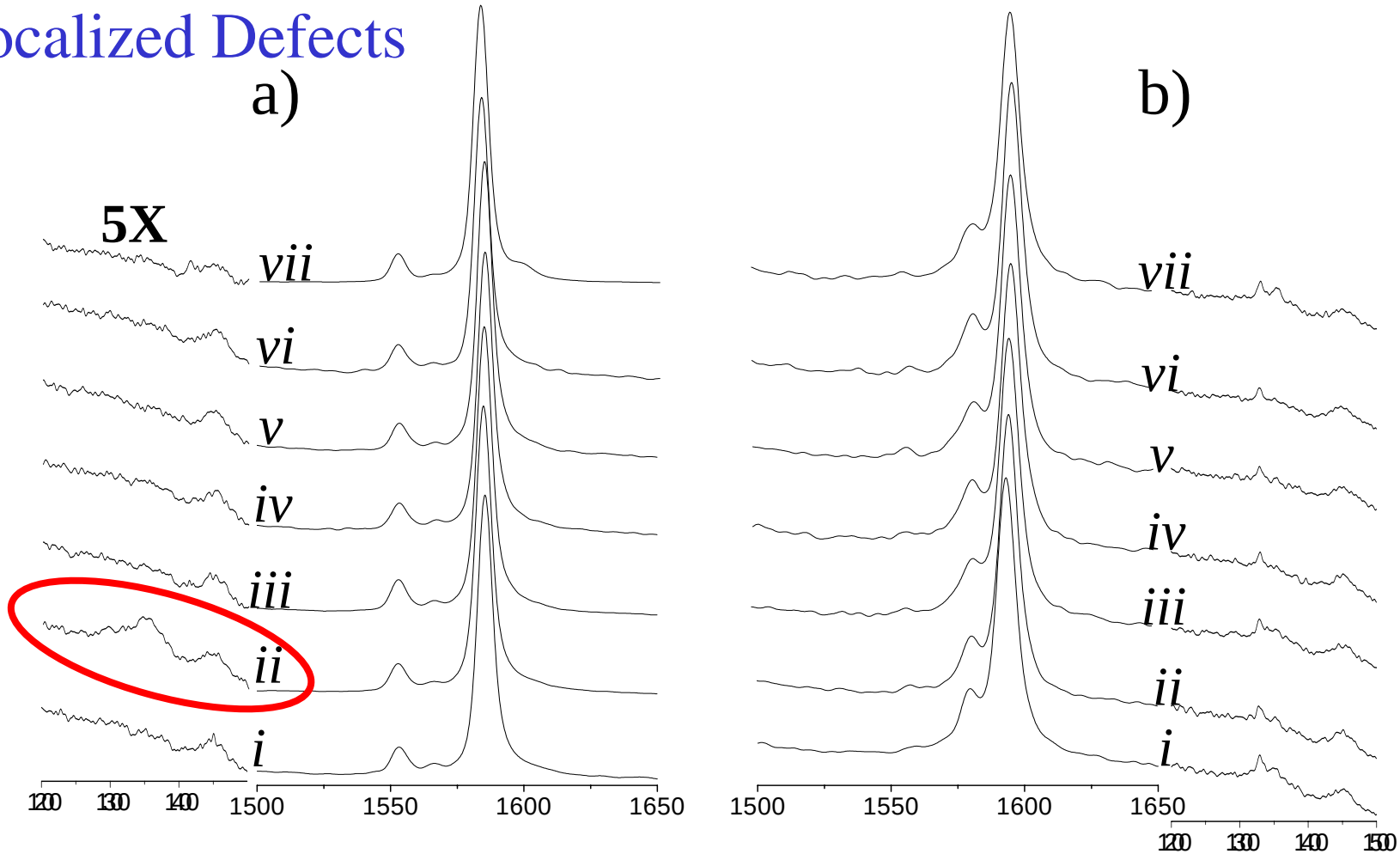

**High Quality Tubes**
**Low Defect Density**

G/D ratios of 60-500

HiPco G/D ~30
Laser Oven G/D ~190

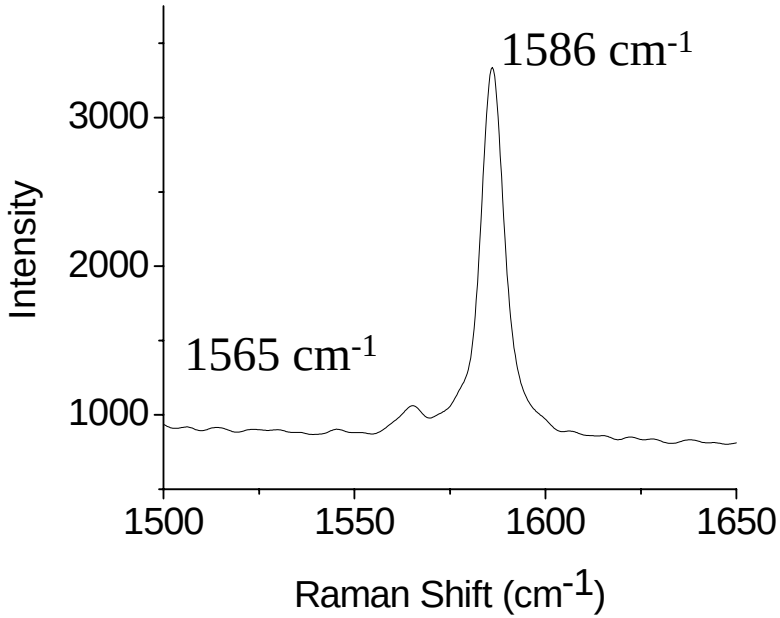
# G & D Band Spectra Probed Along 1 mm Length Of 2 Individual Nanotubes

Localized Defects

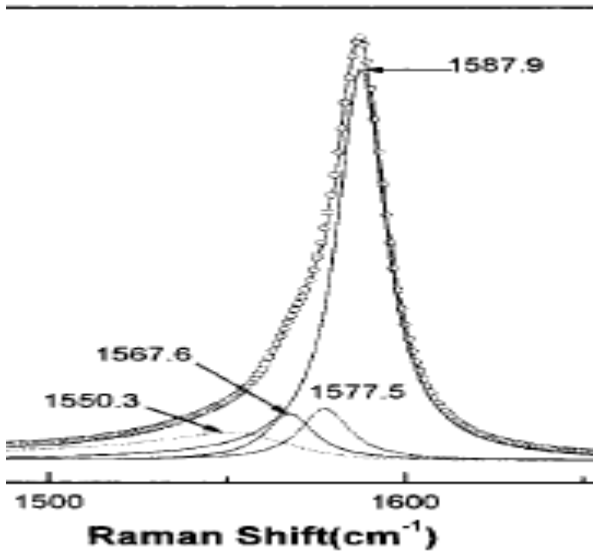


# Single or Double Walled?

Ultralong G-Band



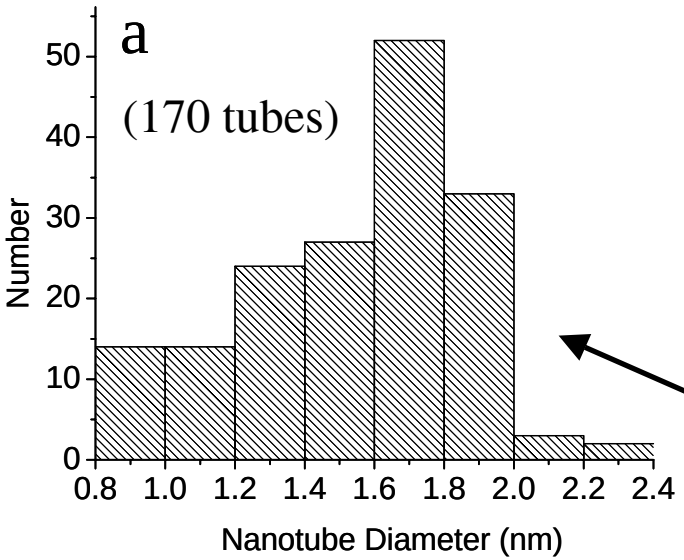

Double Walled G-Band



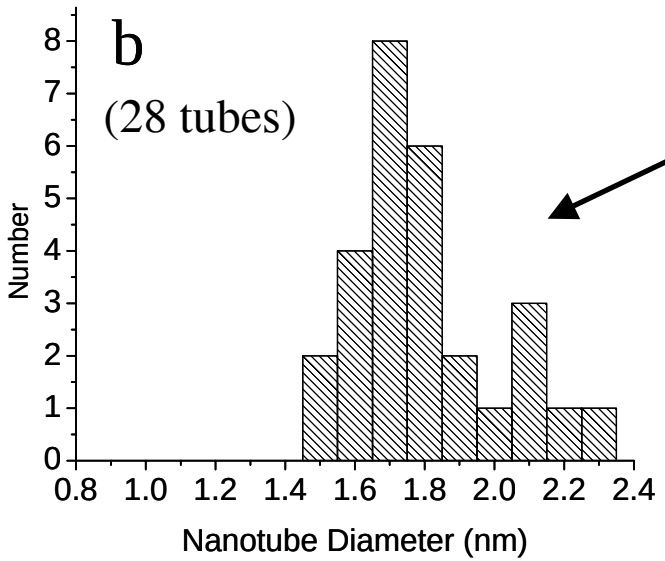

Li *et. al.*, J. Mater. Res., **2003**, *18*, 1251.

TEM needed for definitive result.

# What is Diameter Distribution?

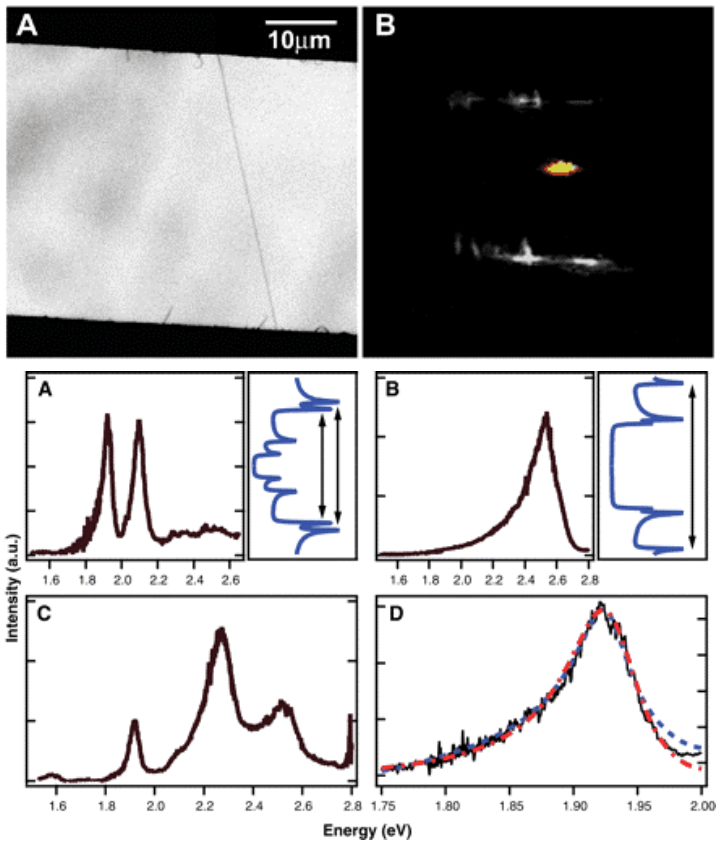



Broad Distribution in Catalyst Region:
--Large range of catalyst diameters.
(Determined by RBM: $\nu_{RBM} = 248/d$)




Narrower Distribution for Long Tubes:
--Narrow range of catalyst diameters.
(Determined by G-band separation)
$\Delta\nu = C/d^2$

# Single Nanotube Chirality Determination Needs

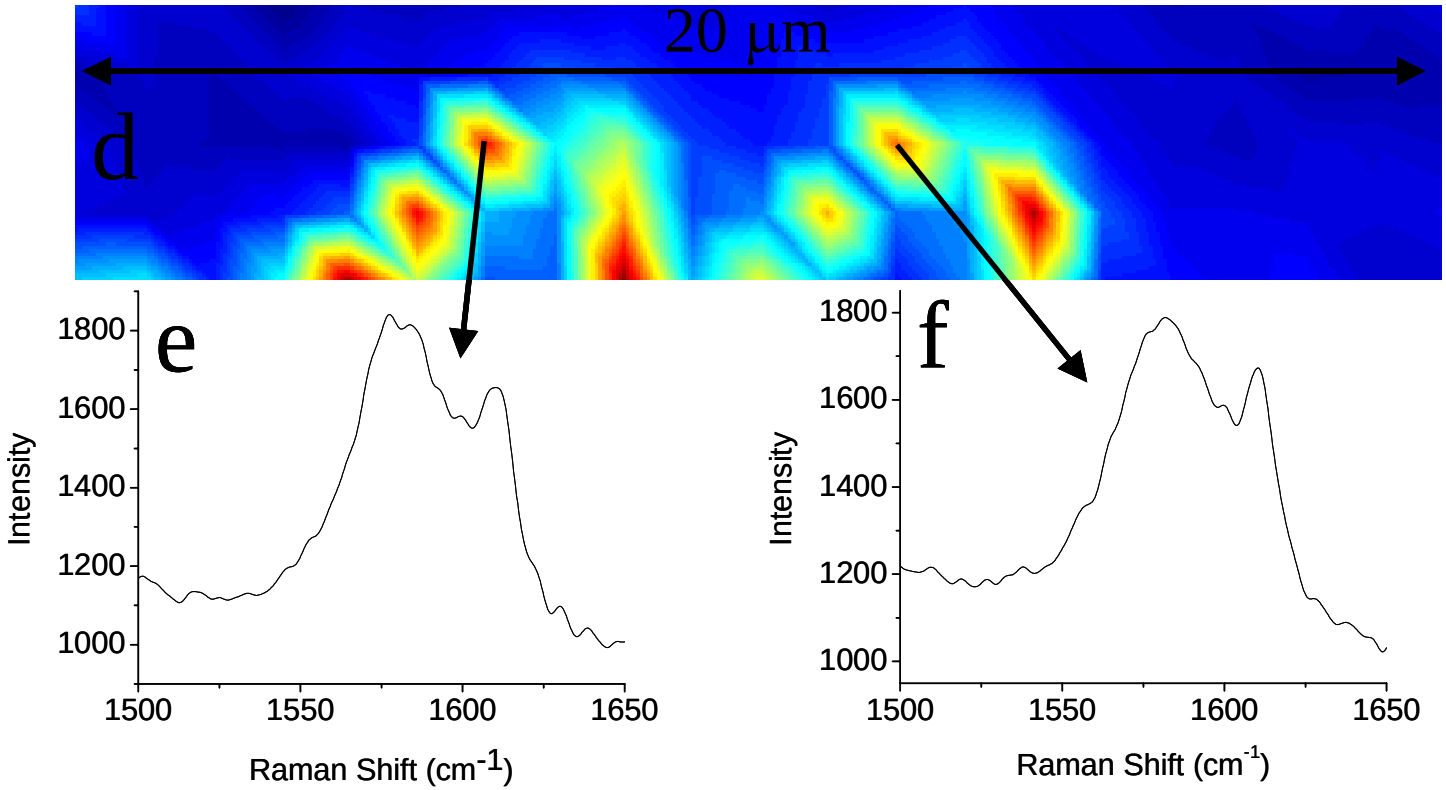



Sfeir, et. al. *Science*, **2004**, *306*, 1540.

--Diameter (RBM)
--Transition Energy
--Theoretical Models
(especially high v.H.)

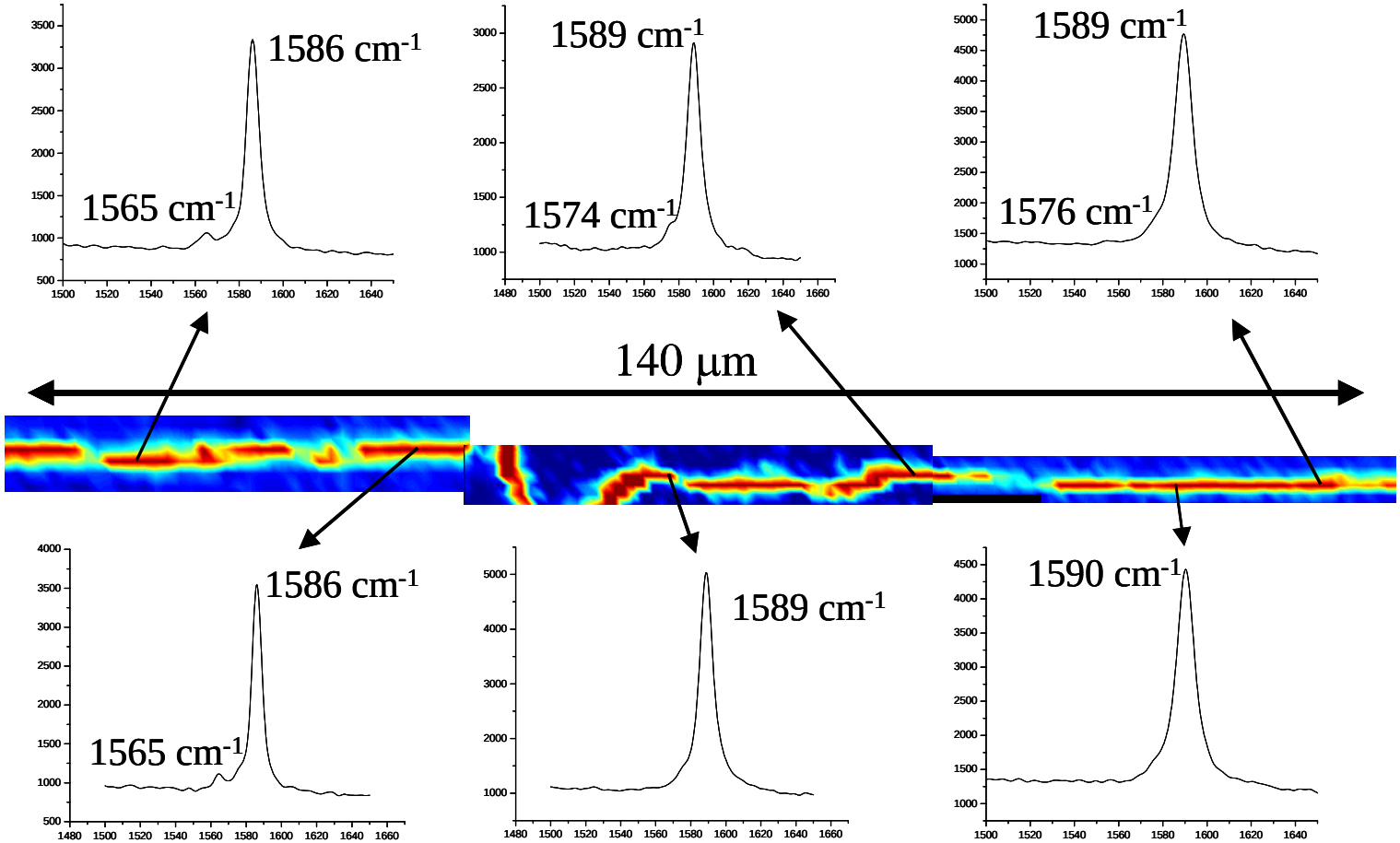
## Transition Energies

--Raman Profiles
--Direct Absorbance
--Fluorescence
--Raleigh Scattering




## Breit-Wigner-Fano Lineshapes: Metallics Present.

## Uniform, Stable Structures Possible

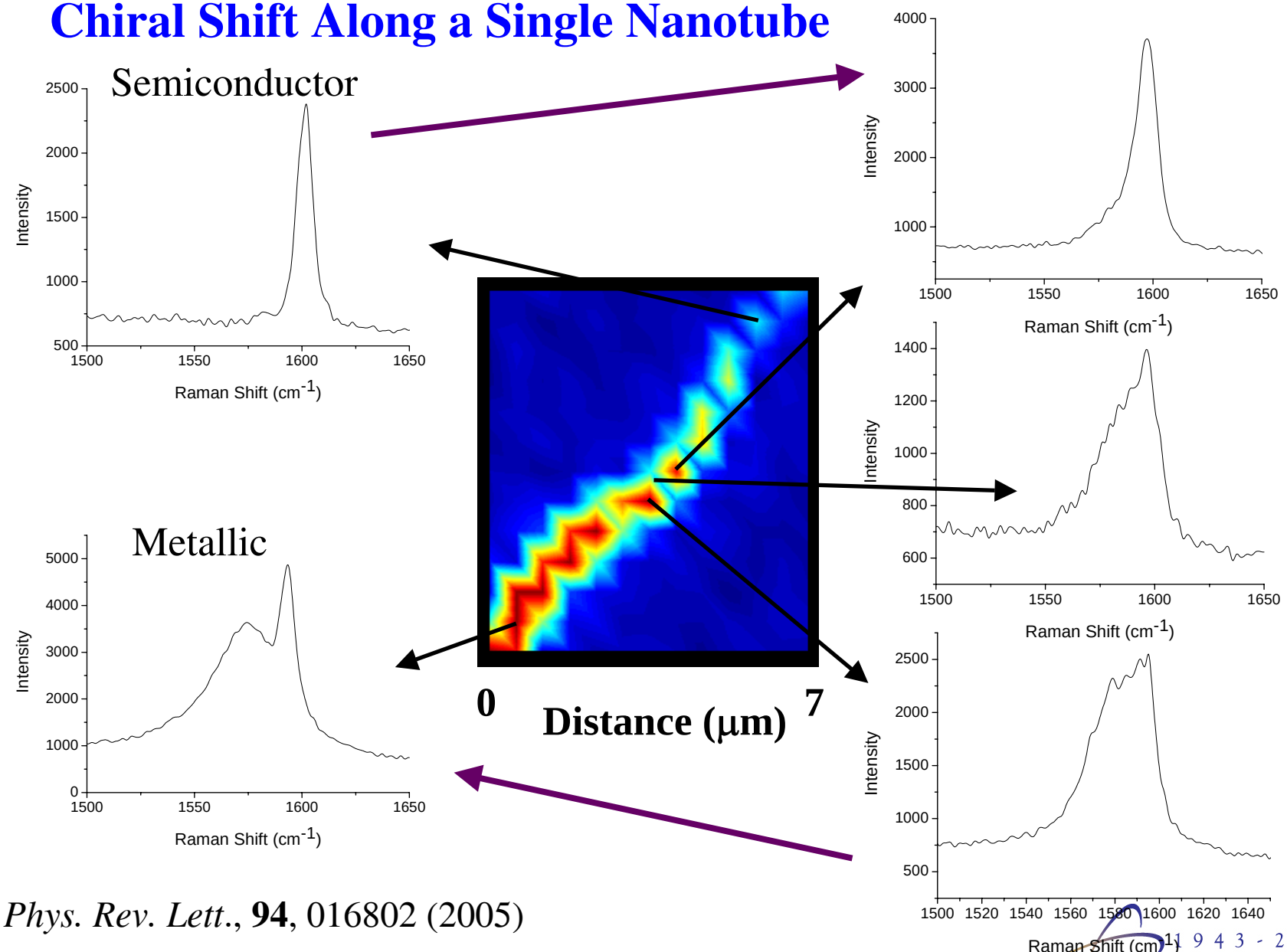



**Narrow Lorentzian G-Bands Dominate: Semiconductors**

**Frequency Shifts: Minor structural changes, substrate interactions.**

**Intensity Changes: Shift in electronic structure, substrate interaction.**

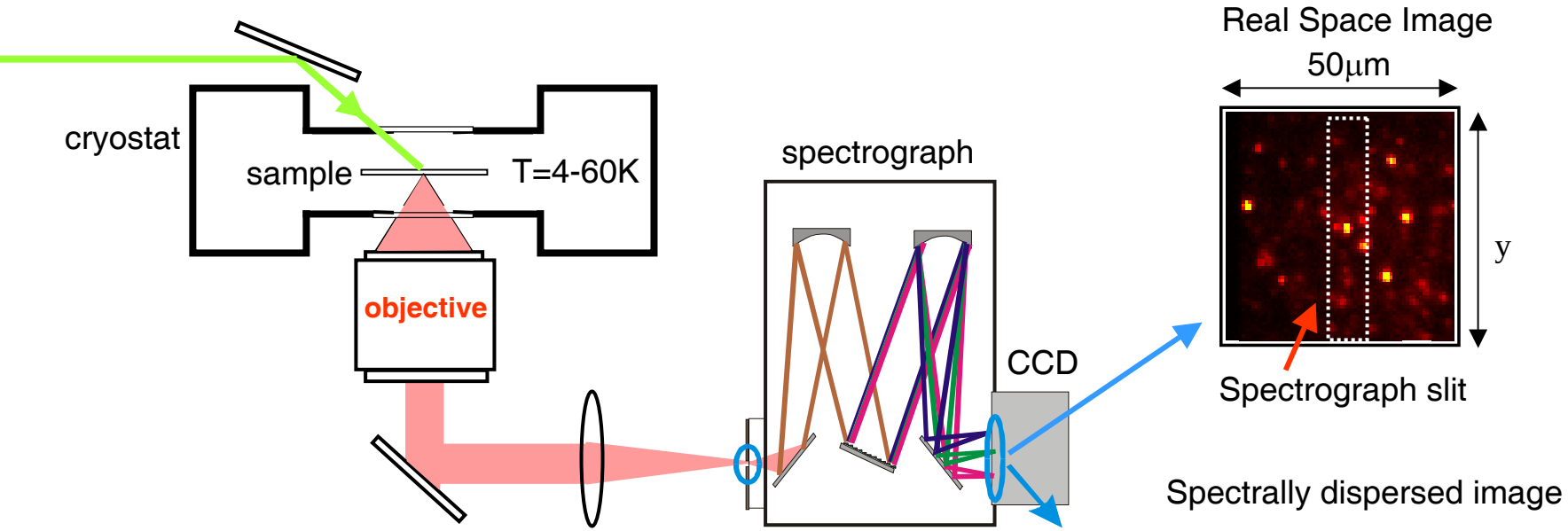
# Chiral Shift Along a Single Nanotube



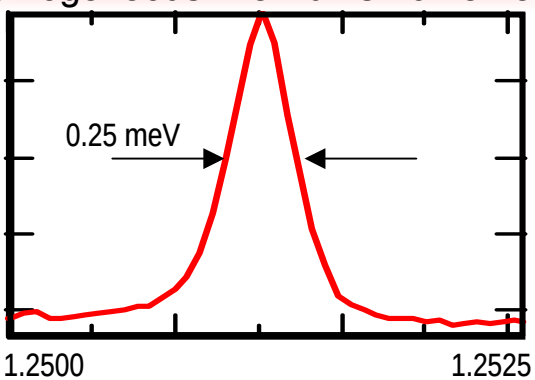

*Phys. Rev. Lett.*, **94**, 016802 (2005)

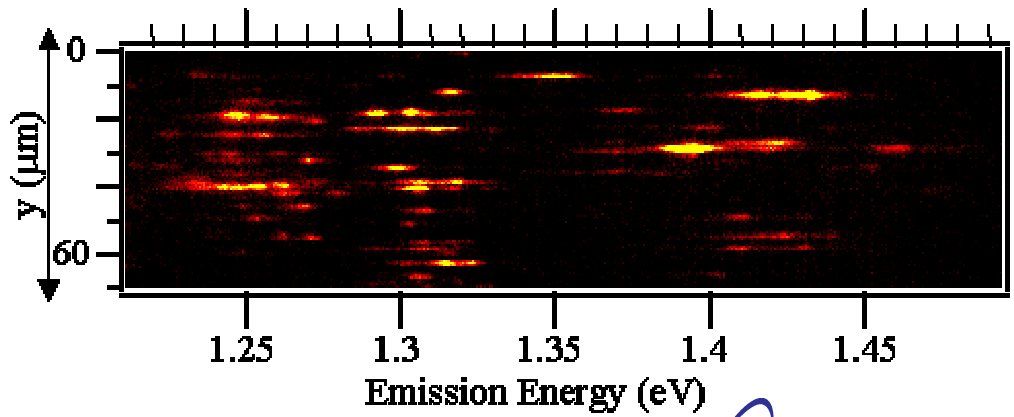
# PL and PLE Spectroscopy of Single Nanotubes at Low Temperature

*Phys. Rev. Lett.*, **93**, 027401 (2004)



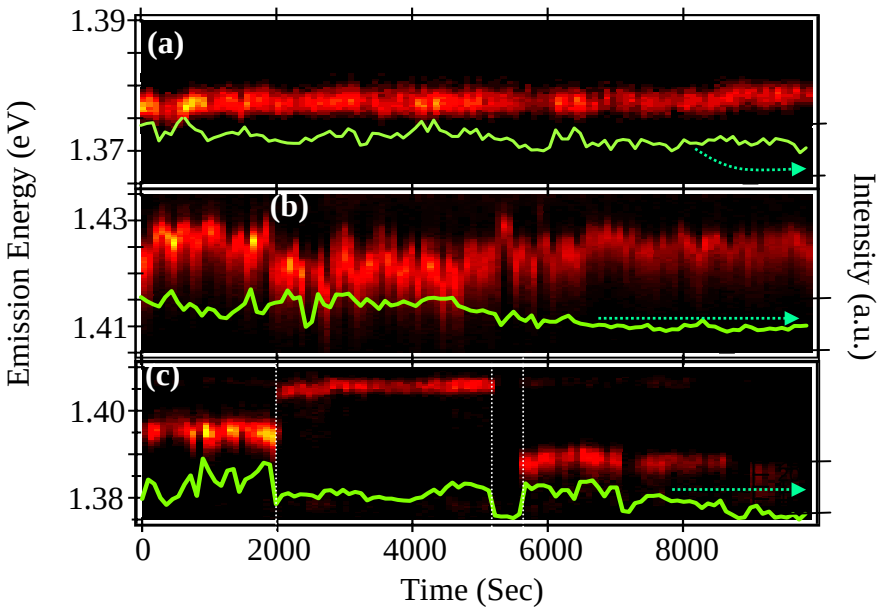

Atomic like spectral peaks with homogeneous linewidths ~0.25meV



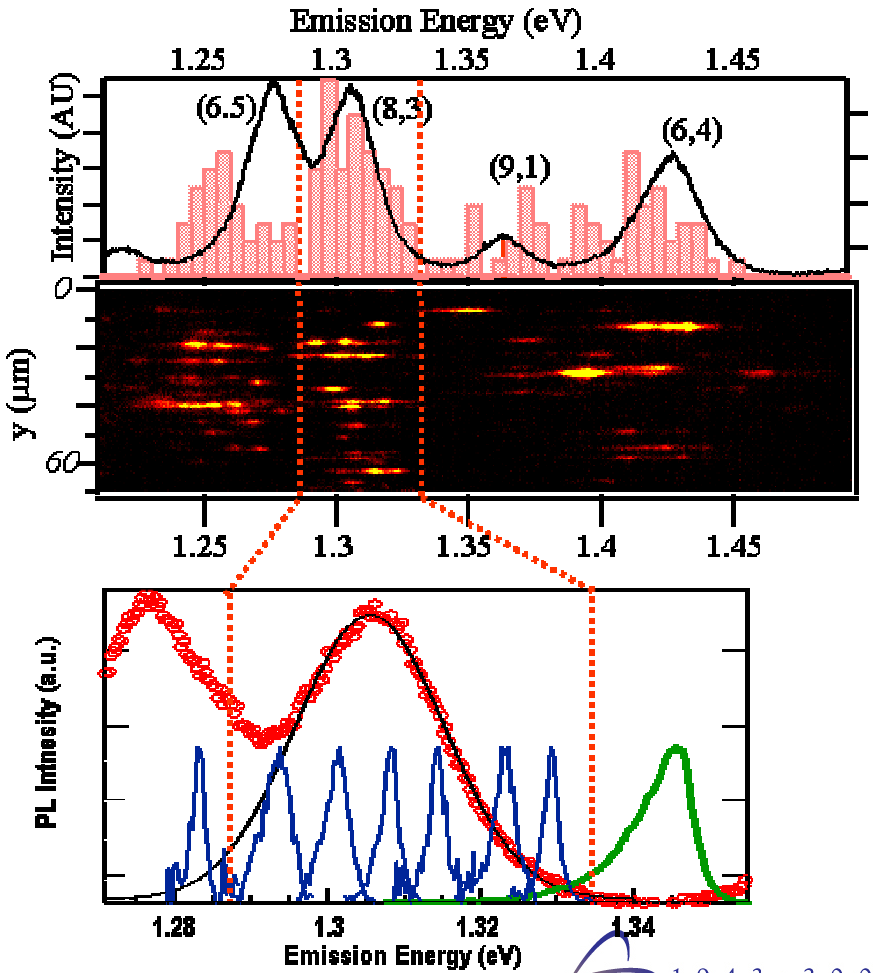



# Ensemble vs. Single Tube Measurements

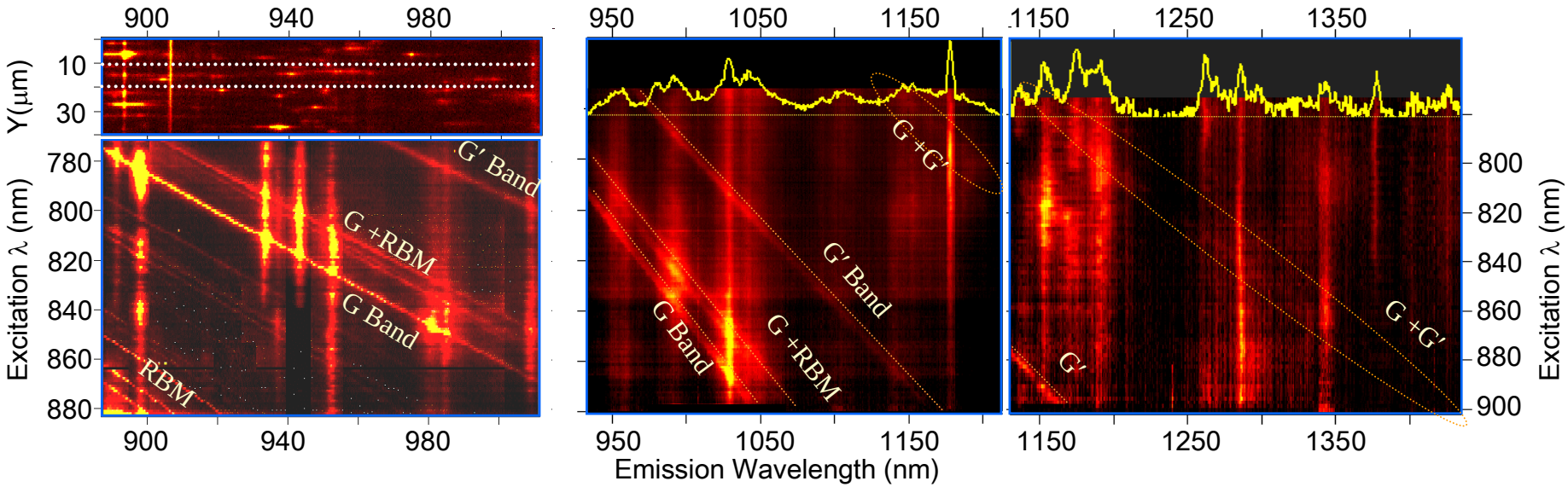
## Blinking and Spectral Diffusion



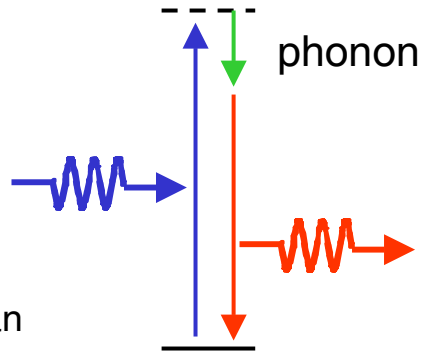

## Spectral Heterogeneities



# Low Temperature PLE Studies of Individual Carbon Nanotubes




- Vertical line segments: PLE spectra
- Diagonal lines ⇒ Peaks appears at a constant energy below the laser
  ⇒ Raman scattering process
- Both Raman and PL signals are enhanced at the intersection points
  ⇒ Phonons have strong contribution in optical absorption
- Large diameter CNTs shows the features that are now associate with Raman lines ⇒ Electronic transitions.



# Single Tube Challenges

--New Tools for Transition Energy Measurement

--Improved and Expanded Kataura Plots

--Separation of Intrinsic Behavior from Environmental Effects

--Correlated Imaging Methods
(Raman + Fluorescence, etc…)

--Couple Rapid Imaging with High Spatial Resolution

# Acknowledgements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