The Ratio Problem In Nanotube Fluorescence Spectroscopy

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What is the ratio problem ?
(previous talk & next slide)

What causes the ratio problem ?
(e-h interaction in excited state)

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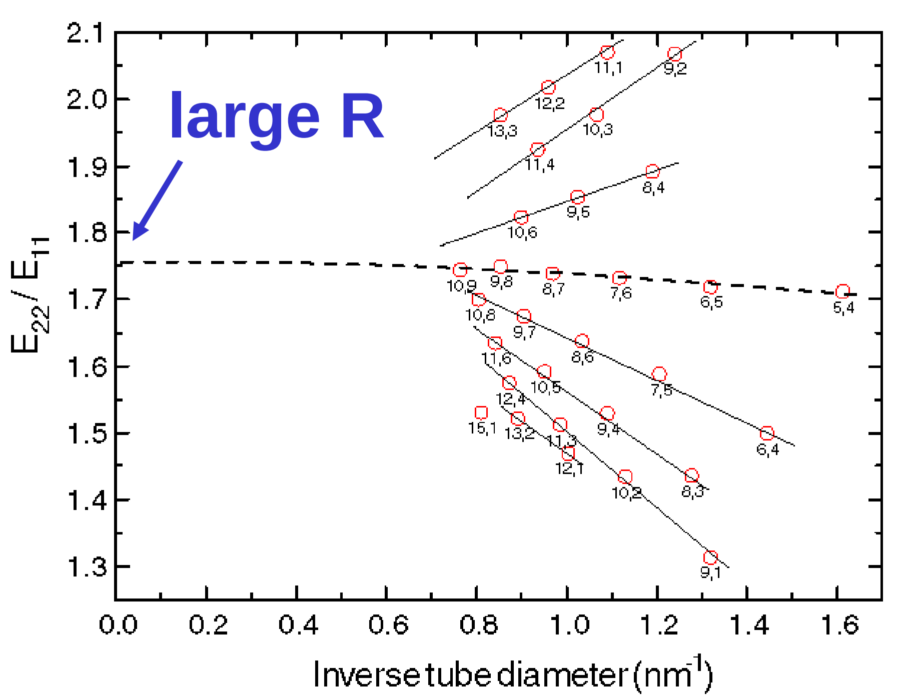
Plot of Ratios of Absorption/Emission Frequencies



One particle excitations on NT's:

- quantized subbands (m)
- conserved crystal momentum (q)
- $E_m(q)$

Interacting e-h's on NT's:

- Intraband scattering:

  only *total* crystal momentum is conserved

  $(q_e - q_h = 0)$

- Intersubband scattering:

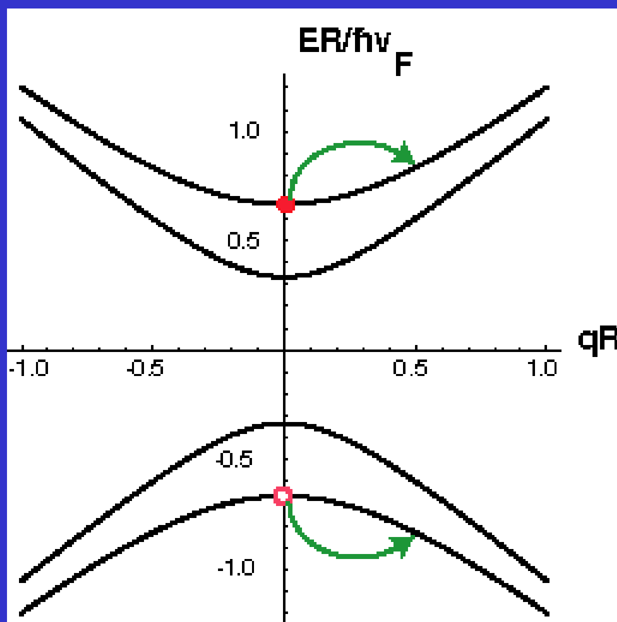
  m is not conserved & higher subband

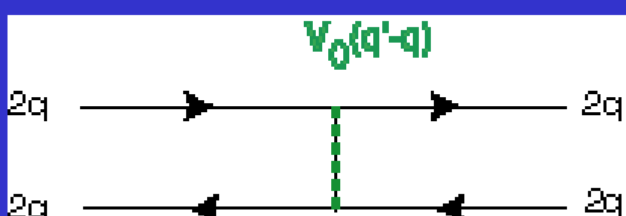
  e-h's *resonate*

- Mix e-h ❿ 2e-2h configurations

  This is Ratio Problem

Intraband scattering❶





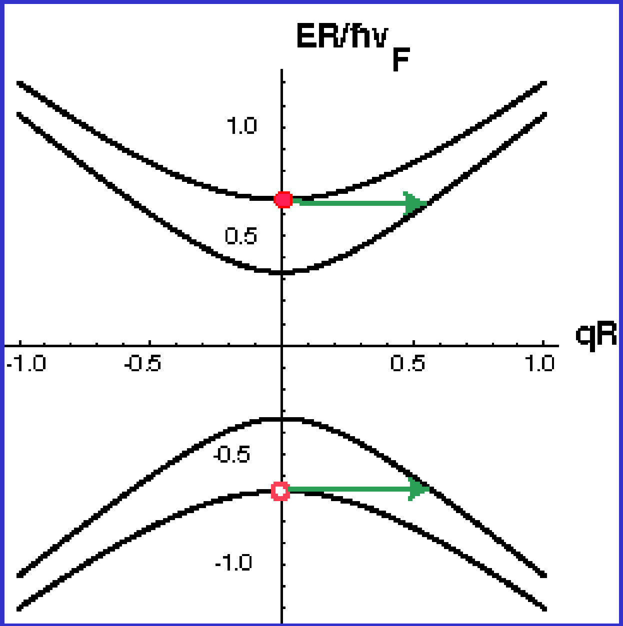
binds e-h pair

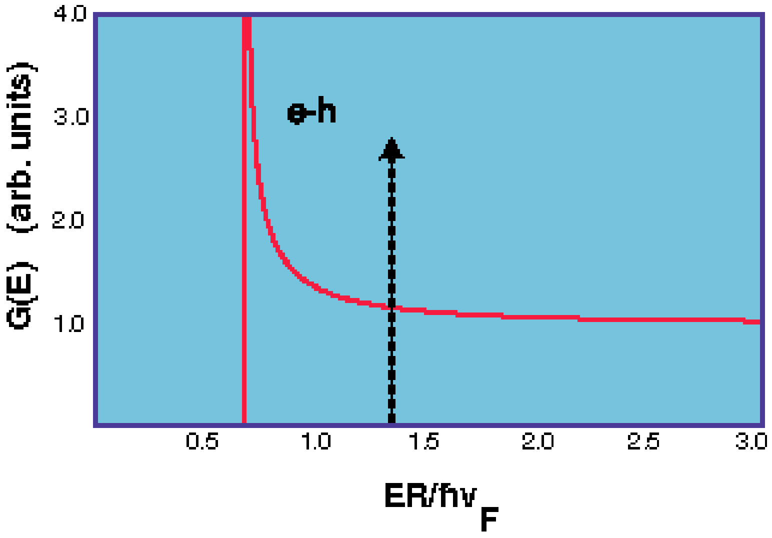
$$\psi(z) = \frac{1}{\sqrt{\xi}} \exp(-\frac{|z|}{\xi})$$

with scaling rules

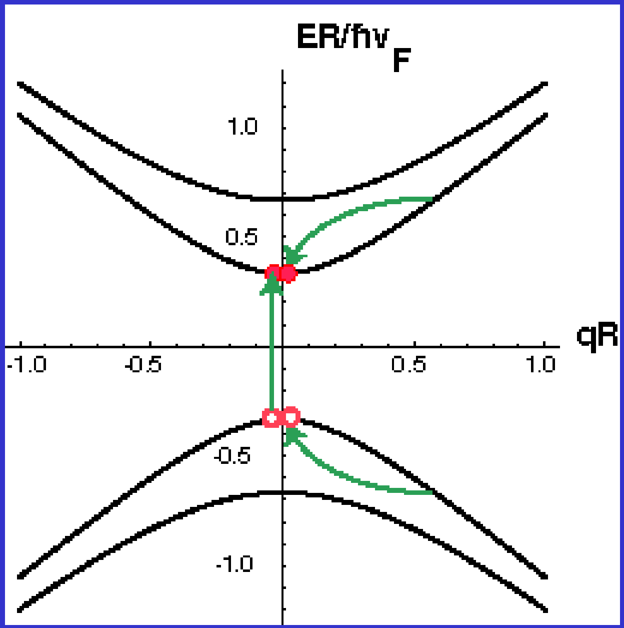
$$\xi_n \propto \frac{1}{n} \qquad \Delta E \propto n$$

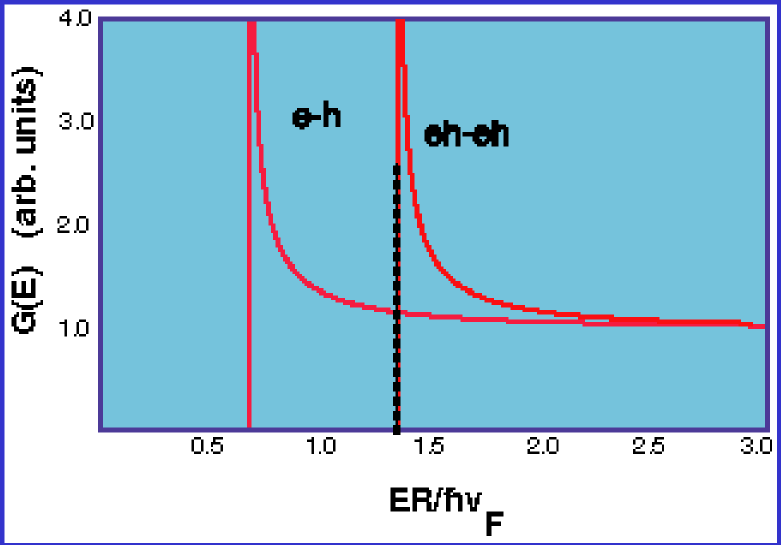
Interband Scattering
Intrinsic Width (Resonant Exciton)



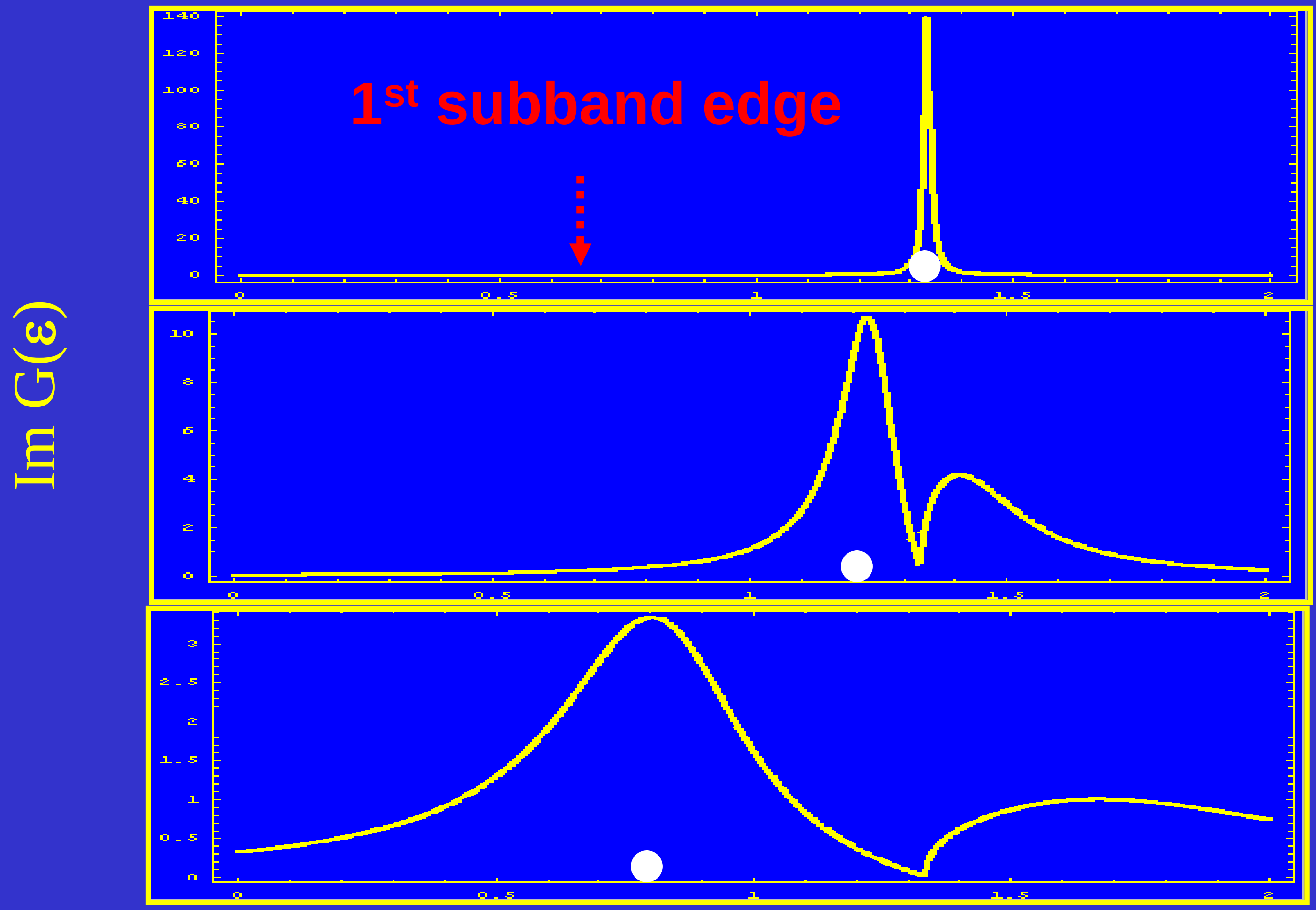


Hybridization of e-h and 2e-2h excitations





Exciton Lineshape



$\varepsilon = ER/\hbar v_F$

Exciton Lineshape Expressed in Natural Units

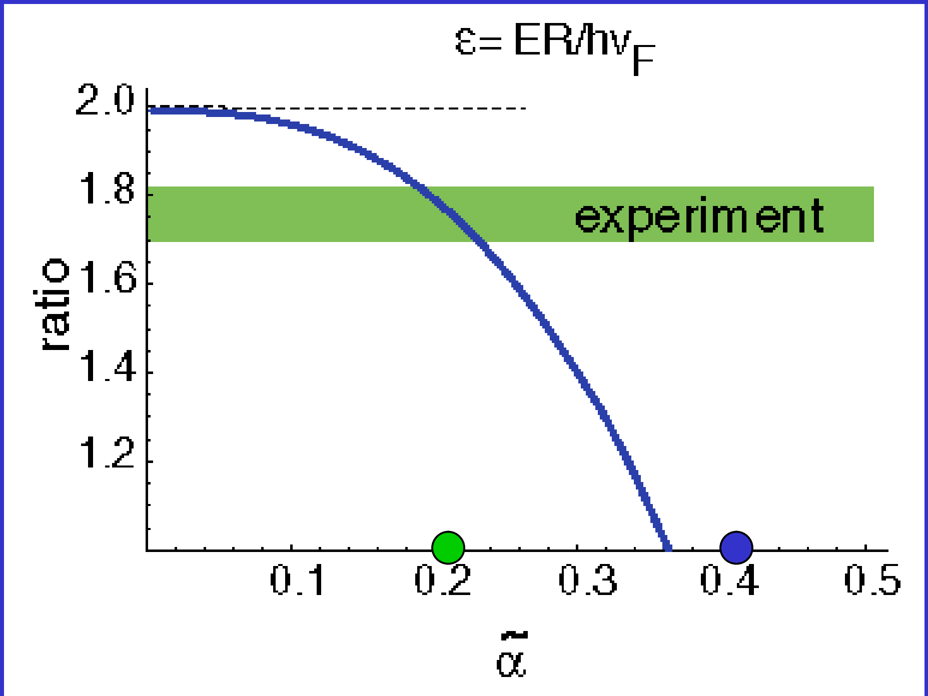
$$\varepsilon = \frac{ER}{\hbar\nu} \qquad \tilde{\alpha} = \frac{e^2}{2\pi\kappa\hbar\nu}$$

$$G(\varepsilon) = \frac{1}{\varepsilon - \varepsilon_0 + iA\tilde{\alpha}^2 + \frac{B^2\tilde{\alpha}^4}{\sqrt{\varepsilon_0^2 - \varepsilon^2}}}$$

$$\Delta\varepsilon = -\frac{3^{1/3}B^{4/3}\tilde{\alpha}^{8/3}}{2} \qquad \gamma = A\tilde{\alpha}^2$$

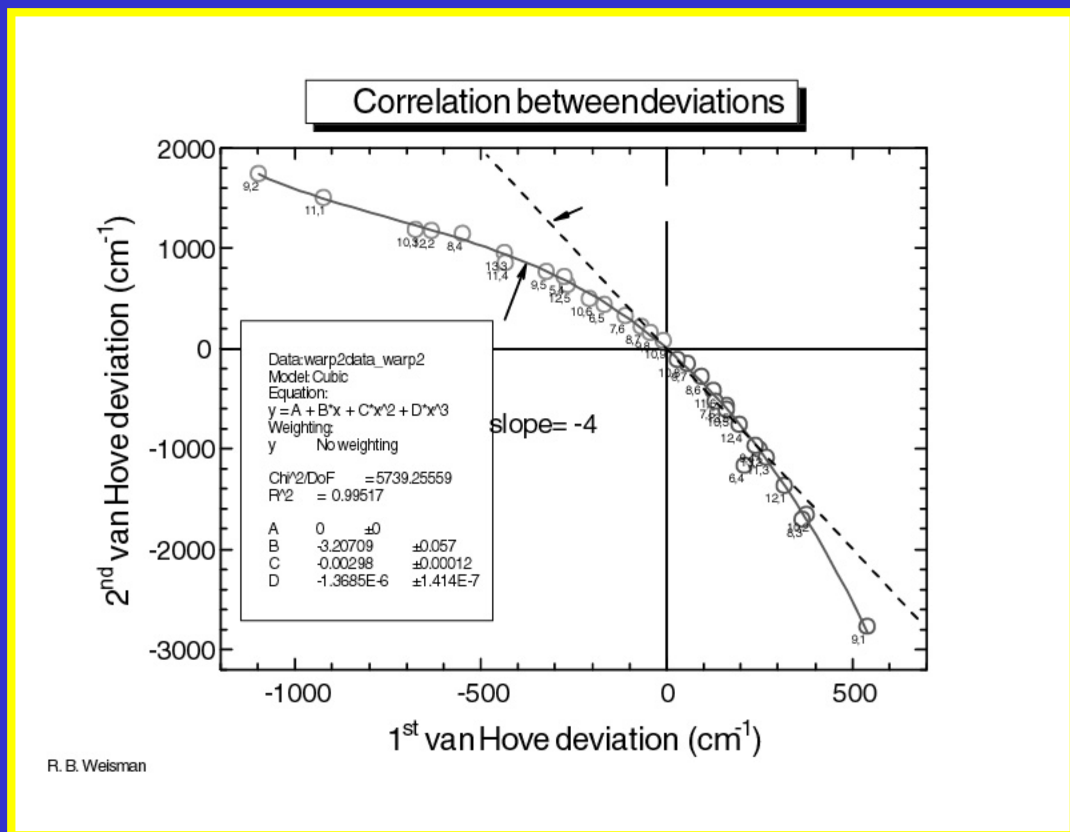
Line shifts and broadens

Nanotube Fluorescence Spectroscopy

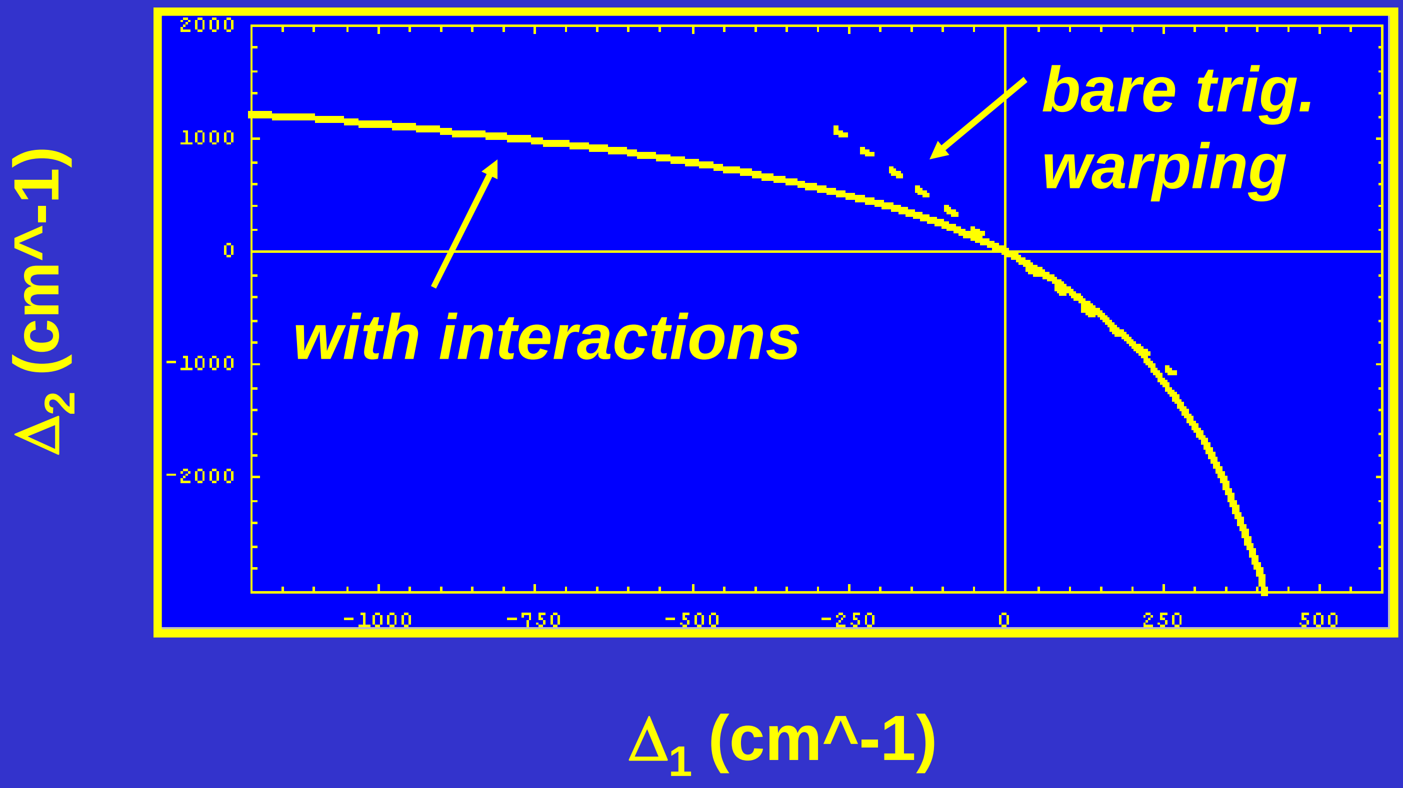


Correlations of `Mod 3` Gap Deviations

(from Structure Assigned Optical Specta of Single Wall Carbon Nanotubes,
S. Bachilo et al., Science <u>298</u>, 2361 (2002))



Nonlinear Scaling of 1st and 2nd Subband Deviations Due to Coulomb & Trigonal Warping



Summary

- FS reveals electronic gap structure outside the conventional band model.
- The "ratio problem"

  Gap Ratio < 2 (asymptote for large diameter tubes)

  Hybridize e-h and 2e-2h excitations

  1D + degeneracy from tube wrapping.

  Long Range Interaction

- "Mod 3" gap deviations

  They are very large… with ± asymmetry

  Curvature, Trig. Warping + Coul. Anisotropy (distinguished by scaling with R, n)

  Nonlinear Scaling in Data gives Coul. Anis. Trig. Warping