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)
Plot of Ratios of Absorption/Emission Frequencies

![Graph showing plot of ratios of absorption/emission frequencies with inverse tube diameter (nm⁻¹) on the x-axis and E₂₂/E₁₁ on the y-axis. The graph indicates a trend for large R.](image-url)
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 \textit{total} crystal momentum is conserved
    \( (q_e - q_h = 0) \)
  • Intersubband scattering:
    \( m \) is not conserved & higher subband
    e-h’s \textit{resonate}
  • Mix e-h \( \otimes \) 2e-2h configurations

\textit{This is Ratio Problem}
Intraband scattering

binds e-h pair

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

with scaling rules

$$\xi n \propto \frac{1}{n} \quad \Delta E \propto n$$
Interband Scattering

Intrinsic Width (Resonant Exciton)
Hybridization of e-h and 2e-2h excitations
Exciton Lineshape

\[ \varepsilon = \frac{ER}{\hbar v_F} \]
Exciton Lineshape Expressed in Natural Units

\[ \varepsilon = \frac{E_R}{\hbar \nu} \quad \tilde{\alpha} = \frac{e^2}{2\pi \kappa \hbar \nu} \]

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

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

Line shifts and broadens
Nanotube Fluorescence Spectroscopy
Correlations of `Mod 3` Gap Deviations

(from Structure Assigned Optical Spectra of Single Wall Carbon Nanotubes,
S. Bachilo et al., Science 298, 2361 (2002))
Nonlinear Scaling of 1\textsuperscript{st} and 2\textsuperscript{nd} Subband Deviations Due to Coulomb & Trigonal Warping

\[ \Delta_1 (\text{cm}^{-1}) \]

\[ \Delta_2 (\text{cm}^{-1}) \]

- \textit{bare trig. warping}
- \textit{with interactions}
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.

• “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