Raman intensities of the first optical transition in carbon nanotubes

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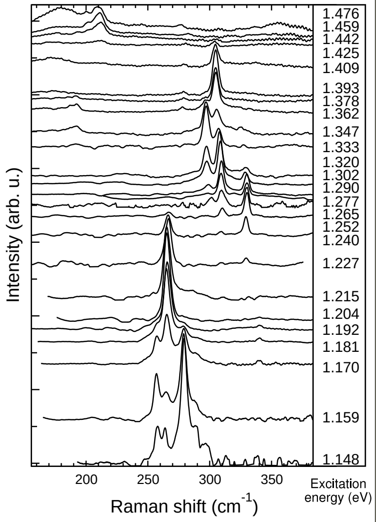
Motivation

- In resonant Raman spectroscopy of the E_{22} transitions a strong intensity difference between nanotubes with $\nu = (n-m) \bmod 3$=-1 and ν=+1 has been observed. [2,3]
- PLE experiments show weak intensities for near zig-zag tubes of ν=+1 families. [4]
- Yet PLE and Raman intensities are often used to probe the relative abundance of different (n,m). [4-6]
- Can we determine the (n,m) distribution in a nanotube sample from E_{11}^S Raman intensities?

RBM resonant with E_{11}^S

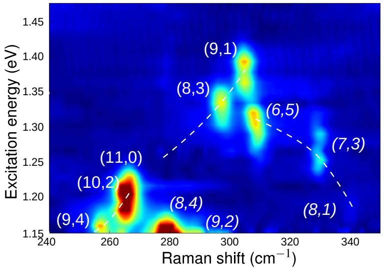


- Tubes were separated in solution using sodium dodecyl sulfate (SDS). [7]
- Data are normalized to integration time, laser power, ω_s^4, phonon energy and Bose-Einstein occupation number.

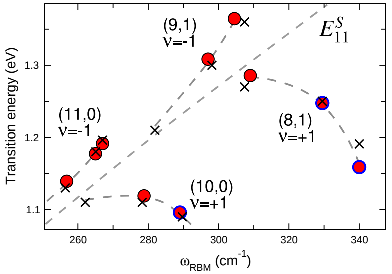
$$I(E_l) \propto \left| \frac{\mathcal{M}_{e-r}\mathcal{M}_{e-ph}\mathcal{M}_{e-r}}{(E_l - E_{ii} - i\gamma/2)(E_l - \hbar\omega_{\mathrm{RBM}} - E_{ii} - i\gamma/2)} \right|^2$$

E_{11}^S transition energies

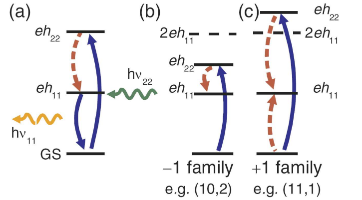
- Contour plot of the Raman spectra.
- (n,m)-assignment based on an assignment of the second optical transitions [2] and a theoretical Katauraplot [1].



- Experimental Kataura plot of the E_{11} optical transitions
- circles: RRS E_{11}, crosses: experimental and empirical data from Ref. [4] and [8]
- Three extra tubes compared to PLE experiments.



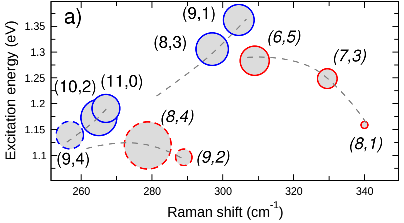
optical absorption strength



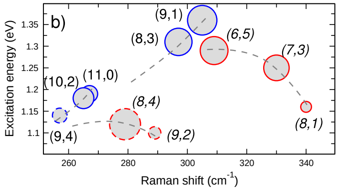
- Figure taken from Ref. [9]
- Absorption into the second subband can be suppressed by exciton resonance.
- Tubes with ν=+1 and small chiral angle, as (8,1), (7,3), (10,0) and (9,2), are not seen in luminescence experiments.

electron-phonon coupling

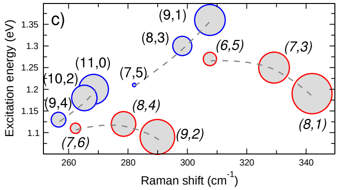
exp. Raman intensities E_{11}^S



exp. normaliezed to Gauss dist.



theo. from Ref. [10]

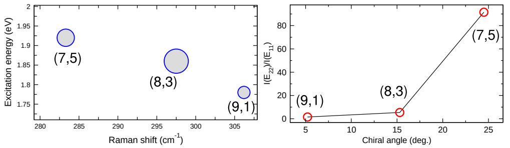


a)
- Intensities of ν=+1 and ν=-1 tubes are similar
- In contrast to E_{22}^S transitions where ν=-1 intensities are approximately 5 times larger than the ν=+1 intensities.
- (7,5) tube is not observed.

b) Intensities normalized to a Gaussian distribution with mean diameter 10 Å and width 3 Å.

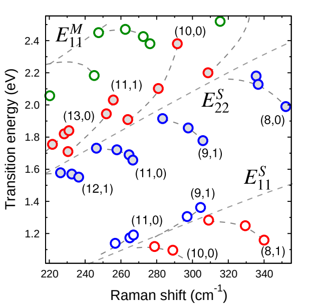
c) Theoretical Raman intensities from Ref. [10]

exp. Raman intensities E_{22}^S



- Ratio between Raman intensities at E_{11}^S and E_{22}^S resonances depends on the chiral angle.
- Agreement with *ab-initio* calculations in Ref. [11]:
  (10,0) ν=+1 ⇒ $|\mathcal{M}_1^{e-ph}|^2/|\mathcal{M}_2^{e-ph}|^2 = 3$
  (8,4) ν=+1 ⇒ $|\mathcal{M}_1^{e-ph}|^2/|\mathcal{M}_2^{e-ph}|^2 = 11$
  we expect an opposite behaviour for the (7,5) and (9,1) tubes $|\mathcal{M}_2^{e-ph}|^2 > |\mathcal{M}_1^{e-ph}|^2$.

exp. Kataura plot



Conclusion

- The (8,1), (7,3) and (9,2) tubes, which are not seen in PLE are observed in resonance Raman.
- Intensity difference between ν=±1 families is much smaller than for the E_{22}^S transitions and possibly reversed, in agreement with predictions from Ref. [9,11]
- Ratio between the electron-phonon coupling of the first and second transition depends on the chiral angle.
- Raman intensities need to be normalized by the electron-phonon coupling strength to obtain abundences of (n,m).

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