Phonon softening in metallic NTs

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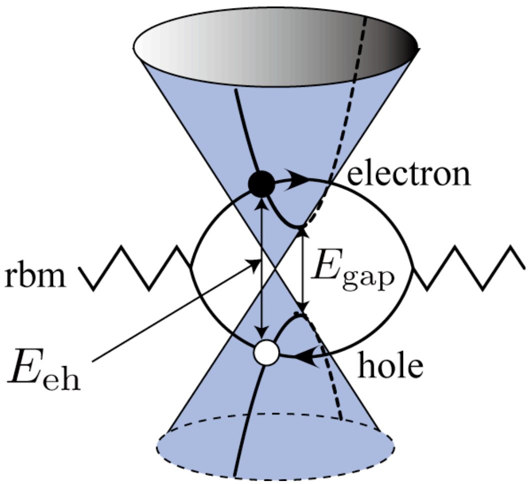
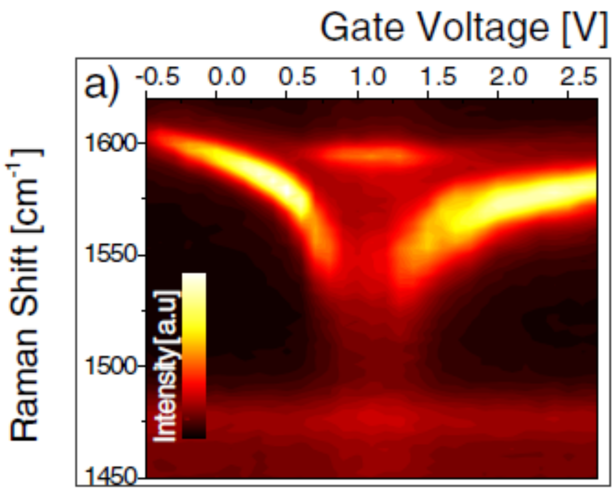
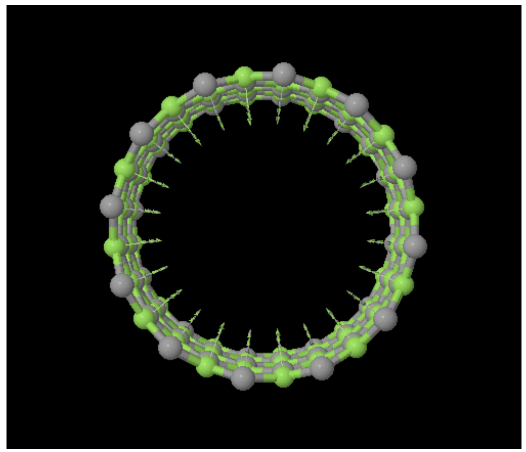
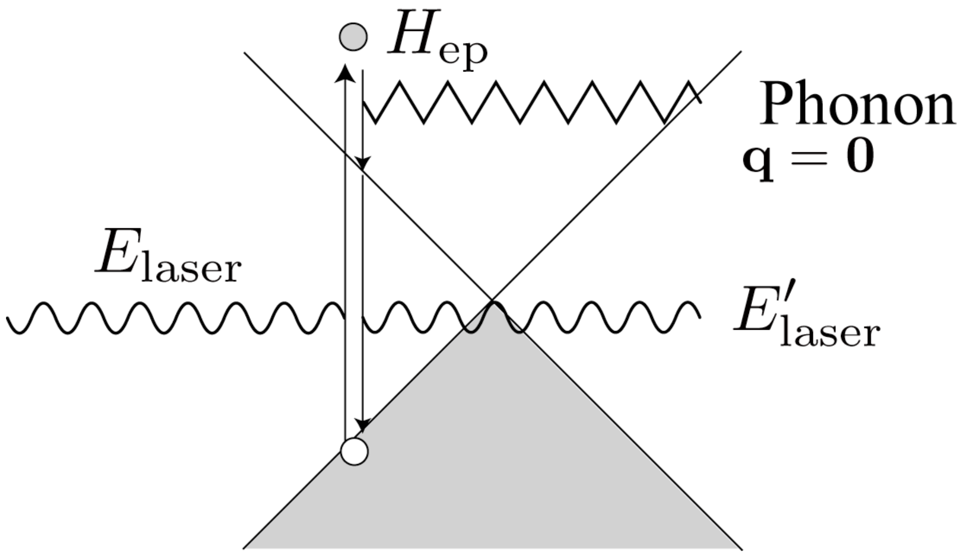






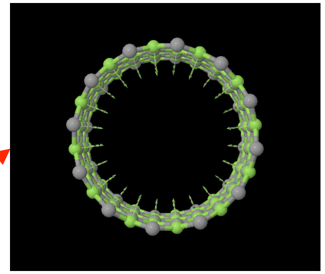
Raman spectroscopy



$$\omega_{\mathbf{q}=\mathbf{0}} = E_{\rm photon} - E'_{\rm photon}$$

A peak in Raman shift gives information on phonon energy

1meV=8/cm



Radial breathing mode (**RBM**)

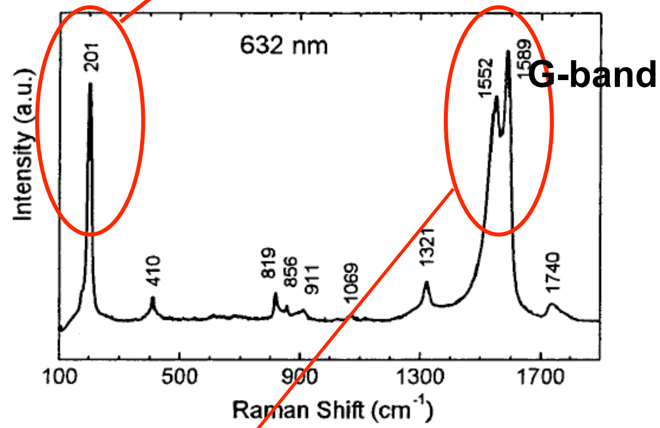
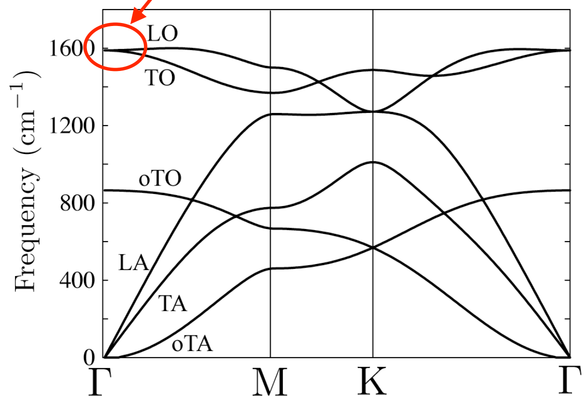


Figure 2. Raman spectrum of a single SWNT bundle obtained by using backscattering confocal Raman microscopy with 20 kW/cm^2 632 nm excitation.

Yu *et al*., J. Phys. Chem. B105 (2001)

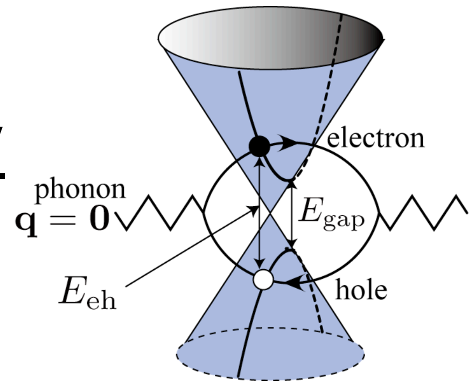


Phonon dispersion relation of graphene

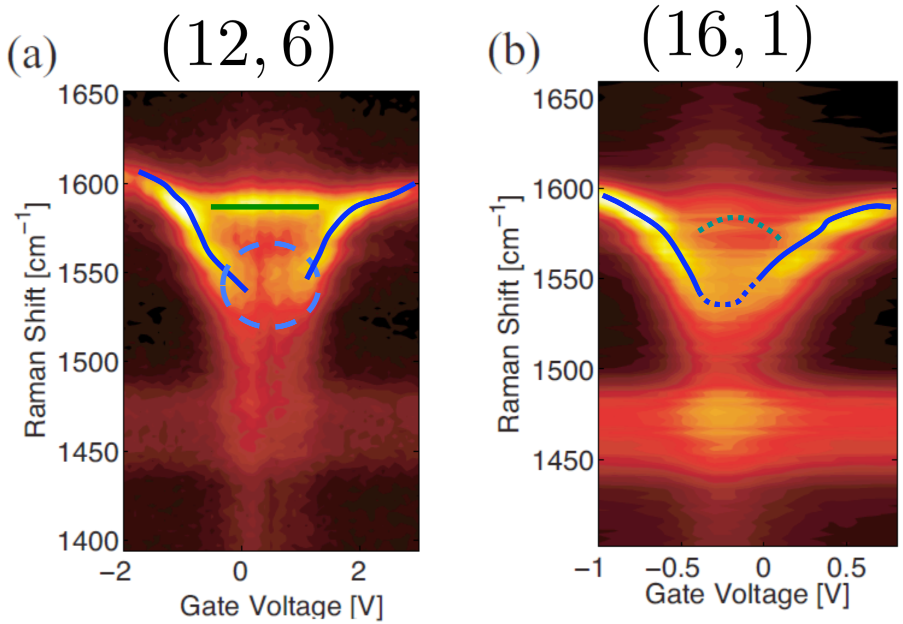
Motivation

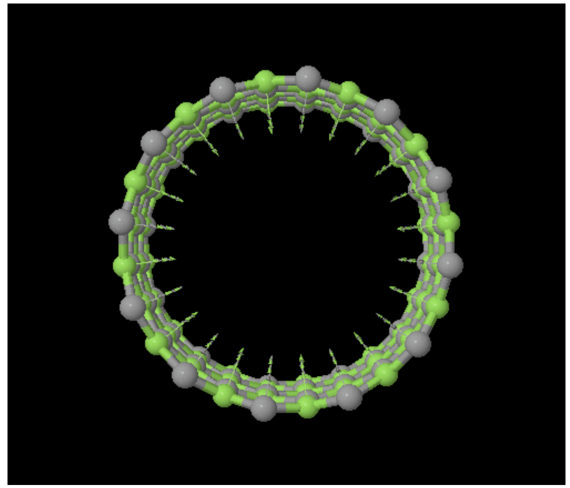
Chirality and **Fermi energy**

$\mathbf{C}_h$ $\qquad\qquad E_F$



G-band : LO/TO modes





$$\omega_{\rm LO/TO}(E_F, \mathbf{C}_h)$$

Chirality dependence originates from

(1) Curvature-induced mini energy gap

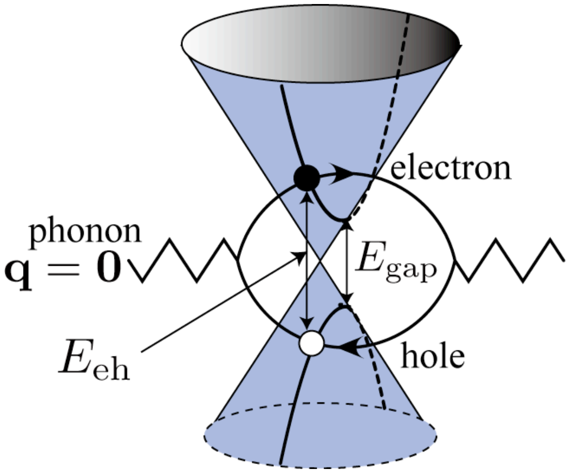
(2) Chirality dependent electron-phonon interaction

How about RBM?

Sasaki *et al*., PRB**78** (2008)

Farhat, Sasaki *et al*., PRL**102** (2009)

Phonon self-energy $\omega + \Delta\omega$



$$\Delta\omega = \sum_{\mathrm{eh}}^{\mathbf{q}=\mathbf{0}} \frac{|\langle \mathrm{eh}|V|\omega\rangle|^2}{\omega - E_{\mathrm{eh}} + i\Gamma}$$

$$\Gamma = -\mathrm{Im}\{\Delta\omega\}$$

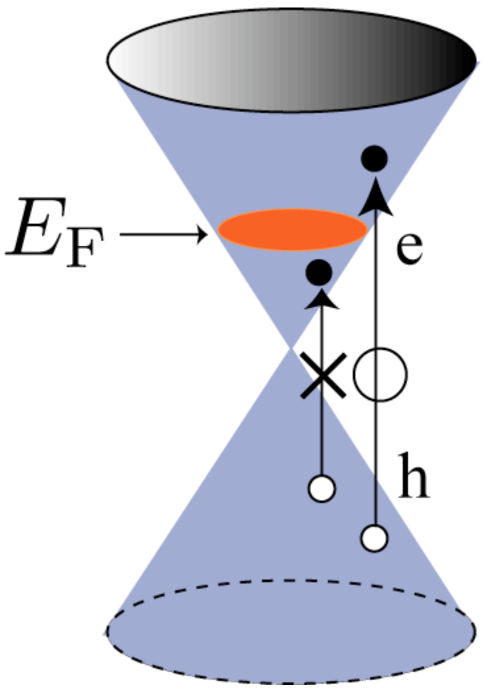
$$e^{-i(\omega+\Delta\omega)t} = e^{-i(\omega+\mathrm{Re}(\Delta\omega))t} e^{-\Gamma t}$$

Fermi energy and Chirality

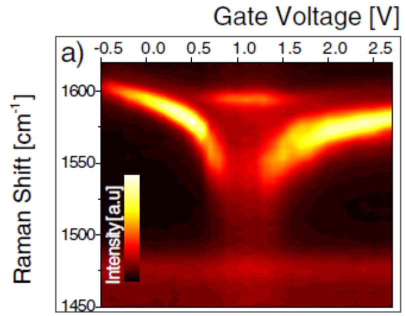
$$\Delta\omega(E_F, \mathbf{C}_h)$$

Many theoretical works on G-band
Fermi energy dependence:
Piscanec *et al*., PRL**93** (2004)
Lazzeri *et al*., PRL**97** (2006)
Ishikawa *et al*., JPSJ**75** (2006)
Popov *et al*., PRB**73** (2006)
Caudal *et al*., PRB**75** (2007)
Das *et al*., PRL**99** (2007)

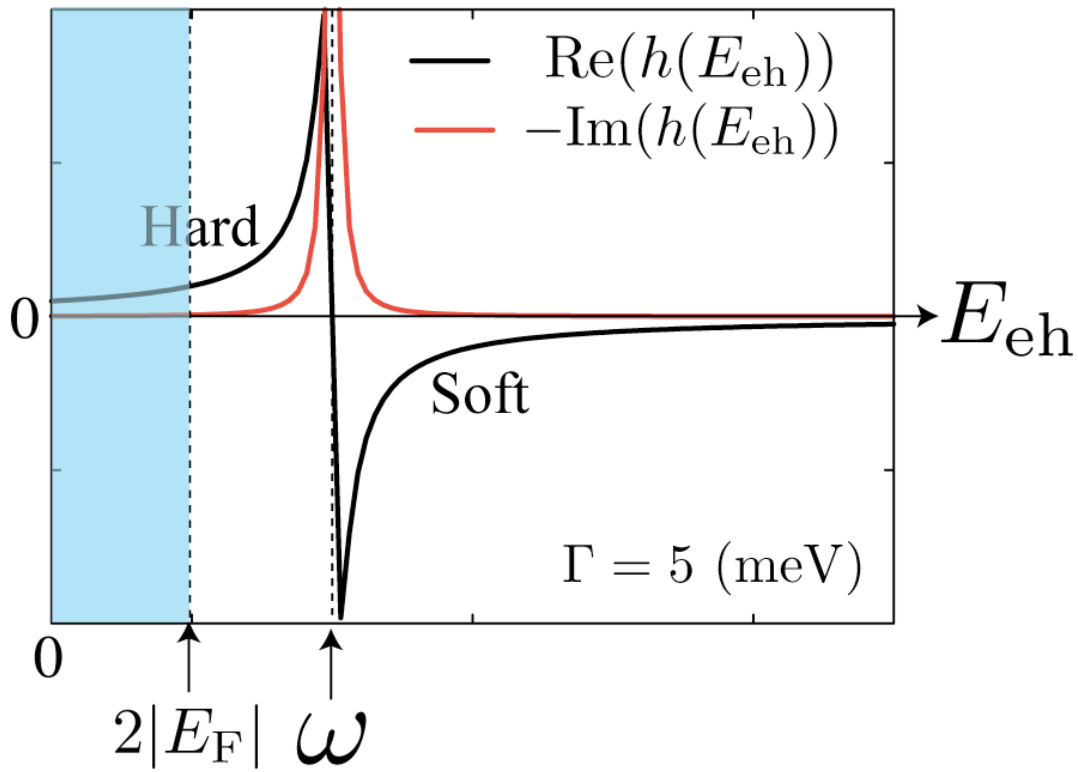
Fermi energy dependence $\Delta\omega(E_F)$



$$\Delta\omega = \sum_{\rm eh}^{\mathbf{q=0}} \frac{|\langle {\rm eh}|V|\omega\rangle|^2}{\omega - E_{\rm eh} + i\Gamma}$$



$$h(E_{\rm eh}) = \frac{1}{\omega - E_{\rm eh} + i\Gamma}$$

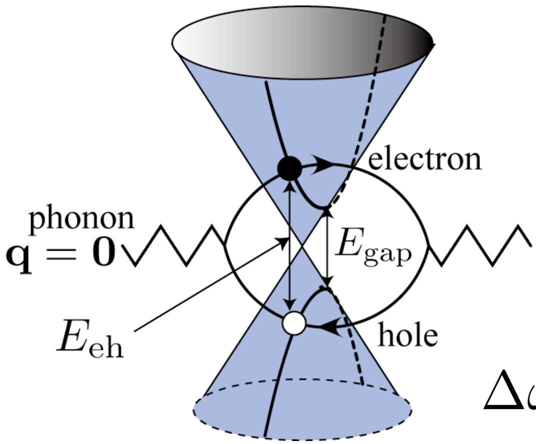


Pauli's exclusion principle

$$E_{\rm eh} \geq 2|E_F|$$

Chirality dependence

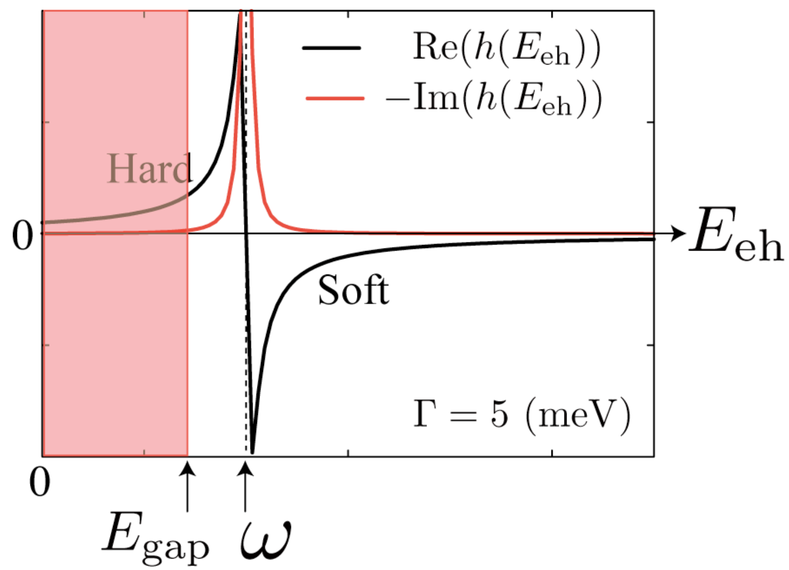
$\Delta\omega(E_F, \mathbf{C}_h)$

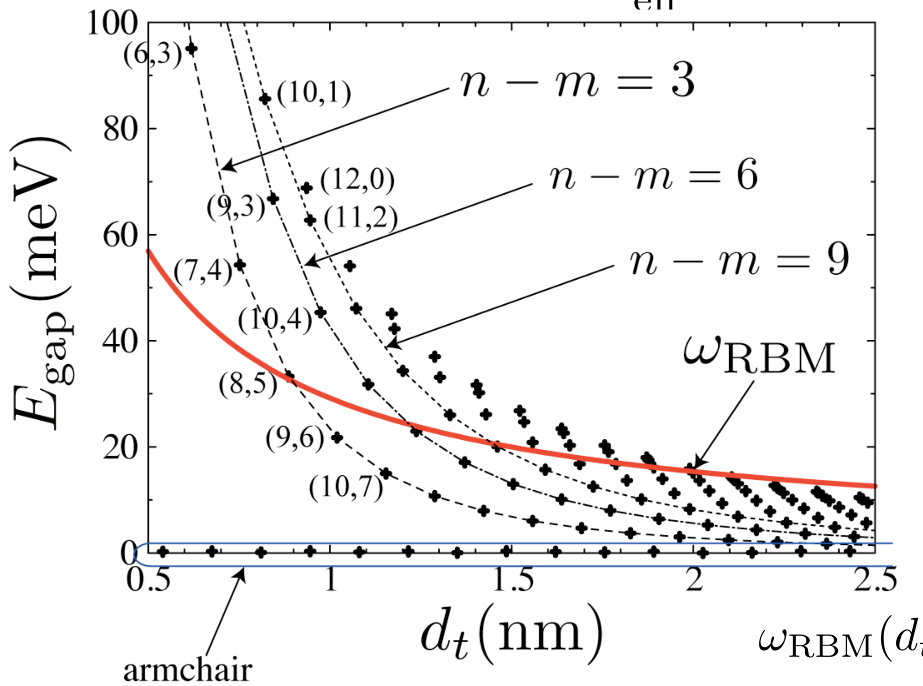


$$E_{\rm eh} \geq E_{\rm gap}$$

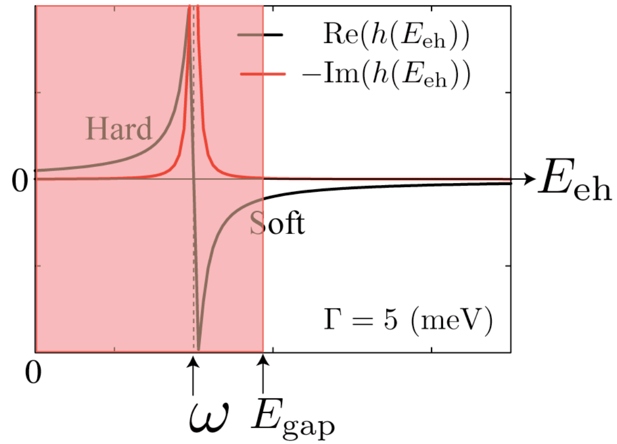
Curvature-induced energy gap

$$\Delta\omega = \sum_{\rm eh}^{\mathbf{q}=\mathbf{0}} \frac{|\langle {\rm eh}|V|\omega\rangle|^2}{\omega - E_{\rm eh} + i\Gamma}$$





$$\omega_{\rm RBM}(d_t) = \frac{c_1}{d_t} + c_2$$



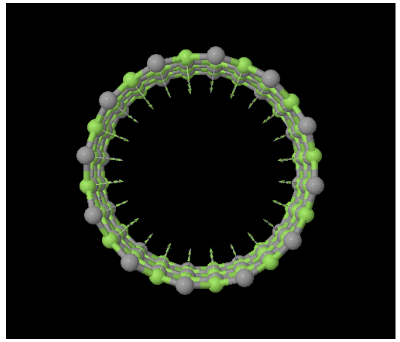
Zigzag SWNTs

$$\Gamma = 0$$

extended tight-binding calculation:
G. G. Samsonidze, Saito *et al.*, APL. 85 (2004)

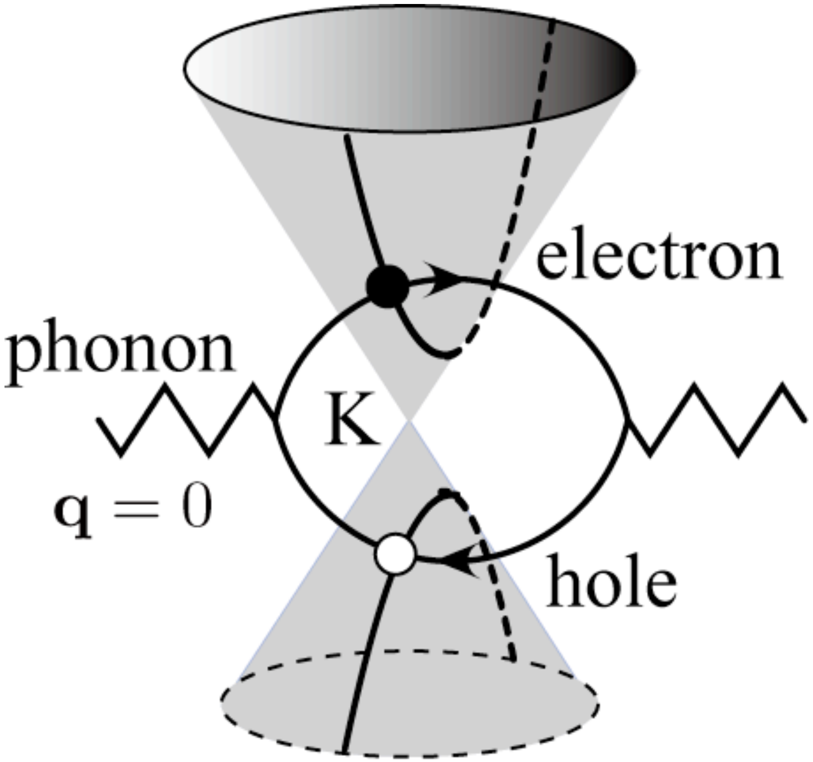
El-ph interaction of RBM

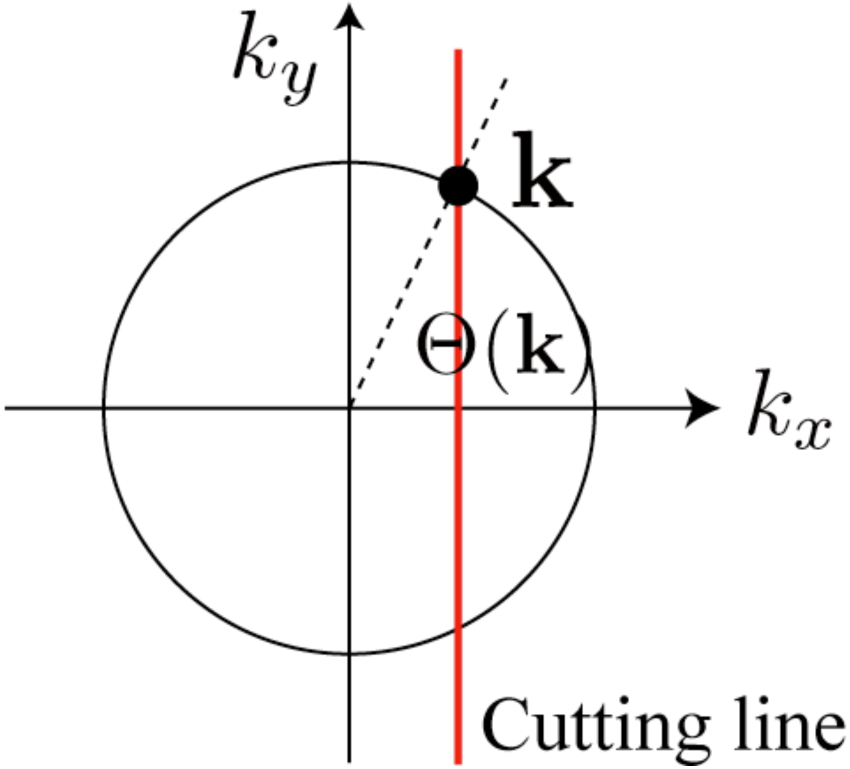
$$\langle \mathrm{eh}(\mathbf{k})|H_{\mathrm{ep}}|\omega_{\mathrm{RBM}}\rangle = i g_{\mathrm{off}} \frac{s_z}{d_t} \sin\left[\Theta(\mathbf{k}) + 3\theta\right]$$



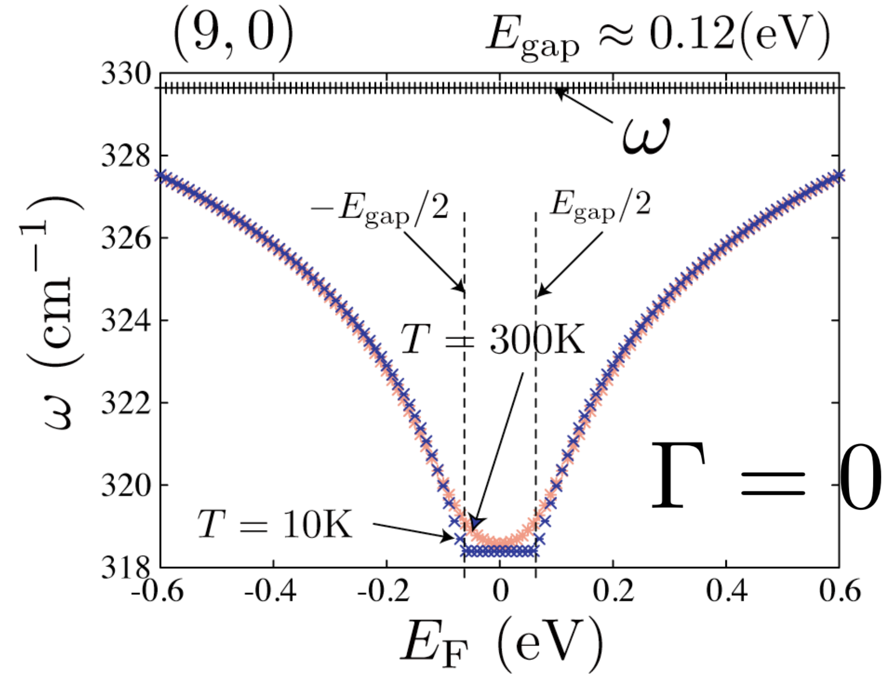
Suzuura *et al*., PRB**65** (2002)
Sasaki *et al*., PRB**78** (2008)

$$s_z = \frac{\hbar}{\sqrt{2M\omega}} \approx 0.4\sqrt{\frac{1\mathrm{meV}}{\omega}}\,\text{Å}$$

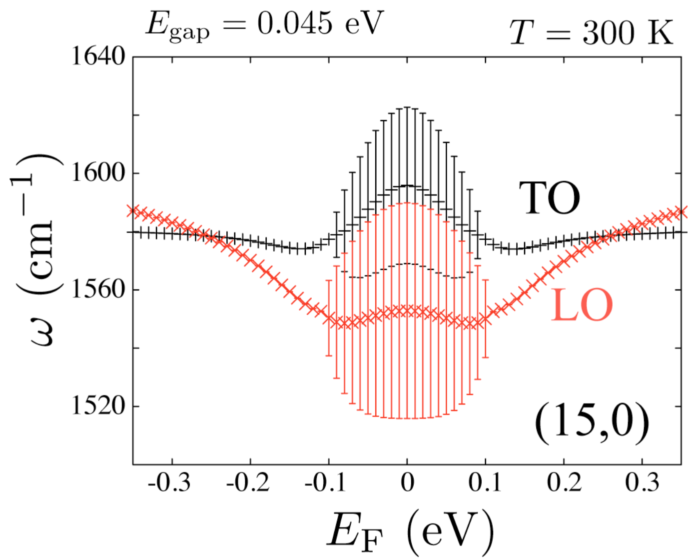




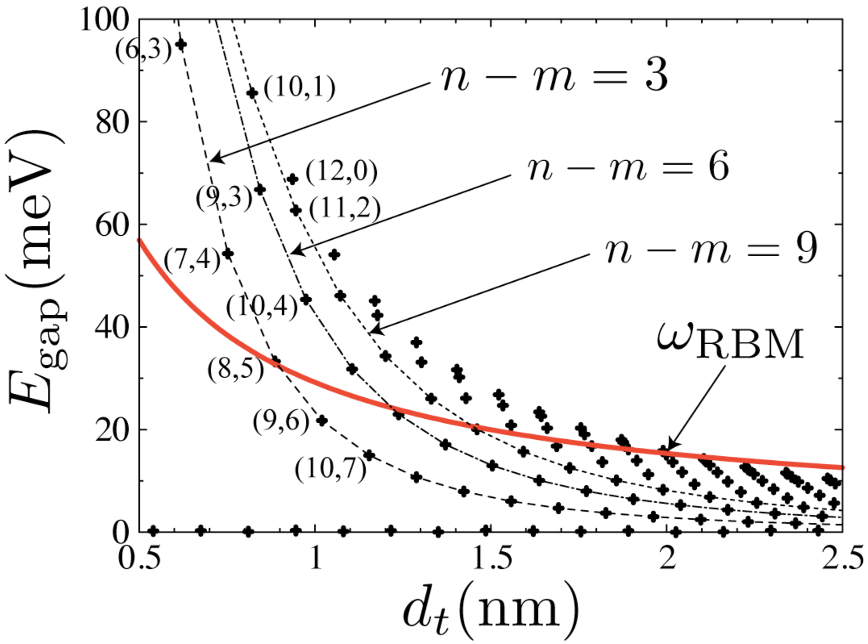
(9,0) SWNT



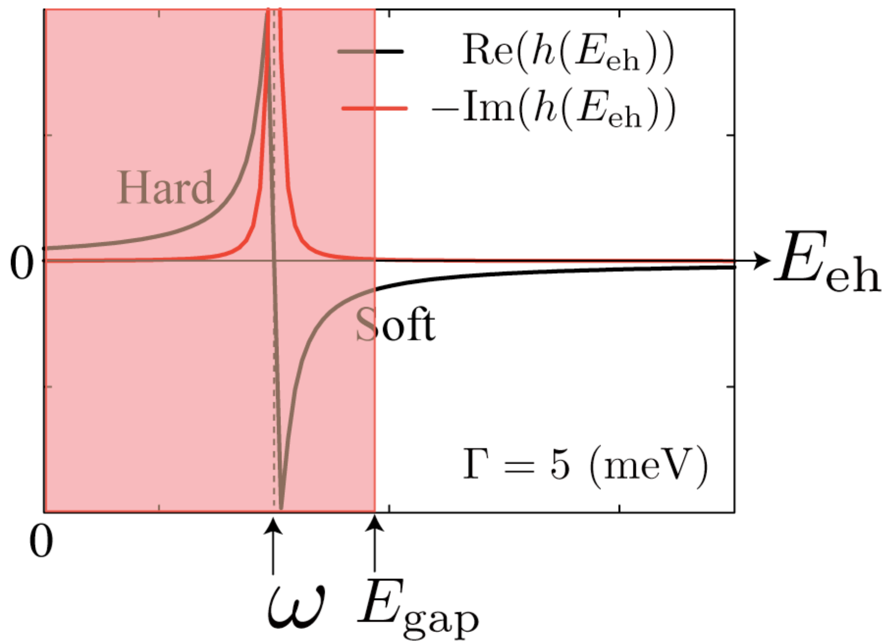
$(9,0)$
$E_{\rm gap} \approx 0.12(\rm eV)$
ω
$-E_{\rm gap}/2$
$E_{\rm gap}/2$
$T = 300$K
$T = 10$K
$\Gamma = 0$
$\omega\ (\rm cm^{-1})$
$E_{\rm F}\ (\rm eV)$



$E_{\rm gap} = 0.045$ eV
$T = 300$ K
TO
LO
(15,0)
$\omega\ (\rm cm^{-1})$
$E_{\rm F}\ (\rm eV)$

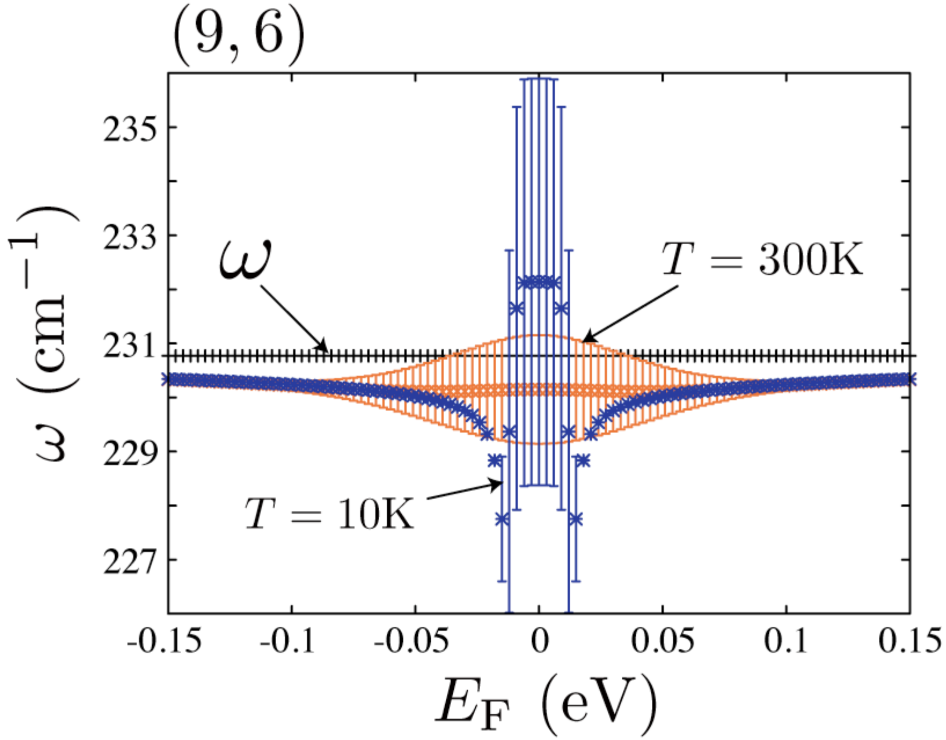


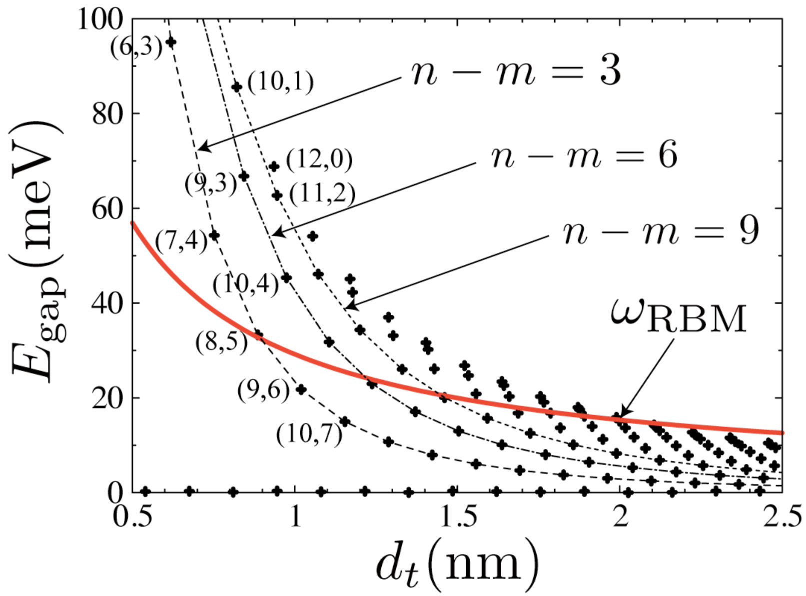
$n - m = 3$
$n - m = 6$
$n - m = 9$
$\omega_{\rm RBM}$
(6,3)
(10,1)
(12,0)
(9,3)
(11,2)
(7,4)
(10,4)
(8,5)
(9,6)
(10,7)
$E_{\rm gap}(\rm meV)$
$d_t(\rm nm)$

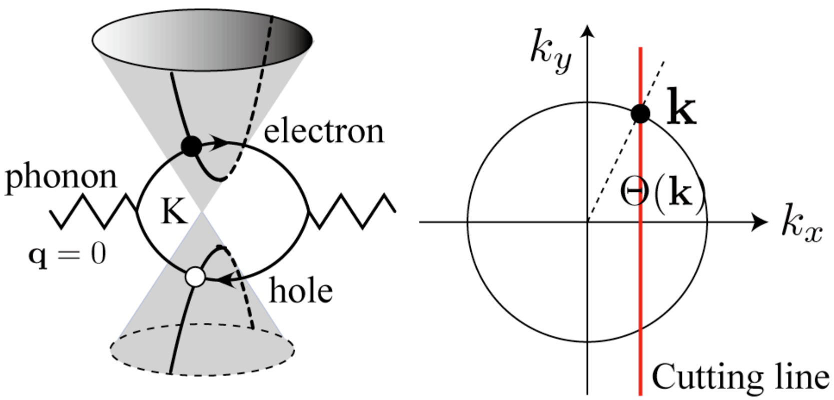


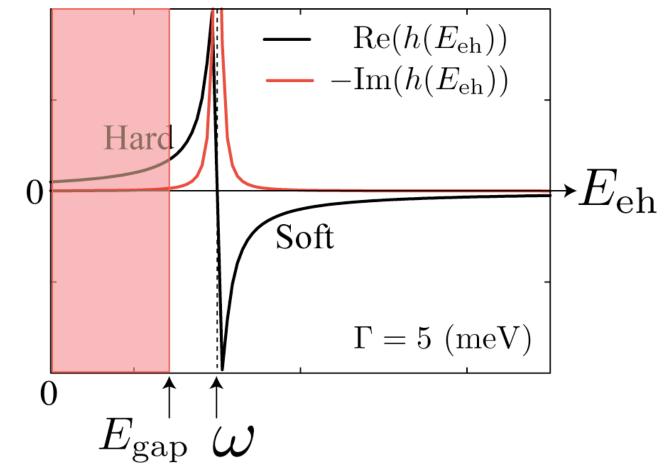
$\mathrm{Re}(h(E_{\rm eh}))$
$-\mathrm{Im}(h(E_{\rm eh}))$
Hard
Soft
$E_{\rm eh}$
$\Gamma = 5$ (meV)
0
ω
$E_{\rm gap}$

(9,6) SWNT $\theta = 24^\circ$









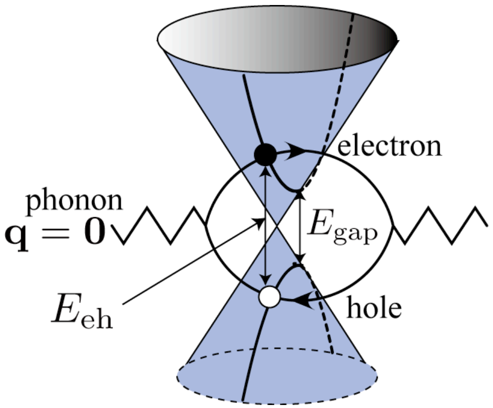
$$\langle \mathrm{eh}(\mathbf{k})|H_{\mathrm{ep}}|\omega_{\mathrm{RBM}}\rangle = i g_{\mathrm{off}} \frac{s_z}{d_t} \sin\left[\Theta(\mathbf{k}) + 3\theta\right]$$

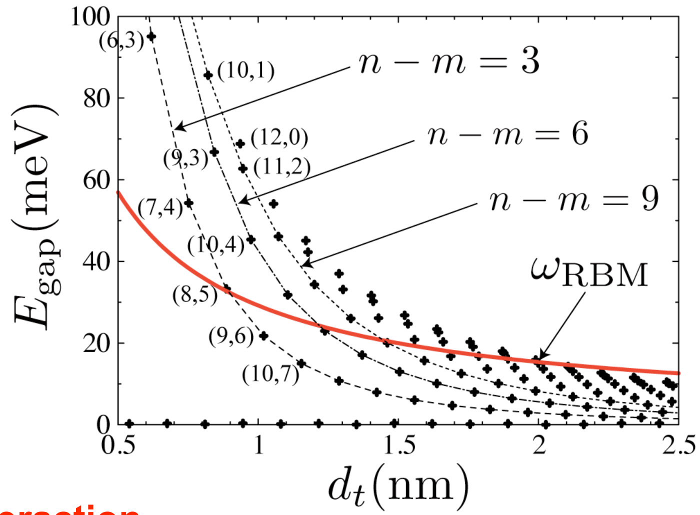
$$\Gamma_{\mathrm{RBM}}(\mathrm{RT}) \leq 2\mathrm{cm}^{-1}$$

Summary

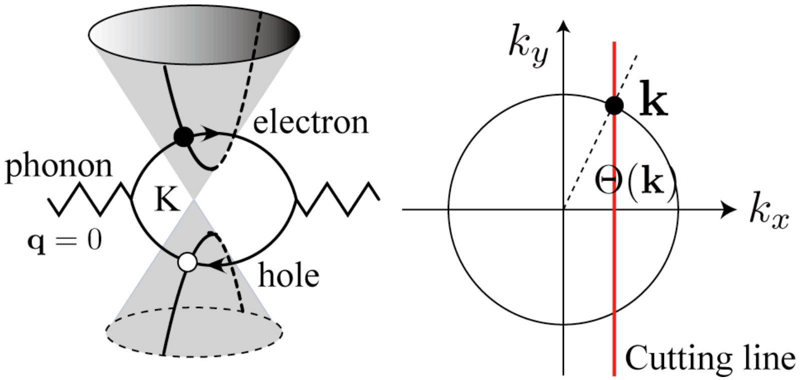
$$\Delta\omega(E_F, \mathbf{C}_h)$$

Softening of RBM in metallic SWNTs





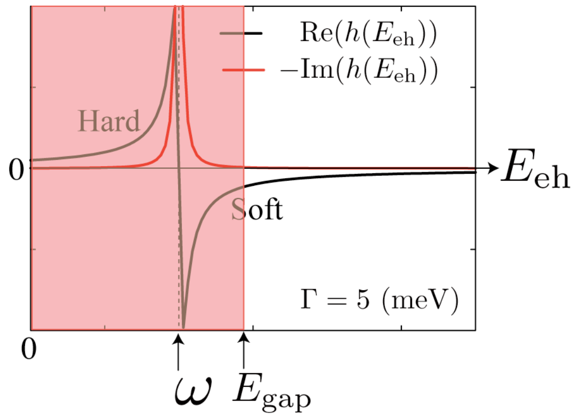
Chirality dependent e-p interaction



$$\langle \mathrm{eh}(\mathbf{k})|H_{\rm ep}|\omega_{\rm RBM}\rangle = i g_{\rm off}\frac{s_z}{d_t}\sin\left[\Theta(\mathbf{k})+3\theta\right]$$

Curvature effect

$$E_{\rm eh} \geq E_{\rm gap}$$



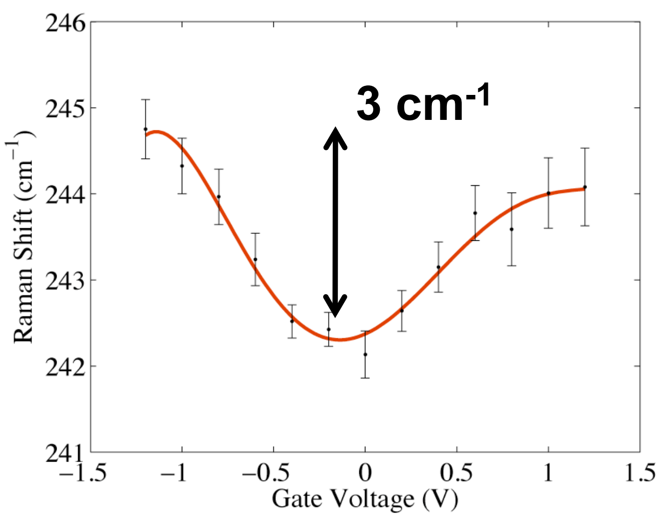
$$\Gamma_{\rm RBM}(\mathrm{RT}) \leq 2\mathrm{cm}^{-1}$$

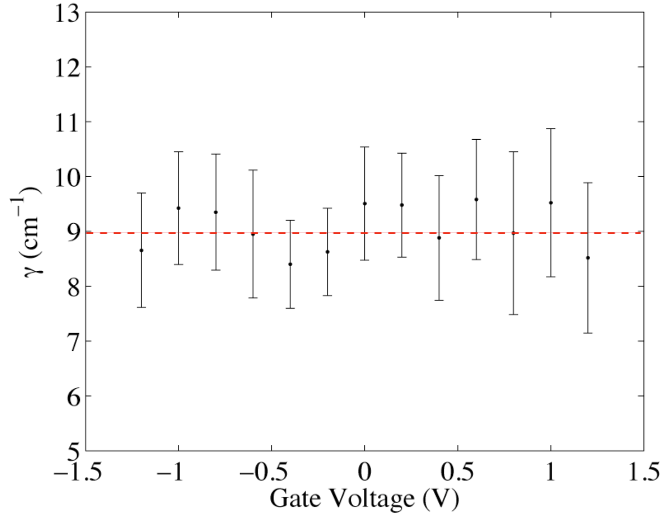
Sasaki *et al.*, PRB**78** (2008)
Farhat, Sasaki *et al.*, PRL**102** (2009)

Experiments

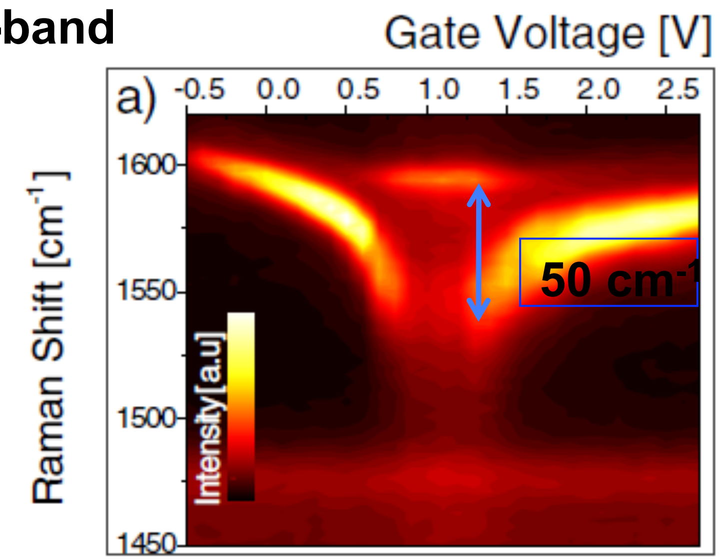
(12,0) or (11,2)

RBM

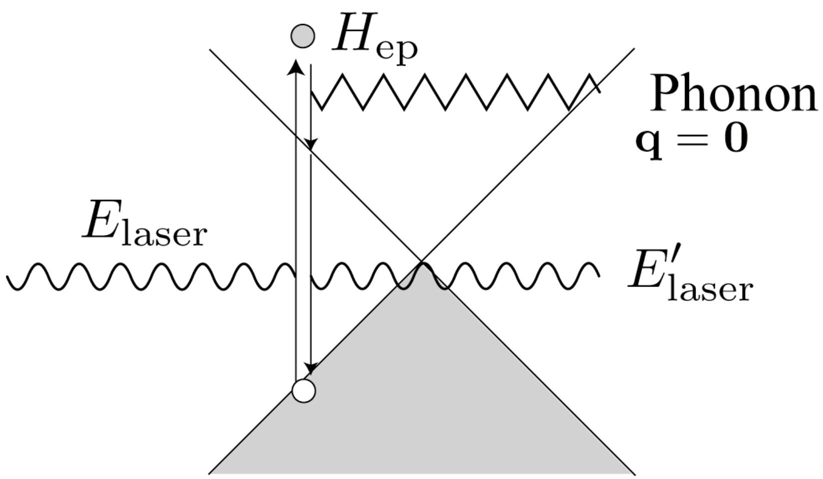




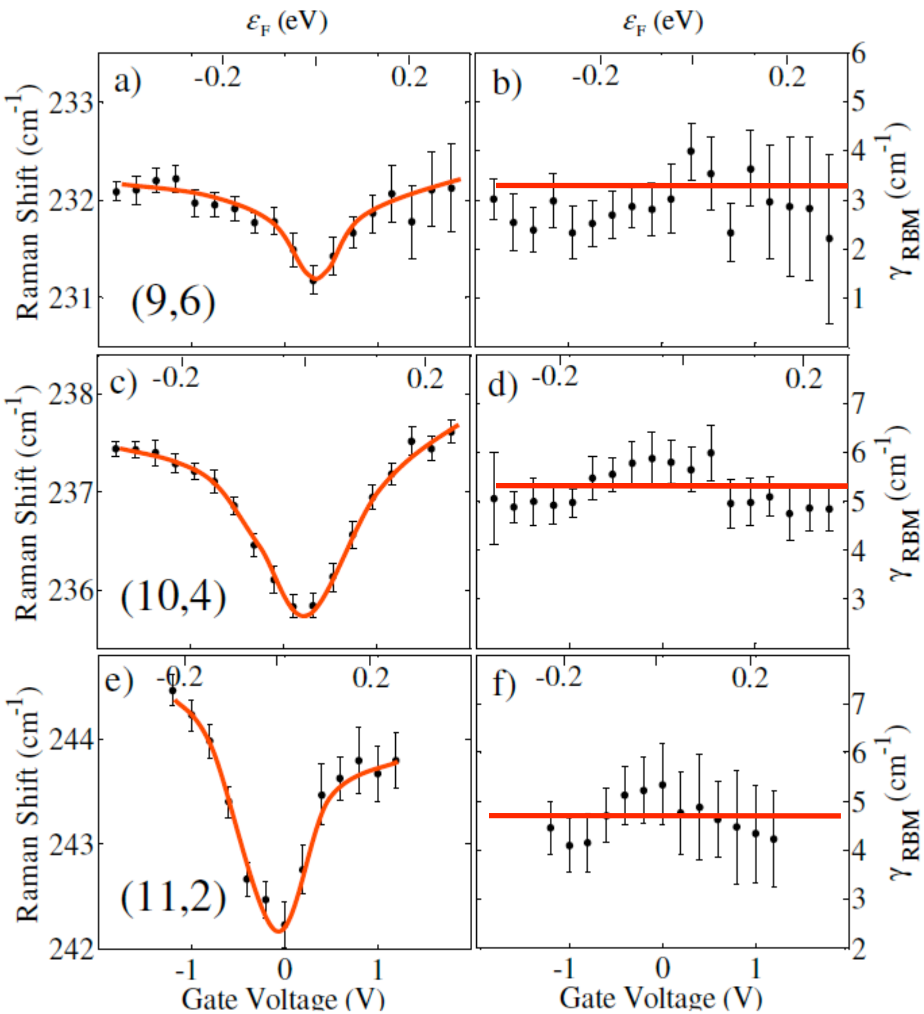
G-band



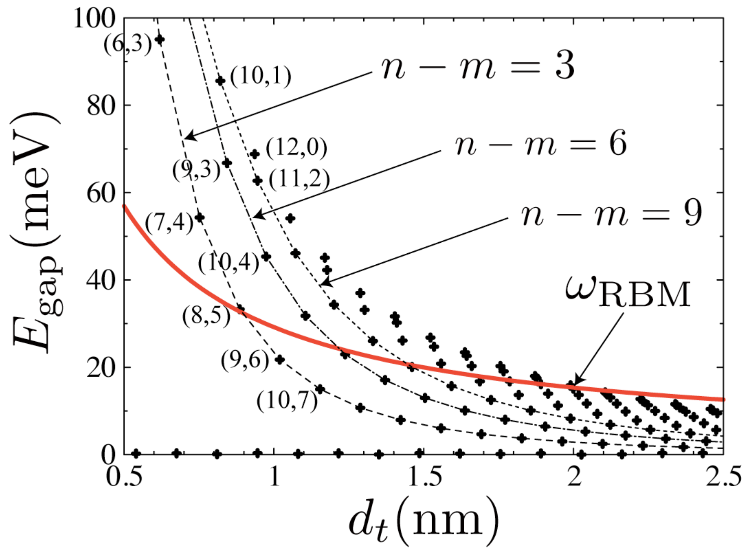
$$\Gamma = \Gamma_{\mathrm{e}} + \Gamma_{\mathrm{RBM}}$$



Metallic-SWNT experiments



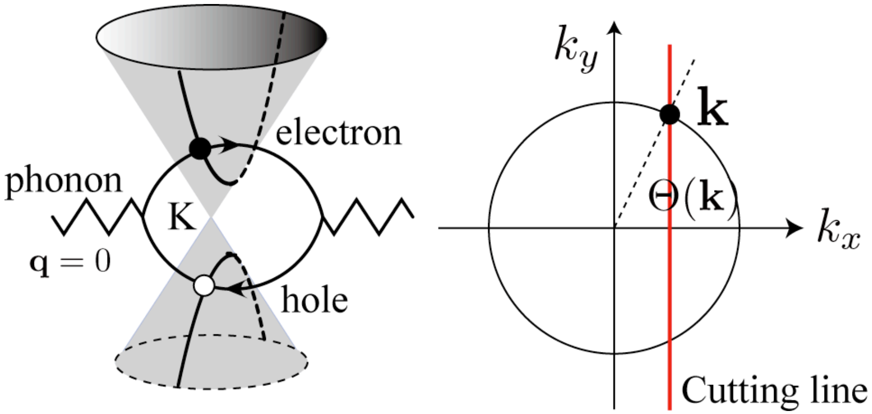
Farhat *et al.*, PRL**102** (2009)

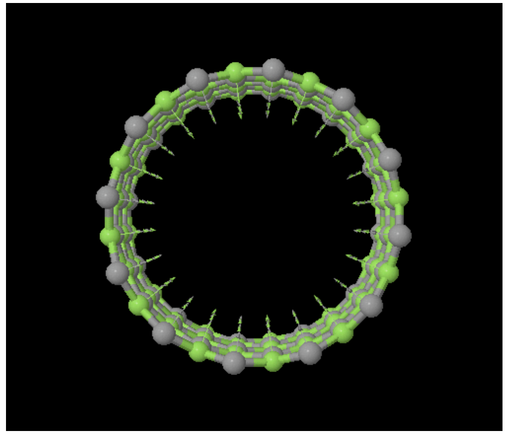


Uniaxial and torsional strain

C. L. Kane and E. J. Mele, Phys. Rev. Lett. 78, 1932 (1997).
L. Yang, *et al.*, Phys. Rev. B 60, 13874 (1999).
L. Yang and J. Han, Phys. Rev. Lett. 85, 154 (2000).

Relationship between RBM and LO/TO





$$\langle \mathrm{eh}(\mathbf{k})|H_{\mathrm{ep}}|\omega_{\mathrm{LO}}\rangle = -ig_{\mathrm{off}}\frac{u_{\mathrm{LO}}}{a_{\mathrm{cc}}}\sin\Theta(\mathbf{k})$$

$$\langle \mathrm{eh}(\mathbf{k})|H_{\mathrm{ep}}|\omega_{\mathrm{TO}}\rangle = -ig_{\mathrm{off}}\frac{u_{\mathrm{TO}}}{a_{\mathrm{cc}}}\cos\Theta(\mathbf{k})$$

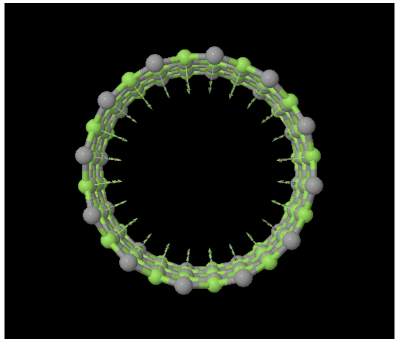
$$|u_{\mathrm{LO/TO}}| = 0.04\text{Å}$$

$$\langle \mathrm{eh}(\mathbf{k})|H_{\mathrm{ep}}|\omega_{\mathrm{RBM}}\rangle = ig_{\mathrm{off}}\frac{s_z}{d_t}\sin\left[\Theta(\mathbf{k})+3\theta\right]$$

$$|s_z| = 0.13\text{Å}$$

$$d_t = 1.4\mathrm{nm}$$

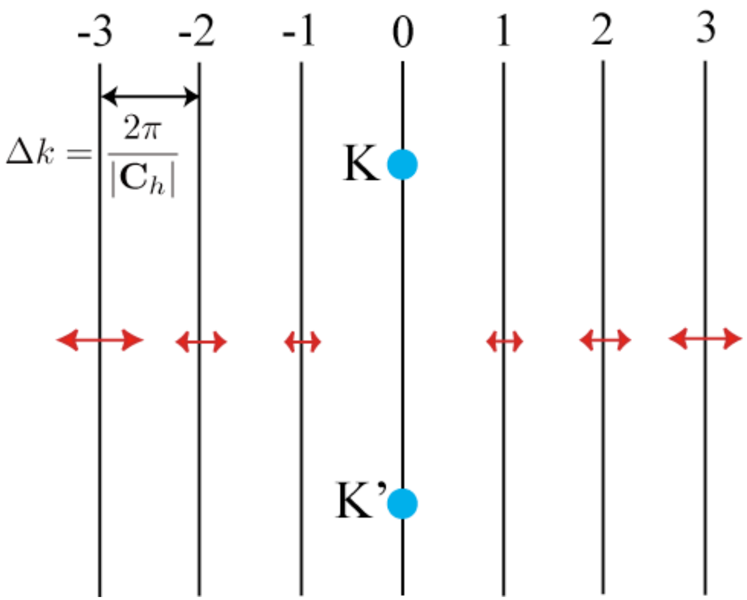
El-ph interaction of RBM



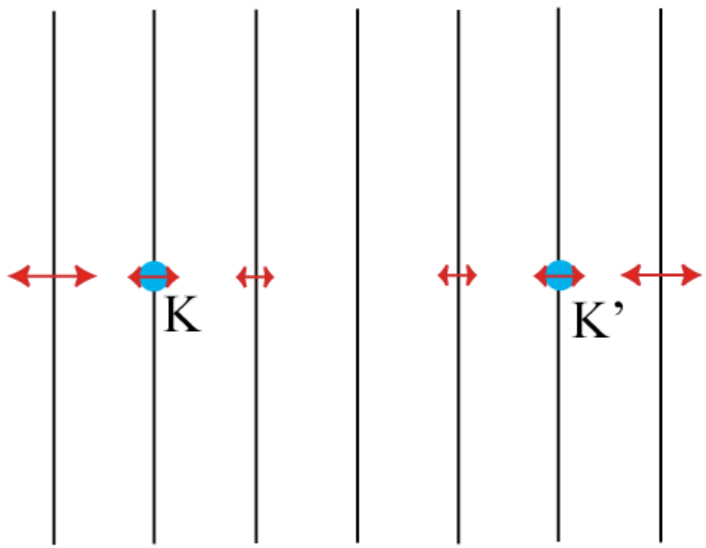
$$k_n = \frac{2n}{d_t}$$

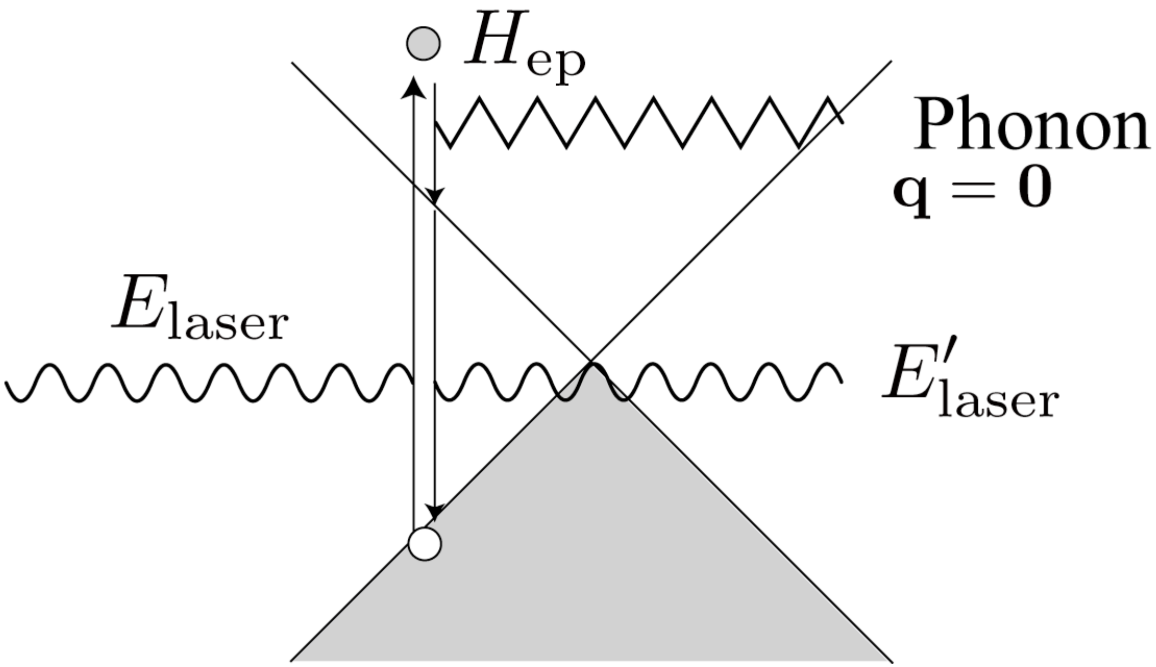
$$k_n = \frac{2n}{d_t + 2s_z} = \frac{2n}{d_t}\left(1 - \frac{2s_z}{d_t} + \cdots\right)$$

armchair SWNT



zigzag SWNT





$$\Gamma = \Gamma_{\mathrm{e}} + \Gamma_{\mathrm{RBM}}$$

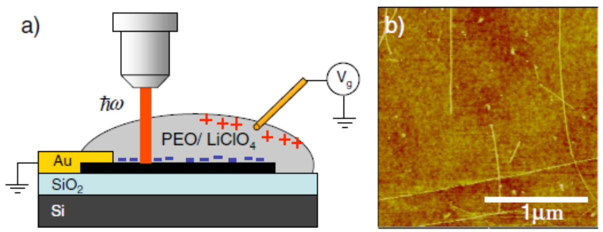


FIG. 1 (color online). (a) Schematic diagram of the experimental setup. The excitation laser shines through the PEO/$LiClO_4$ polymer electrolyte. (b) An AFM image indicating that the nanotubes are spaced out and are typically isolated from one another.