Coherent phonon model for radial breathing mode in carbon nanotubes

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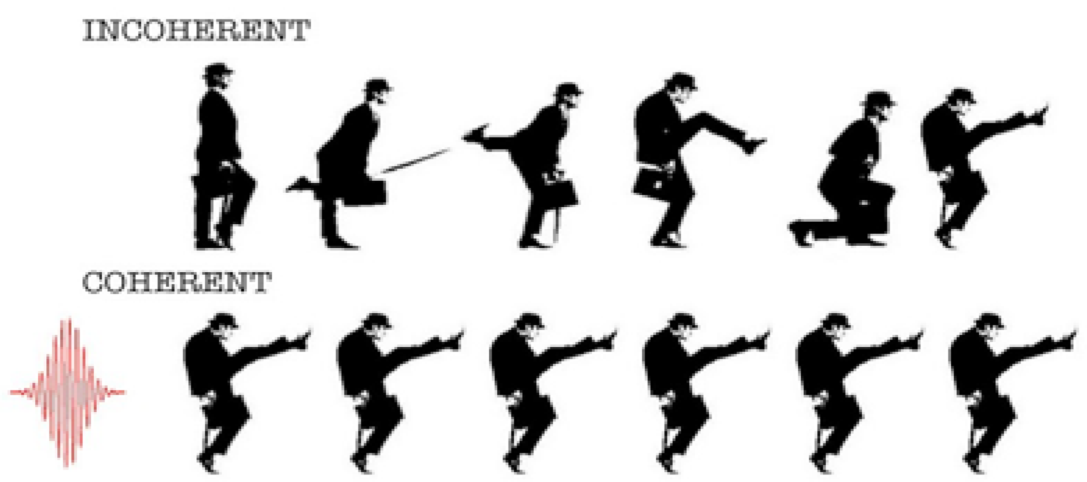
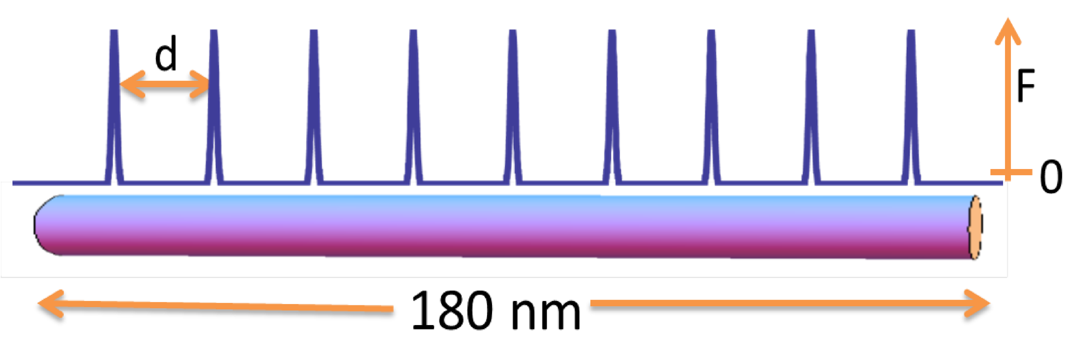
Purpose

- Force applied to carbon nanotube
- Solve equation of motion for tube
- Coherence of radial breathing mode



Coherent Phonon and RBM

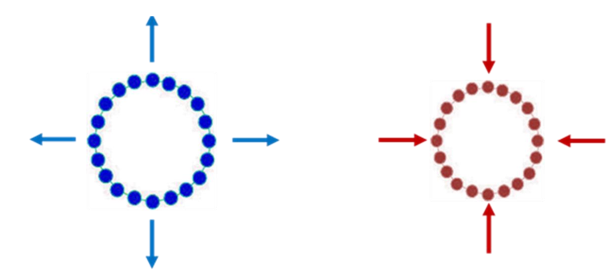
Exciton Force(x,t₀) along tube length

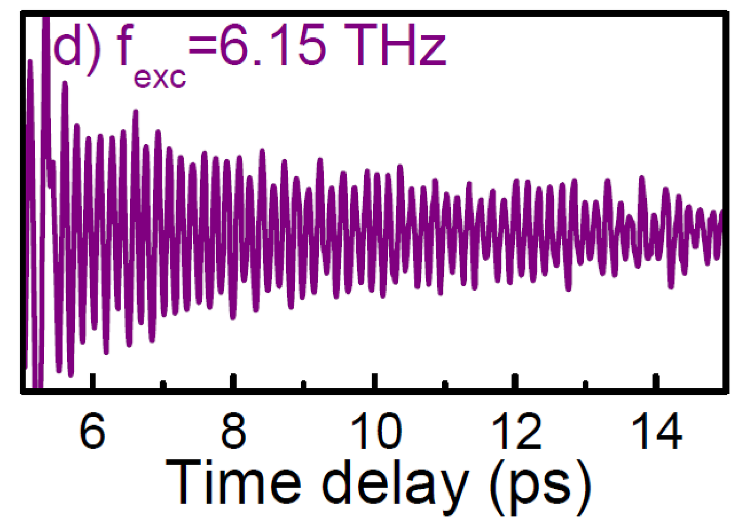


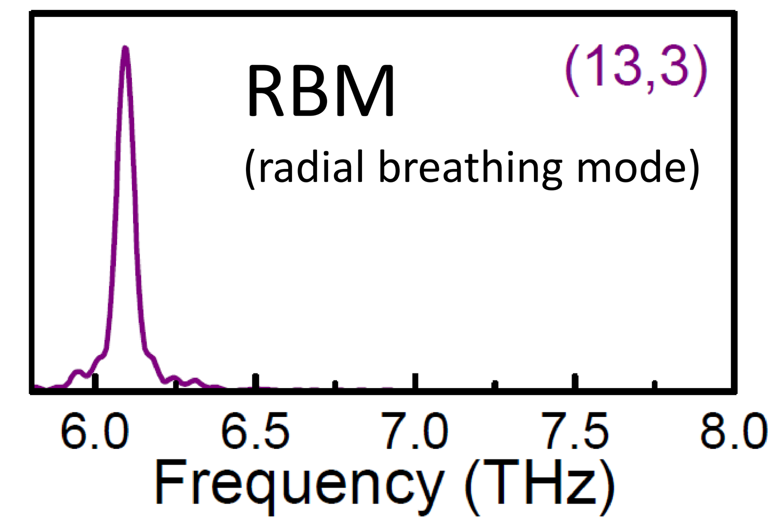
d = 18 nm

Gaussian force:

$$F_1 = Be^{-x^2/\alpha^2}\theta(t)$$







Results and Discussion

Fourier transform (with F = 0) shows RBM dispersion:

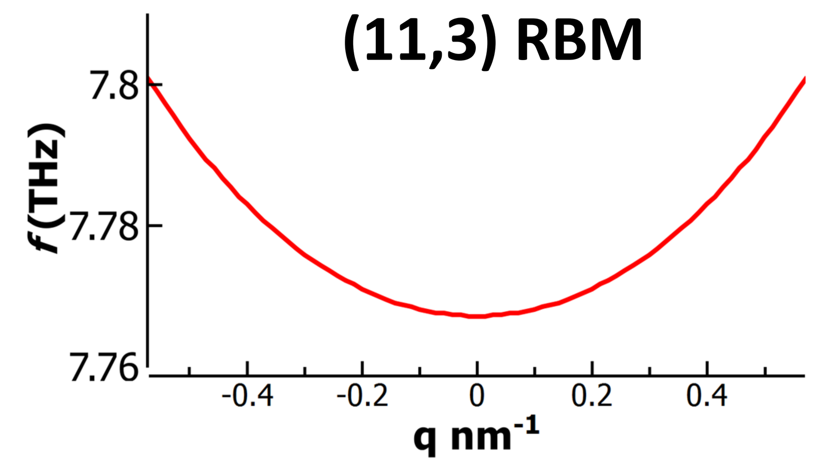
$$Q_{tt} - c^2 Q_{xx} = F - \kappa Q$$

$$\omega(q) = \sqrt{c^2 q^2 + \kappa}$$

Taylor expansion

$$\omega = \left(\frac{c^2}{2\sqrt{\kappa}}\right)q^2 + \sqrt{\kappa}$$

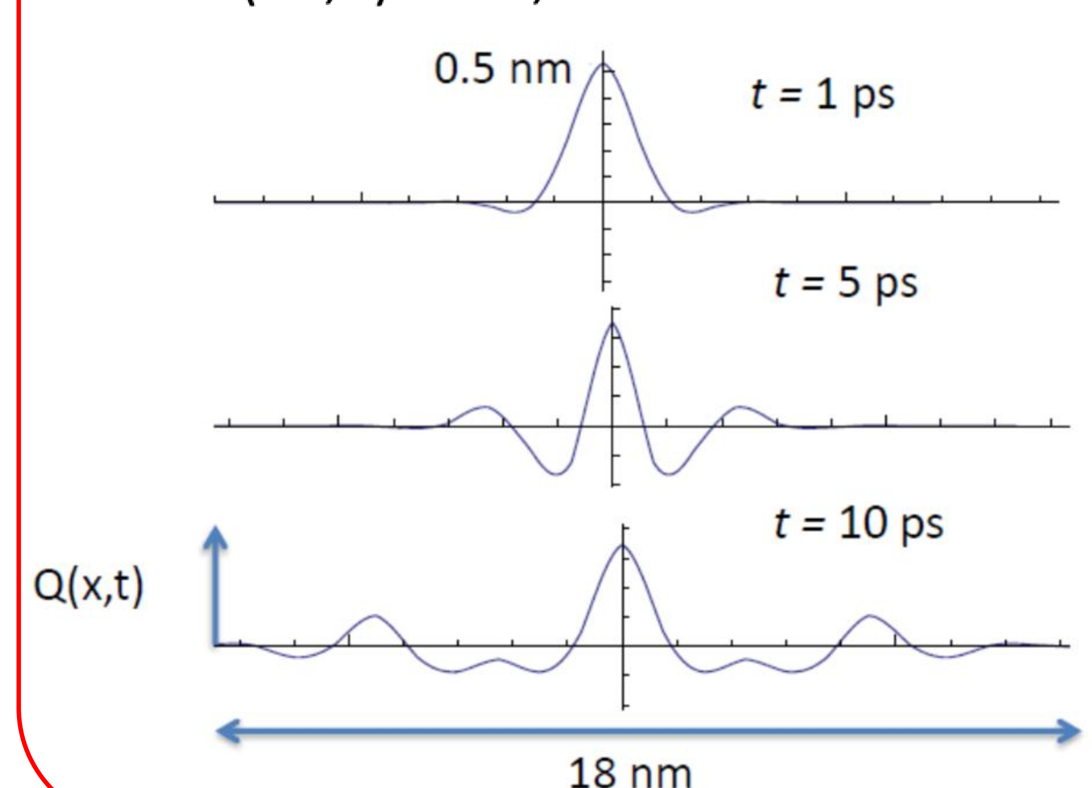
$$\omega = 0.101 q^2 + 7.767$$

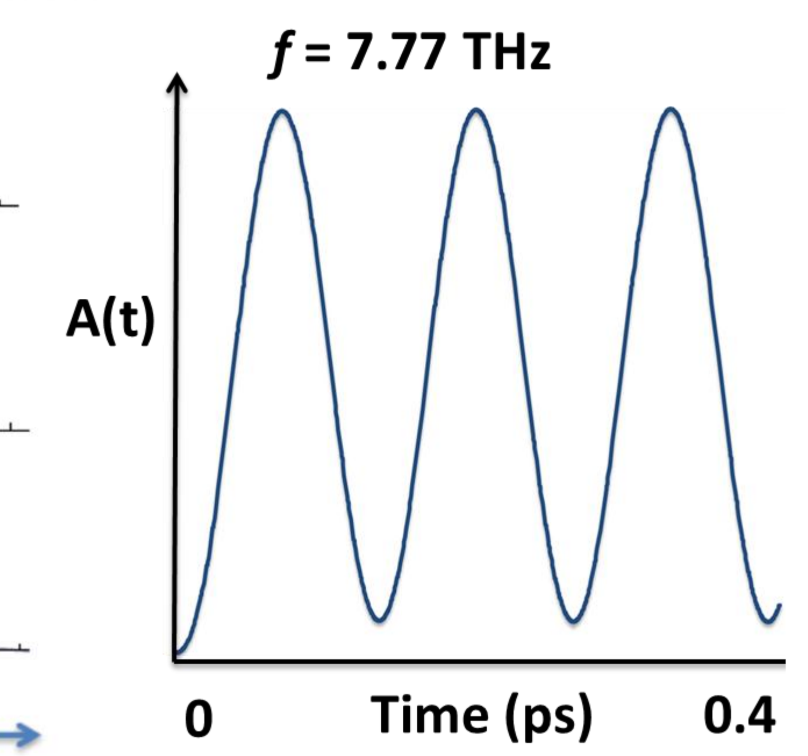


Quadratic fit, $R^2 = .999$

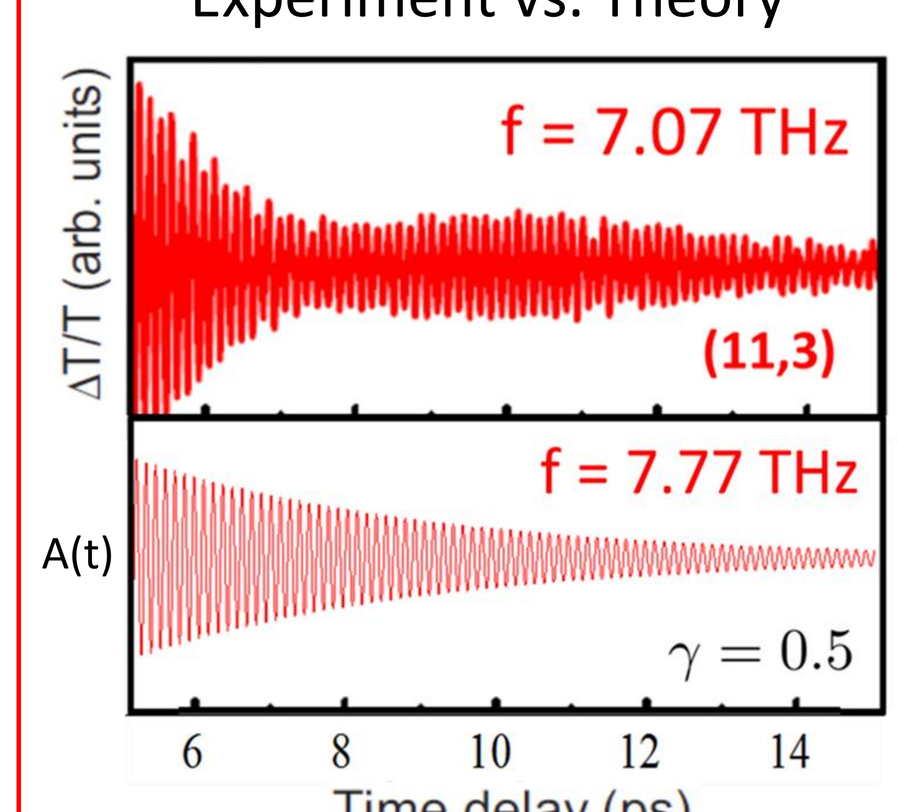
$$c = 1.255 \text{ nm/ps}$$
$$\kappa = 60.328 \text{ ps}^{-2}$$

(11,3) tube, Klein-Gordon solution





Experiment vs. Theory

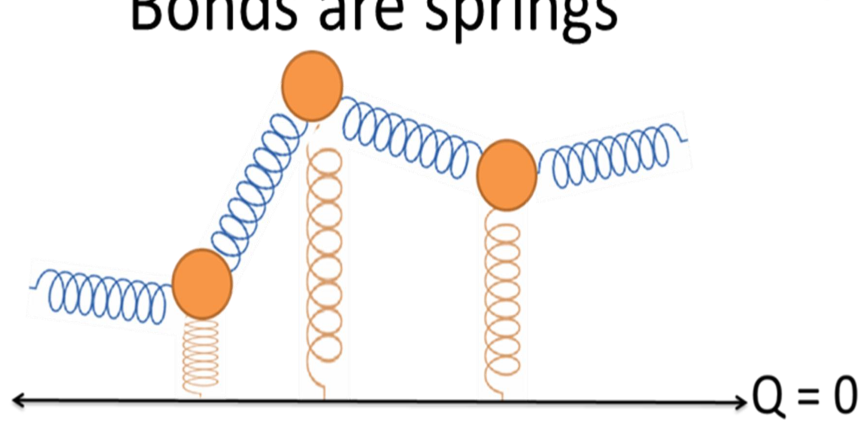


Summary

- Klein – Gordon equation to model RBM
- Cross section displacement shows coherent phonon

Calculation Method

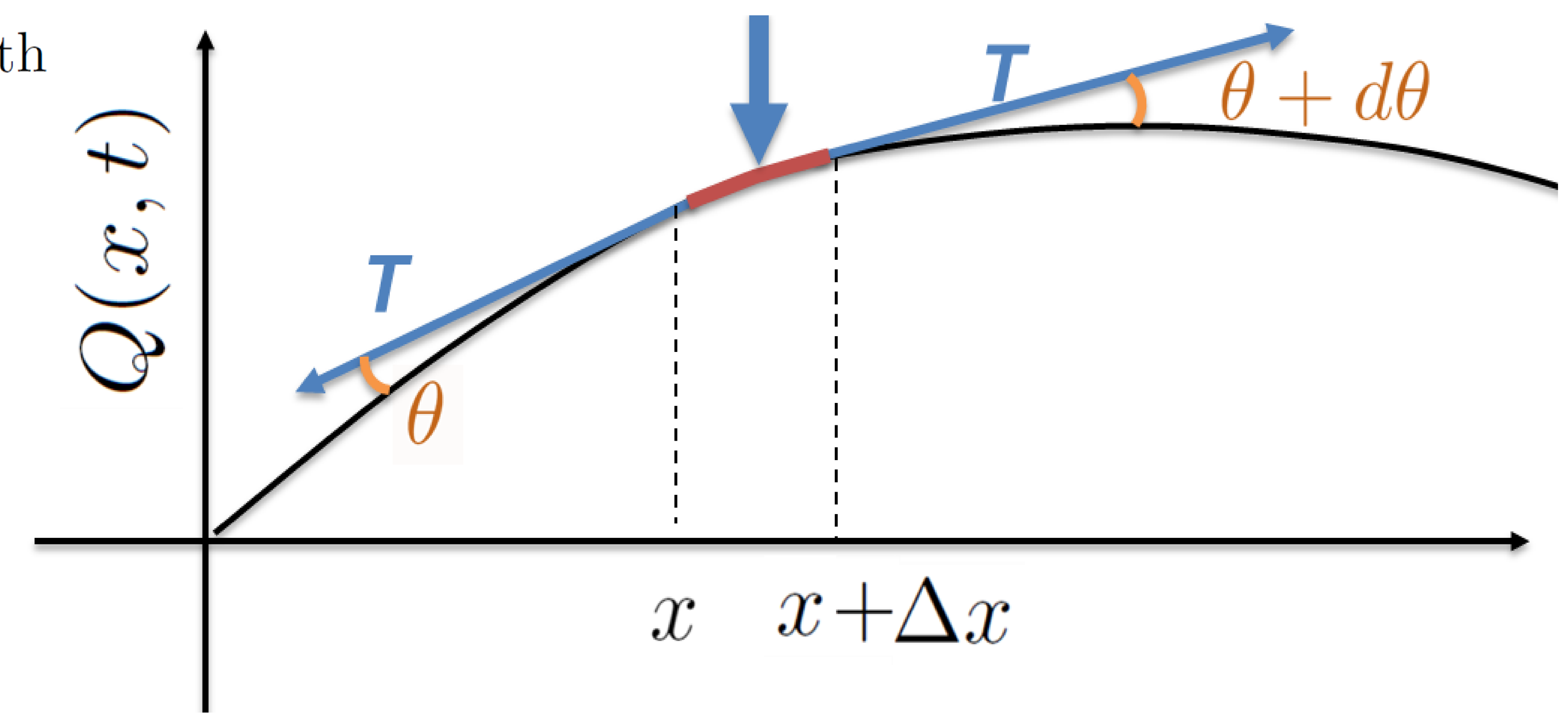
Bonds are springs



Nanotube is a 1D string:

K = spring const/length
T = tensile strength
ρ = mass/unit length
c = wave speed
κ = constant
γ = damping factor

$$F = -K\Delta x Q$$



$$\sum F = T[\sin(\theta + d\theta) - \sin(\theta)] - QKdx + F_{ex}$$

Small angle: $\sin(\theta + d\theta) = \sin(\theta) = \tan(\theta) = \frac{dQ}{dx}$

$$= T\left[\left(\frac{\partial Q}{\partial x}\right)_{x+dx} - \left(\frac{\partial Q}{\partial x}\right)_x\right] - QKdx + F_{ex} = Q_{tt}\rho dx$$

$$T\frac{\partial^2 Q}{\partial x^2}dx$$

Wave equation becomes Klein-Gordon equation:

$$\rightarrow \quad Q_{tt} - c^2 Q_{xx} = F - \kappa Q - \gamma Q_t$$

Cross sectional area: $A(t) = \int_L Q(x,t)dx$

$$\rightarrow \quad A_{tt} + \gamma A_t + \kappa A = C$$

Solved numerically using Mathematica

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A femtosecond laser pulse interacting with a single wall carbon nanotube (SWNT) sample can induce photo-excited carriers called excitons, which are electron-hole pairs bound by the Coulomb interaction. Before exciton recombination, the lattice starts to vibrate coherently through the exciton-phonon interaction. This interaction becomes the driving force for coherent vibrations of the nanotube. Of the many different types of phonon modes, we specifically consider the radial breathing mode (RBM) of the SWNT in which the tube diameter expands and contracts in the radial direction.

The excitonic driving force is spatially localized in a Gaussian shape along the tube axis, but we expect diffusion of displacement because experiments show uniform displacement of diameter. To understand the coherent RBM motion we model the nanotube as a one dimensional string with string displacement equivalent to change in nanotube diameter. We solve for the vibration amplitude as a function of space and time using the Klein-Gordon equation, which reproduces the RBM phonon dispersion. We solve this equation numerically using Mathematica, and considering realistic parameters of a nanotube. The calculated coherent motion provides a physical explanation for why a localized driving force gives the oscillation of reflectance in coherent phonon experiments.