Nanoelectromagnetics of low-dimensional nanostructures

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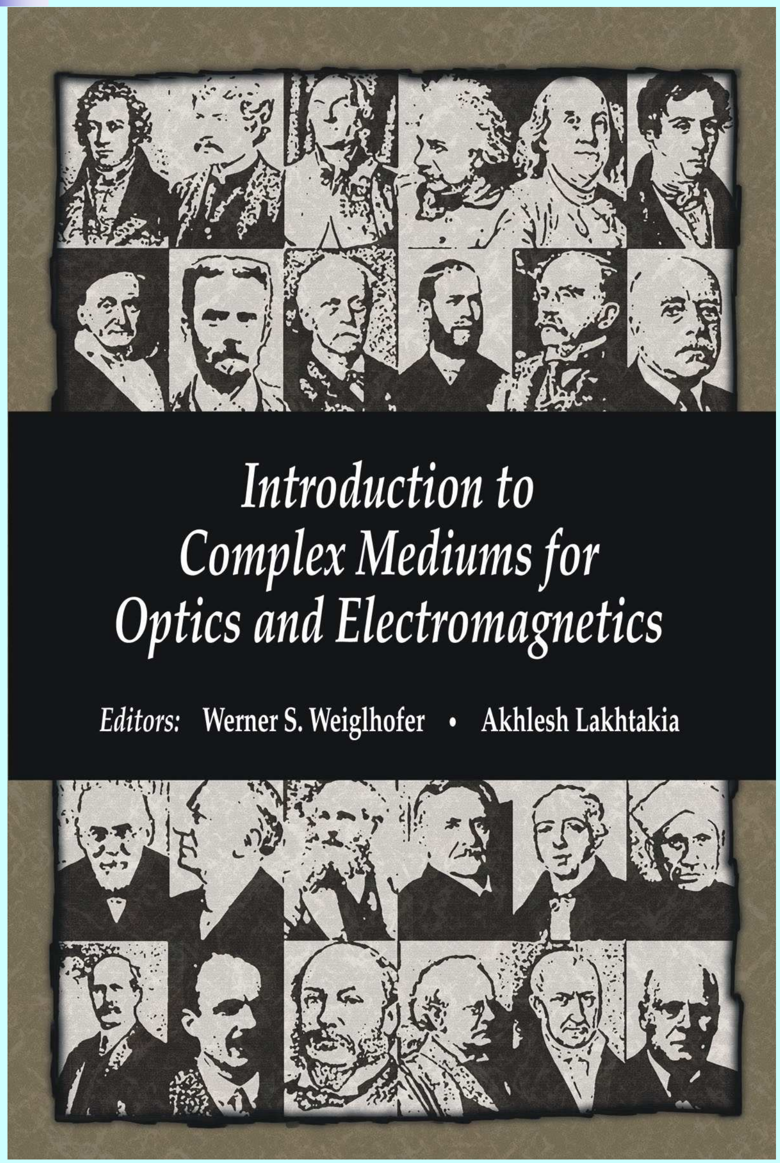
Dresden, CoPhen, MPI-PKS, 2004

MOTIVATION

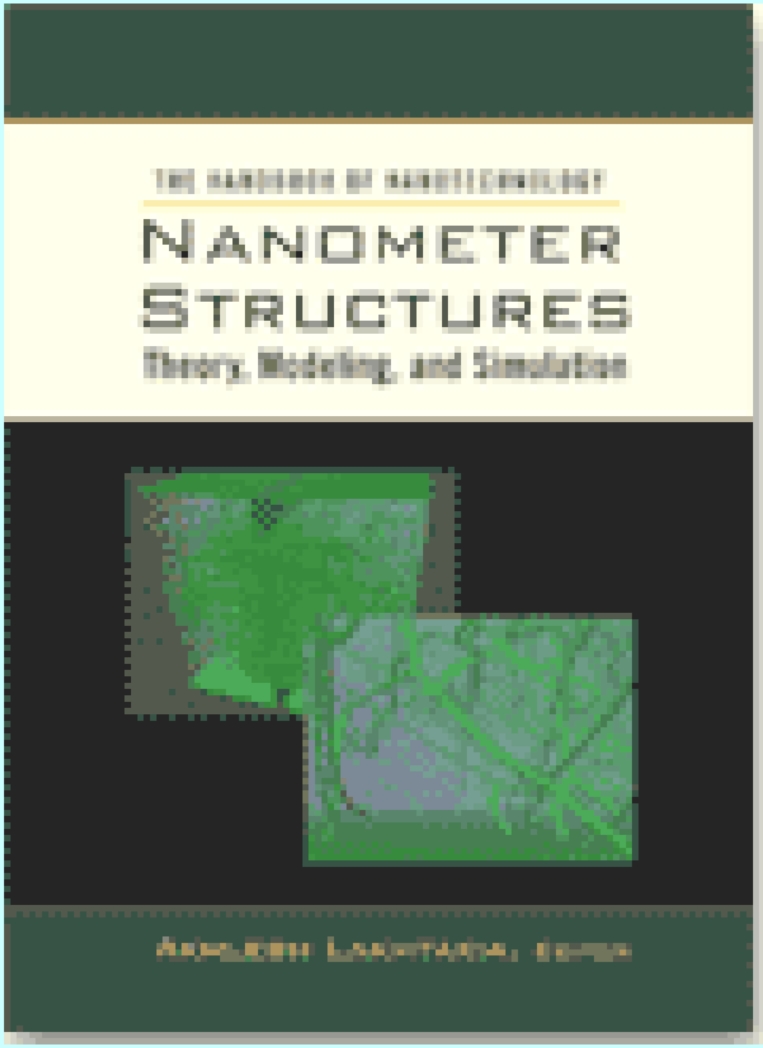
- To stress the role of nanoscale nonhomogeneity of electromagnetic fields in nanostructures
- To demonstrate the close connection between traditional problems of classical electrodynamics of microwaves and new problems arising in nanostructures
- To elucidate the peculiarities of electromagnetic problems in nanostructures irreducible to problems in classical ED due to the complex conductivity law

Electromagnetic effects in nanotubes



S.A.Maksimenko and G.Ya.Slepyan, Electromagnetics of Carbon Nanotubes, in "Introduction to Complex Mediums for Optics and Electromagnetics", SPIE Press Vol. PM 123, 2003.

Electromagnetic effects in nanotubes



S.A.Maksimenko and G.Ya.Slepyan, Nanoelectromagnetics of low-dimensional structures, in "Hand-book of Nanotechnology: Theory, Modeling and Simulation", Ed. by: A. Lakhtakia, SPIE Press, 2004, pp. 145-206 (in press).

NANOSTRUCTURES:

quantum wires and quantum dots, fullerenes, nanotubes, sculptured thin films, atomic clusters, nanocrystallites, etc.

Spatial nonhomogeneity

- Confinement of the charge carrier motion
- Electromagnetic field diffraction

complex geometry
complex electronics

Electromagnetic effects in nanotubes

Diffraction Theory

Boundary-value problems for complex-shaped regions:
Complex geometry, ordinary electronics

Solid State Physics

Quasi-particle concept:
Electrons, phonons, magnons…
Complex electronics, ordinary geometry

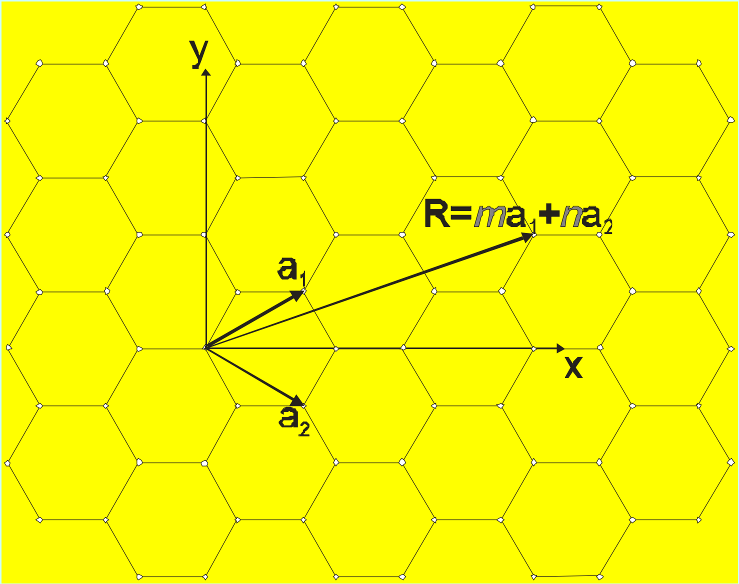
NANOELECTRODYNAMICS

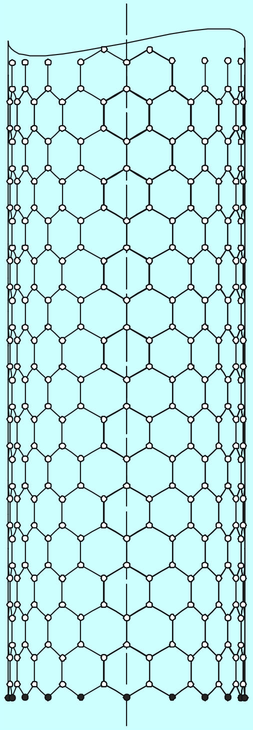
Main topics

- Linear electrodynamical response
- Nonlinear optics
- Quantum electrodynamics of CNTs

Electromagnetic effects in nanotubes

CARBON NANOTUBE





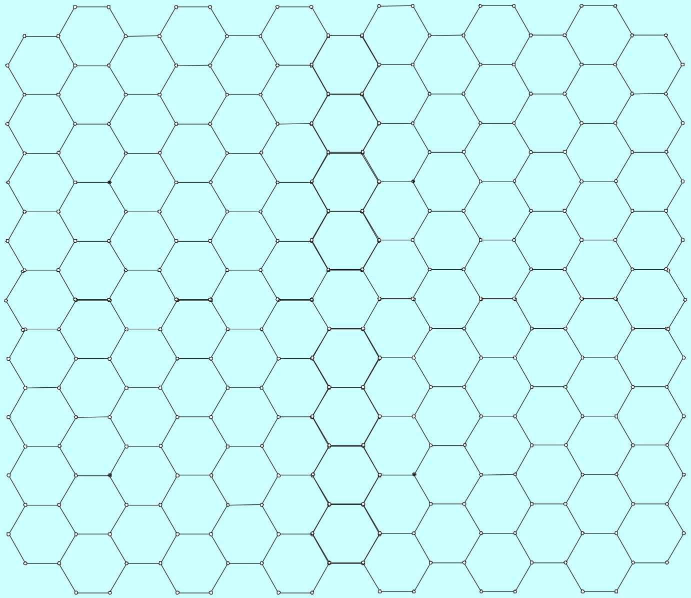
Graphene crystalline lattice

SWCNT *(m,n)*

Electromagnetic effects in nanotubes

Basic concept

Classical metallic grid screen



Graphene crystalline lattice

$$\varepsilon(p_z, s) = \pm\gamma_0 \left[1 + 4\cos\left(\frac{3bp_z}{2\hbar}\right)\cos\left(\frac{\pi s}{m}\right) + 4\cos^2\left(\frac{\pi s}{m}\right) \right]^{1/2}$$

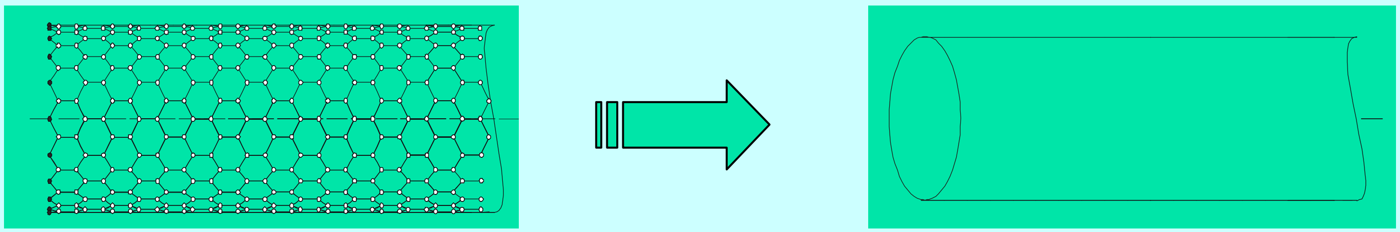
Electromagnetic effects in nanotubes

$\lambda >> b, \quad \lambda >> R_{cn}$

$b = 1.42\, A^0$

Effective boundary conditions:

universal tool for solving of ED problems in CNTs



$$\left(1+\frac{l_0}{k^2(1+i/\omega\tau)^2}\frac{\partial^2}{\partial z^2}\right)\left(H_\phi\Big|_{\rho=R+0}-H_\phi\Big|_{\rho=R-0}\right)=\frac{4\pi}{c}\sigma_{zz}E_z\Big|_{\rho=R},$$

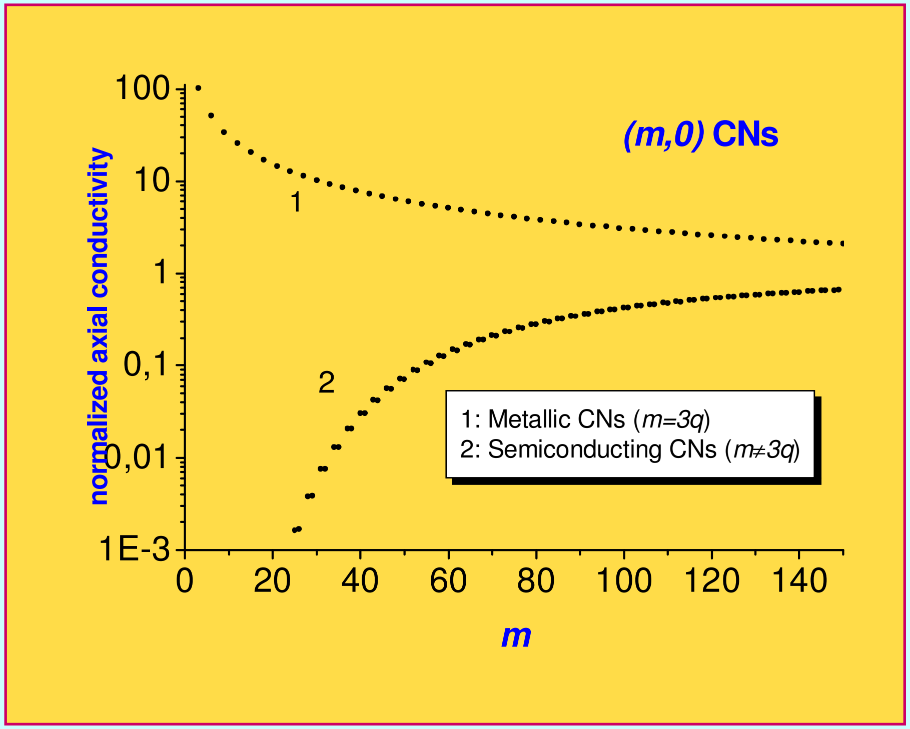
$$H_z\,|_{\rho=R-0} - H_z\,|_{\rho=R+0}=0,\;\; E_{z,\varphi}\,|_{\rho=R-0}-E_{z,\varphi}\,|_{\rho=R+0}=0$$

Spatial dispersion parameter $l_0 \sim 10^{-5}$ for metallic CNTs and not too large *m*

Electromagnetic effects in nanotubes

Dynamical conductivity of a single CNT

$$\sigma_{zz}(\omega,h) = -i\frac{e^2}{\sqrt{3}\pi\hbar m b}\sum_s \int_{-2\pi\hbar/3b}^{2\pi\hbar/3b} \frac{\partial F(p_z,s)}{\partial \mathcal{E}} \frac{\mathrm{v}_z^2(p_z,s)\quad dp_z}{\omega - h\,\mathrm{v}_z(p_z,s) + i/\tau}$$



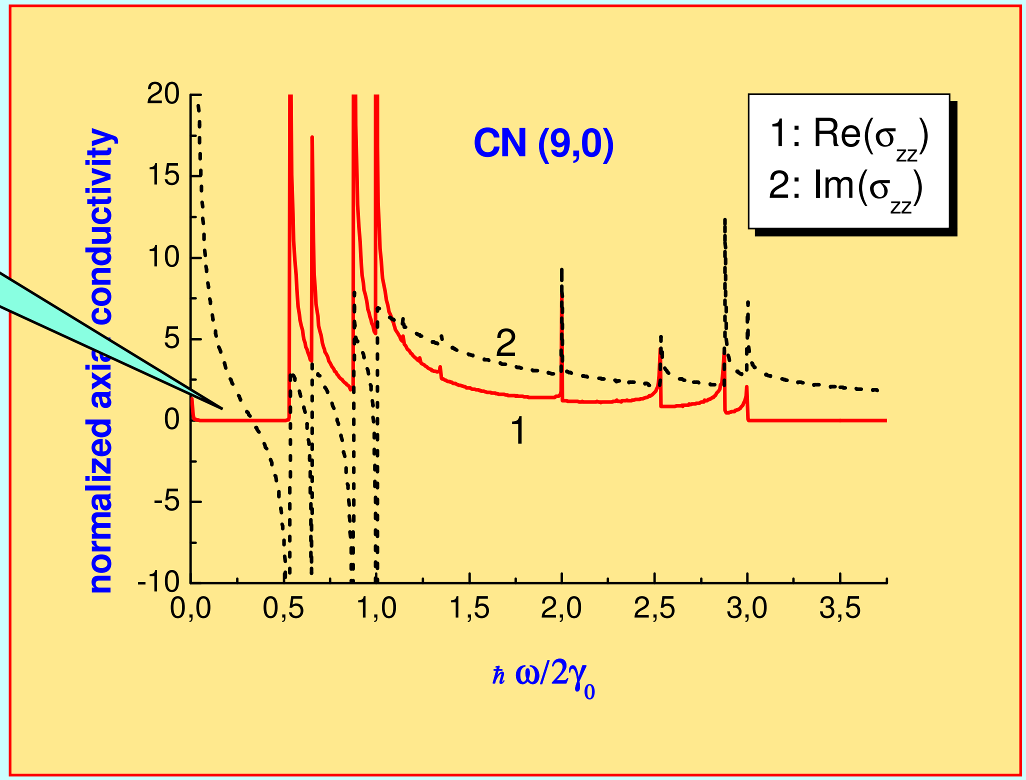
Phys. Rev. B 60, 17136, 1999

Electromagnetic effects in nanotubes

Dynamical conductivity of a single CNT:

the role of interband transitions

Semiclassical theory



Phys. Rev. B 60, 17136, 1999

Surface electromagnetic waves in CNT

Hertz potential

$$\mathbf{\Pi}_{\varepsilon} = A\mathbf{e}_z \begin{Bmatrix} I_q(\kappa\rho)K_q(\kappa R) \\ I_q(\kappa R)K_q(\kappa\rho) \end{Bmatrix} e^{ihz} e^{iq\phi} \qquad \kappa = \sqrt{h^2 - k^2}$$

Dispersion equation

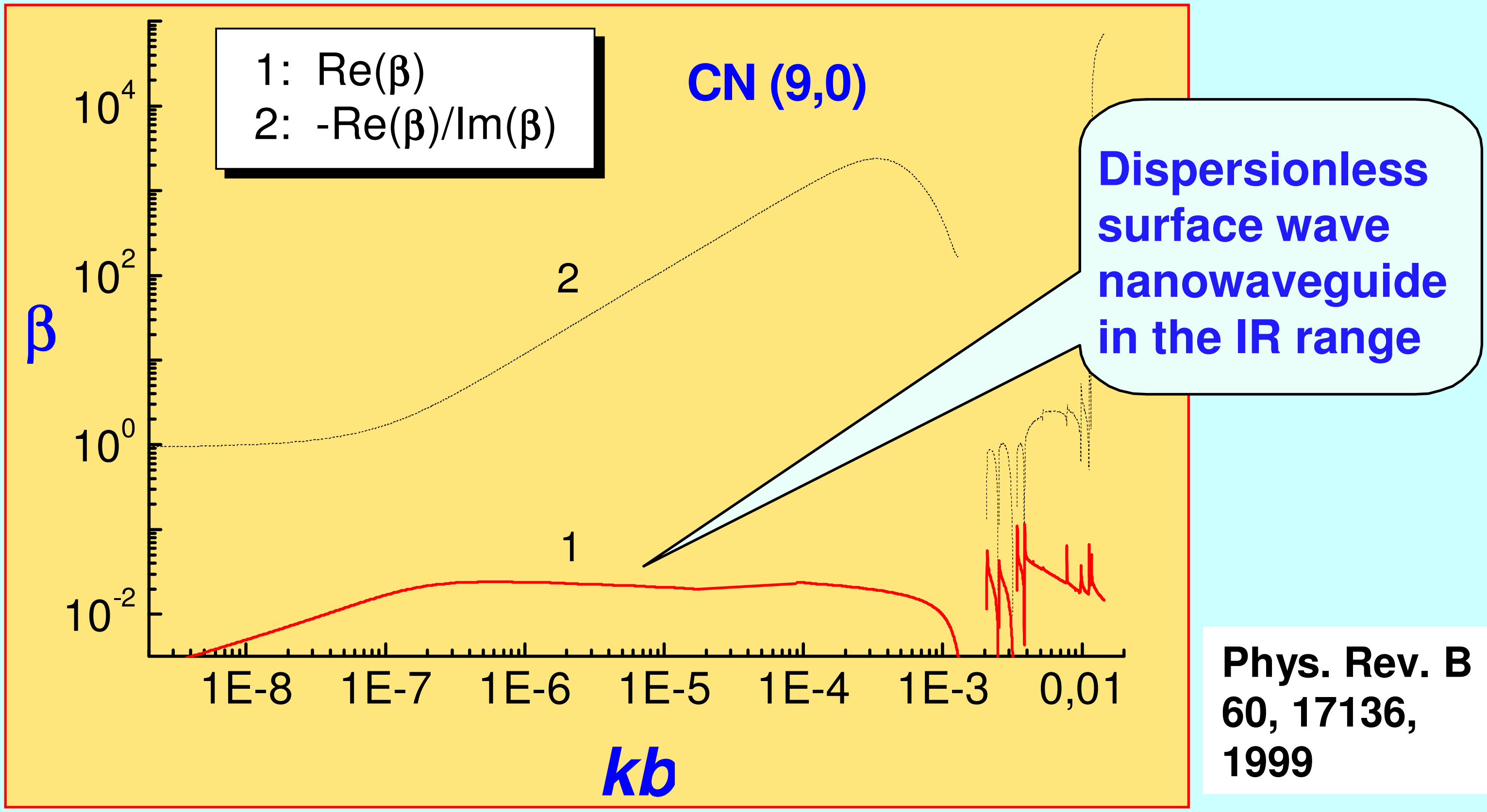
$$\left(\frac{\kappa}{k}\right)^2 I_q(\kappa R) K_q(\kappa R) = \frac{ic}{4\pi\kappa R_{cn}\sigma_{zz}} \left(1 - \frac{\kappa^2 + k^2}{(\omega + i/\tau)^2} c^2 l_0\right).$$

Slow-wave coefficient

$$\beta = \frac{k}{h} = \frac{k}{h' + ih''}$$

Electromagnetic effects in nanotubes

Complex-valued slow-wave coefficient β for a polar-symmetric surface wave



Phys. Rev. B 60, 17136, 1999

CNT is the optical delay-line: $1/\beta \sim 100$

Finite-length effects in CNTs

At optical frequencies, the CNT's cross-sectional radius and the length satisfy the following conditions:

$$kR_{cn} << 1, \quad kl_{cn} \sim 1, \quad k = \omega / c$$

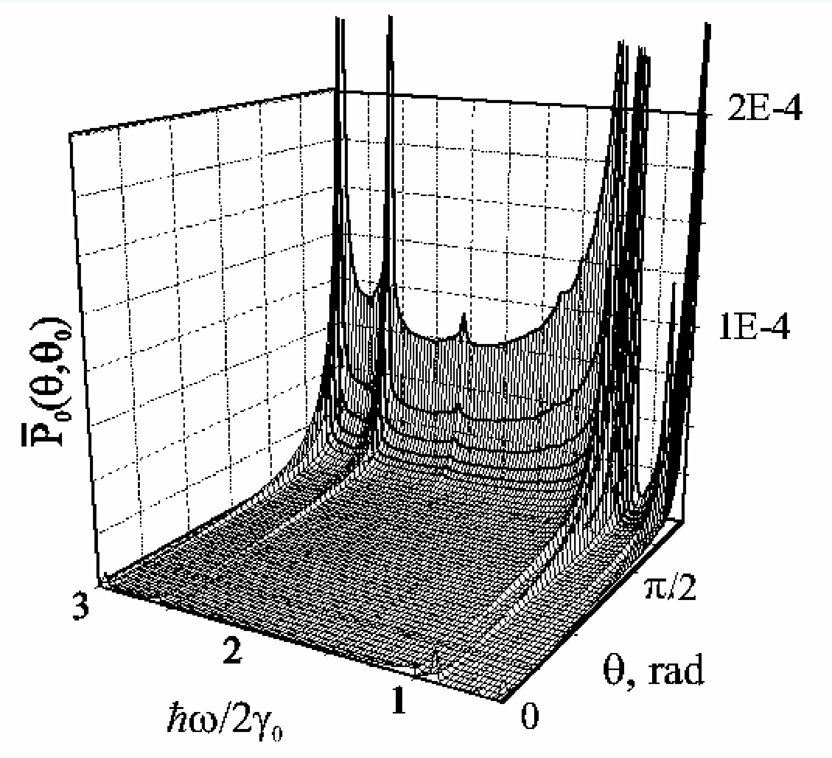
Clearly, although the cross-sectional radius is electrically small, the length is electrically large - conditions that are characteristic of wire antennas. Thus,

an isolated CNT is a wire nano-antenna at optical frequencies.

The key problem for the optical response of isolated CNTs and CNT arrays is the calculation of the scattering pattern of an isolated CNT of finite length.

Semi-infinite wire antenna

- analytical Wiener-Hopf technique is applied to a semi-infinite CNT;
- derivation of the finite-wire scattering amplitude using the edge-



Normalized density of the scattered power for the metallic (9, 0) CNT at frequencies of interband transitions

figures demonstrate sharp oscillations in the vicinity of optical resonances.

AEU Int. J. Electron. & Commun. 55, 273 (2001).

CNT as a travelling wave tube in the IR

- **Large retarding: $1/\beta > 100$**
- **Ballistic electron motion**

gain

$$g \sim \frac{k l_{cn}}{\beta} \left(\frac{\varpi}{c} l_{cn} \right)^2$$

V. Becker et al, Phys. Rev. A 25, 956 (1982)

Estimate:

l_{cn}= 30 mkm
V~ 10 V
I ~ 1 nA

$g \sim 0.3$

Nonlinearity: motivation

Pronounced nonlinearity of CNTs

X. Liu, *et al.*	Appl. Phys. Lett. 74, 164 (1999)
Y.-C. Chen, *et al.*	Appl. Phys. Lett. 81, 975 (2002)
C. Stanciu, *et al.*	Appl. Phys. Lett. 81, 4064 (2002)
J.-S. Lauret, *et al.*	Phys. Rev. Lett. 90, 057404 (2003)

Experimental observations

Semi-classical theory does not respond to crucial questions

Nonlinearity in the vicinity of optical resonances?

R.-H. Xie and J. Jiang,	Appl. Phys. Lett, 71, 1029 (1997).
V.A. Margulis, *et al.*	Physica B, 245, 173 (1998).
V.A. Margulis,	J. Phys.: Cond. Mat. 11, 3065 (1999)
G.Ya. Slepyan, *et al.*	Phys. Rev. A 61, 777 (1999).
O. E. Alon, *et al.*	Phys. Rev. Lett. 85, 5218, 2000.
G.Ya. Slepyan, *et al.*	Phys. Rev. A, 63, 053808 (2001).
S. M. O'Flaherty, *et al.*	J. Opt. Soc. Am. B 20, 49 (2003).

NONLINEAR EFFECTS: general approach

Schrödinger equation for electrons in the CNT lattice potential

$$i\hbar \frac{\partial \Psi}{\partial t} = -\frac{\hbar^2}{2m}\Delta\Psi + [W(\mathbf{r}) - e(\mathbf{E}\cdot\mathbf{r})]\Psi(\mathbf{p},\mathbf{r})$$

Bloch wave expansion

$$\Psi(\mathbf{p},\mathbf{r}) = \frac{1}{\sqrt{\hbar}} \sum_{l,\,\mathbf{p}} C_l(\mathbf{p})\, e^{i\mathbf{pr}/\hbar}\, u_l(\mathbf{r})$$

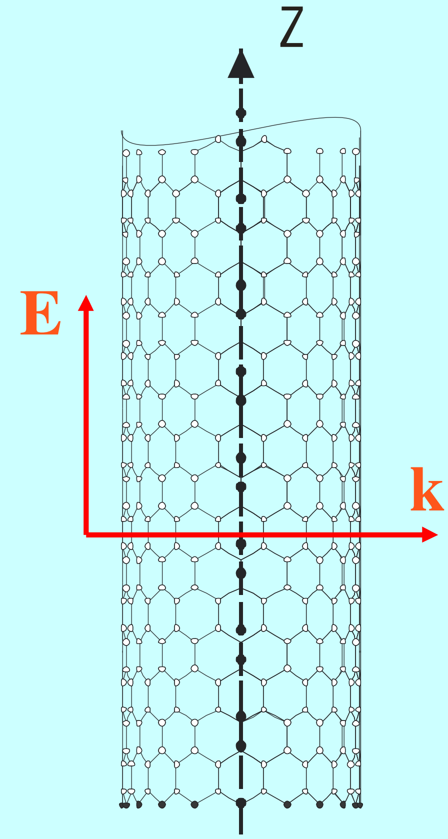
Standard representation of the density matrix

$$\rho_{ll'}(\mathbf{p}) = C_l(\mathbf{p}) C^*_{l'}(\mathbf{p})$$

Restrictions and approximations

- infinitely long rectilinear single-wall CN exposed to $\mathbf{E}(\mathbf{r},t) = \mathbf{e}_z E(x,t)$
- tight-binding approximation for π-electrons
- quantum-mechanical dispersion law
- CN radius is small compared with the driving field wavelength, $R_{cn} << \lambda_1$
- To avoid CNT damage by the driving field, its amplitude is accepted to be much less than the interatomic field strength:

$$|\mathbf{E}| << m^2 e^5/\hbar^4 = 5\text{x}10^9 \text{ V/cm}$$



Basic equation of nonlinear optics of CNTs

$$\frac{\partial\rho_{cc}}{\partial t}+eE_z\frac{\partial\rho_{cc}}{\partial p_z}=-\frac{i}{\hbar}eE_z(R_{cv}^{*}\rho_{cv}-R_{cv}\rho_{vc}),$$

$$\frac{\partial\rho_{cv}}{\partial t}+eE_z\frac{\partial\rho_{cv}}{\partial p_z}=-\frac{i}{\hbar}eE_z[R_{cv}(\rho_{vv}-\rho_{cc})-(R_{cc}-R_{vv})\rho_{cv}]-i\omega_{cv}\rho_{cv},$$

$$R_{ll'}=\frac{i\hbar}{2}\int_{\Omega}\left(u_l^{*}\frac{\partial u_{l'}}{\partial p_z}-\frac{\partial u_l^{*}}{\partial p_z}u_{l'}\right)d^3\mathbf{r}$$

Indices l and l' take the values v and c which correspond to valence and conduction bands

Explicit expressions for $R_{ll'}$ are available

Electromagnetic effects in nanotubes

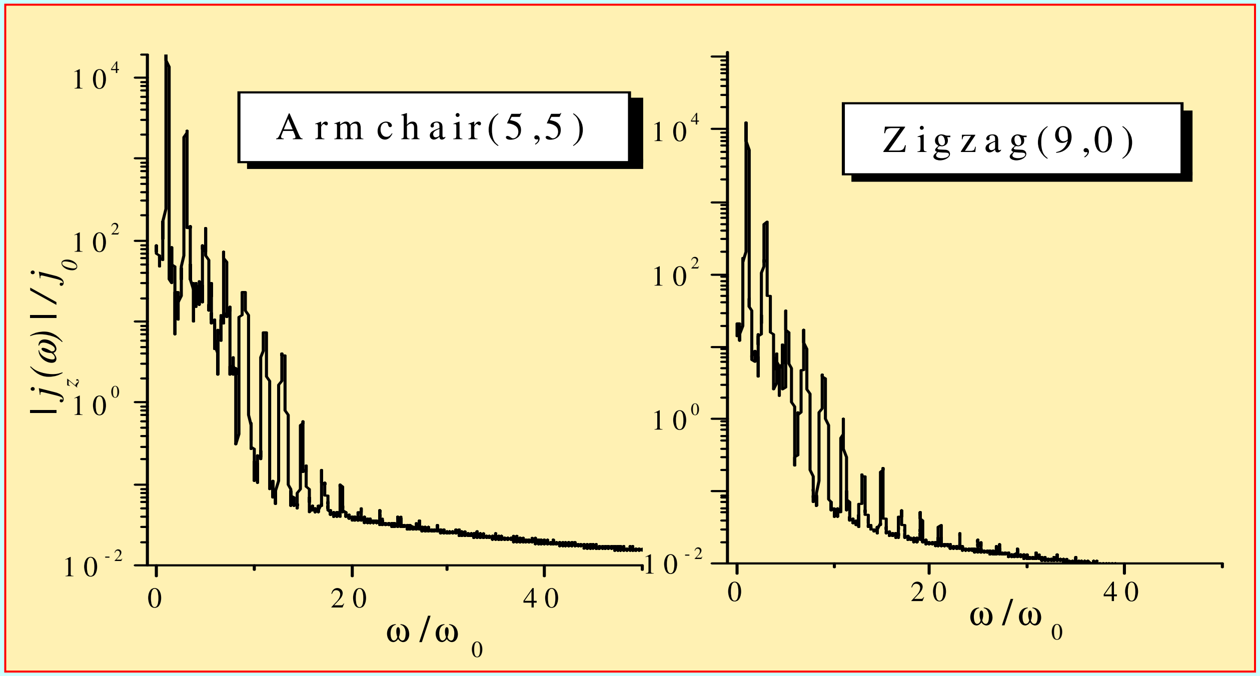
Axial current in CNT

$$j_z = j_z^{(1)} + j_z^{(2)},$$

Intraband transitions

Interband transitions

$$j_z^{(1)} = \frac{4e}{(2\pi\hbar)^2}\int \frac{\partial \mathcal{E}_c}{\partial p_z}\rho_{cc} d^2\mathbf{p}, \qquad j_z^{(2)} = \frac{8e}{(2\pi\hbar)^2\hbar}\int \mathcal{E}_c R_{cv}\,\mathrm{Im}(\rho_{cv}) d^2\mathbf{p}$$



Induced current spectrum of CNTs illuminated by a Ti:Sapphire laser pulse

Electromagnetic effects in nanotubes

Third-order harmonics

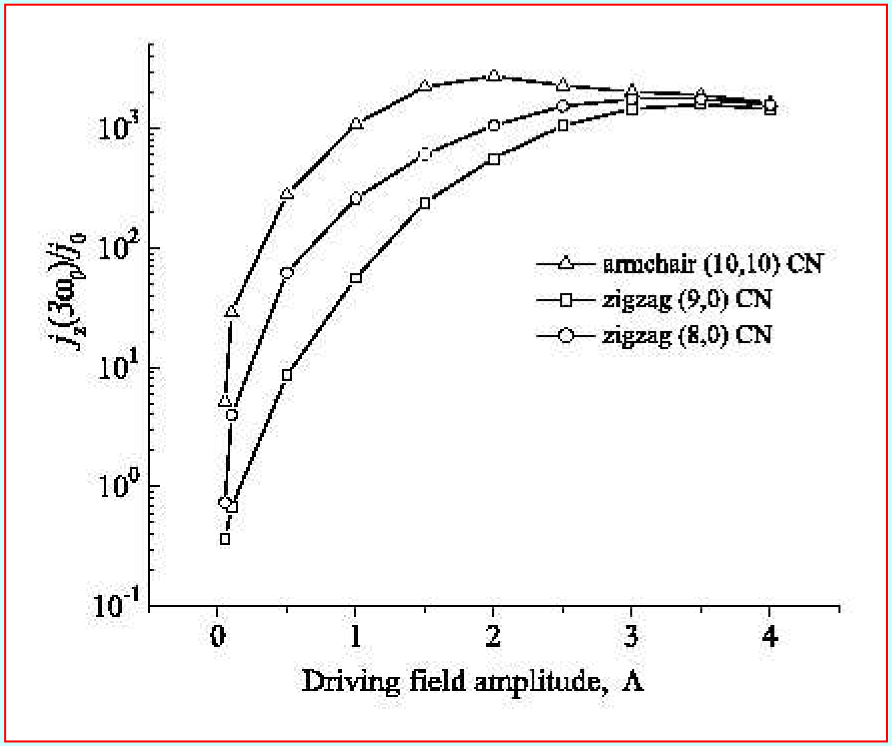


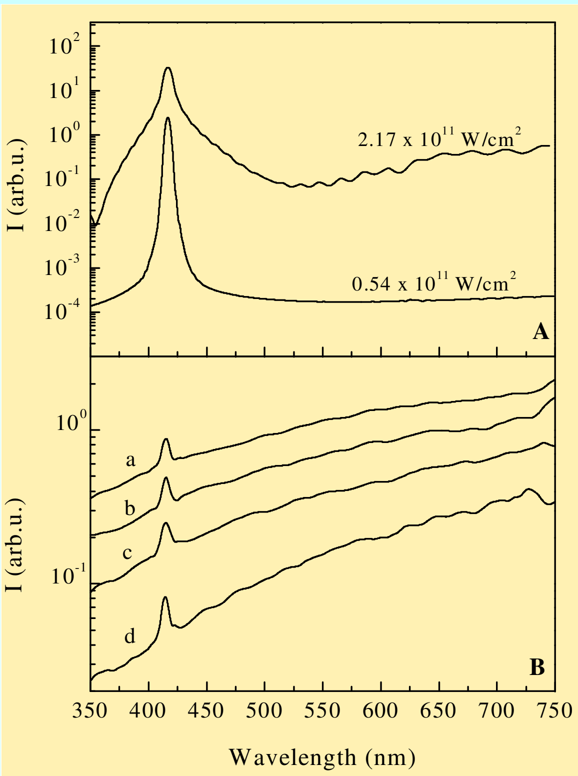
Figure:
Amplitude of the third harmonic current as a function of the driving field strength

$$j_z(N\omega) \sim E_0^p,$$
$$N \neq p$$

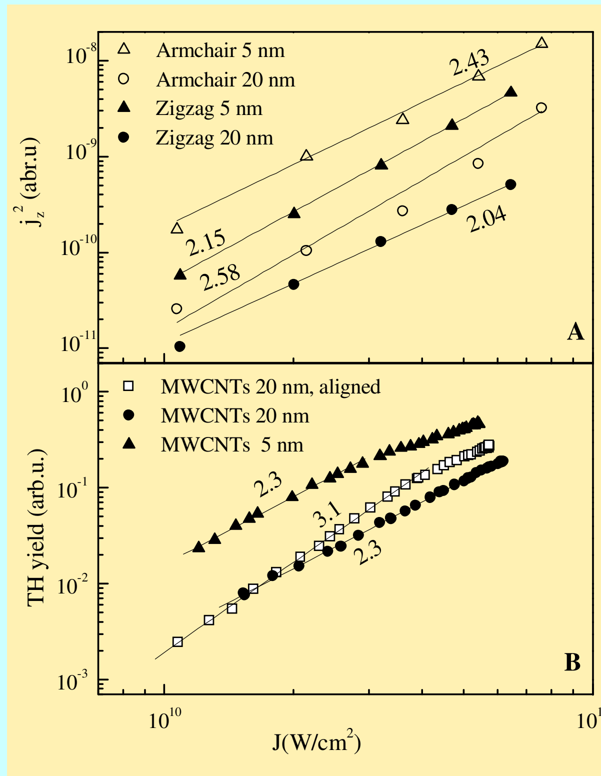
Picture demonstrates that the third-order current is not proportional to the third degree of the field amplitude: **high-order nonlinearities are of importance**

Electromagnetic effects in nanotubes

TH: Theory and experiment



Broad background and TH-signal;
(A) theory, (B) experiment



TH generation efficiency;
(A) theory, (B) experiment

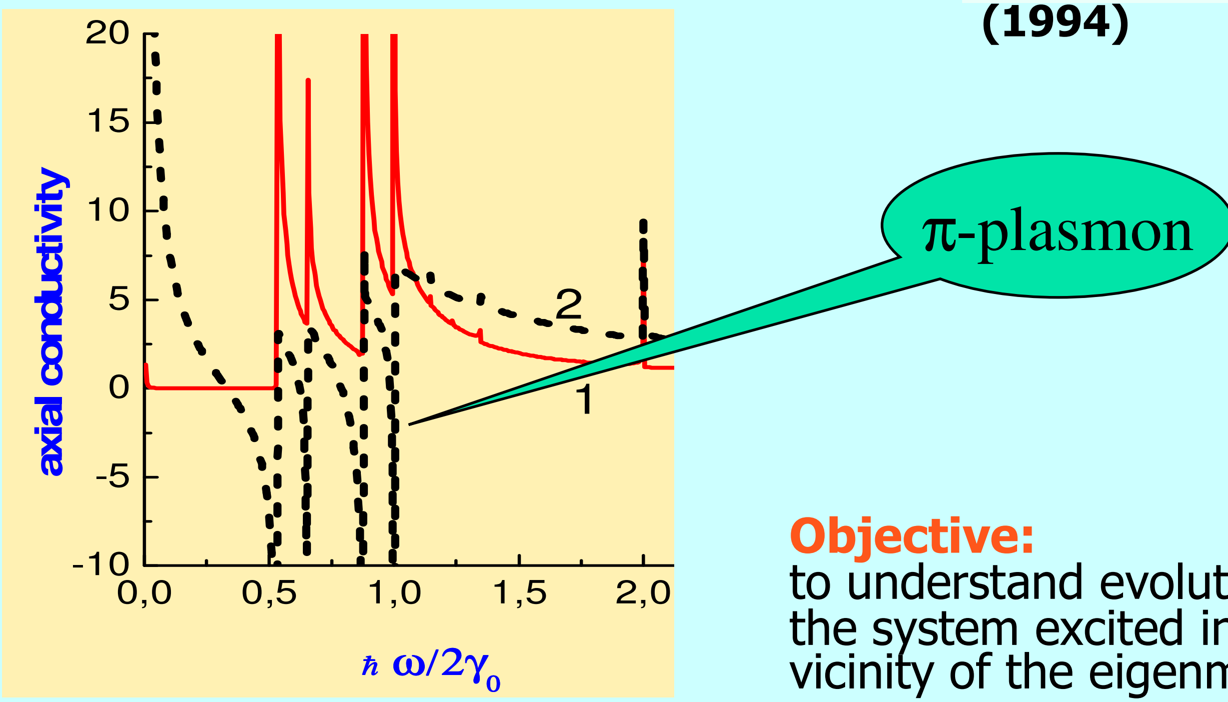
Experiment:
Max-Born Institute, Berlin, Germany
Chalmers University of Technology, Sweden

Appl. Phys. Lett. 81, 4064, 2002

Electromagnetic effects in nanotubes

Plasma resonance

M.F.Lin, K.W-K.Shung, Phys. Rev. B 50, 17774 (1994)



$$\hbar\omega_p = 2\gamma_0$$

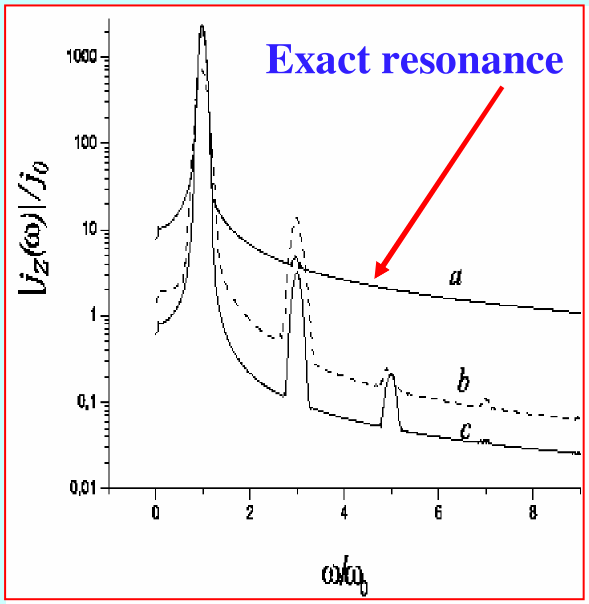
Objective:
to understand evolution of the system excited in the vicinity of the eigenmode.

Electromagnetic effects in nanotubes

Manifestations

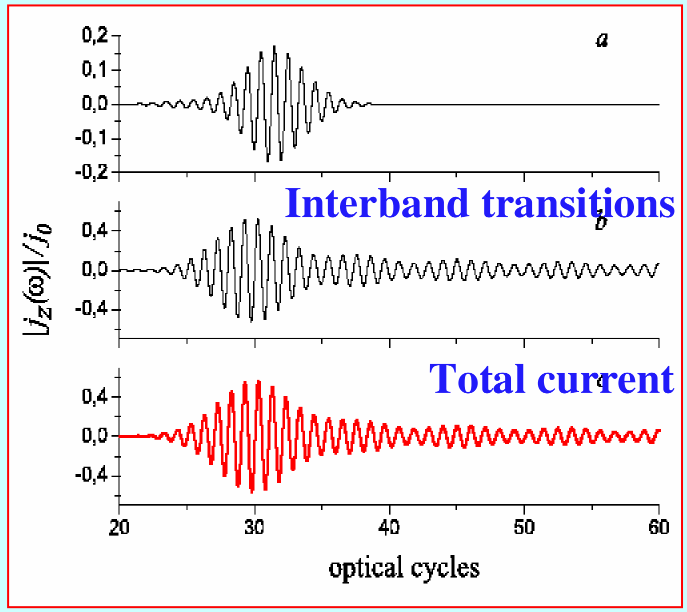
$$\hbar\omega_p = 2\gamma_0$$

Suppression of the HHG



HH spectra for different carrier frequencies. $w=w_p$ (a), $w=w_p$ /3(b), $w= 1.27\ w_p$ (c); $J=5x\ 10^{11}\ W/cm^2$.

Pulse deformation



Pulse evolution in (9,0) zigzag CNT at the plasma resonance

The behaviour of the current at the plasma resonance in a CNT is a typical behavior of any resonant system where eigenmodes can propagate after switching off the source: the current does not decay after the laser pulse is gone.

Electromagnetic effects in nanotubes

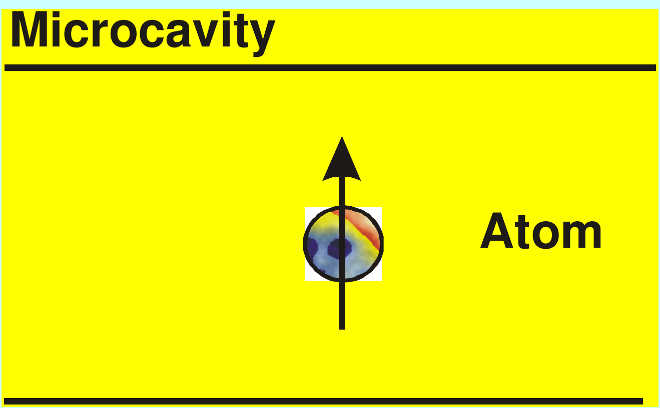
Purcell effect

Purcell effect: enhancement of the spontaneous decay rate of an atom located near media interface and/or optical nonhomogeneity

Realizations: microcavities, optical fibers, photonic crystals

Example: atom in a microcavity

$$\xi = \frac{\Gamma}{\Gamma_0} = \frac{6\pi Q}{k^3 V} \cong Q$$



$$\Gamma_0 = \frac{4}{3\hbar} k_A^3 |\mu|^2$$

$$Q \sim 10^4 - 10^8$$

Pioneering experiments

P.Goy et al. Phys. Rev. Lett. 50, 1903, 1983

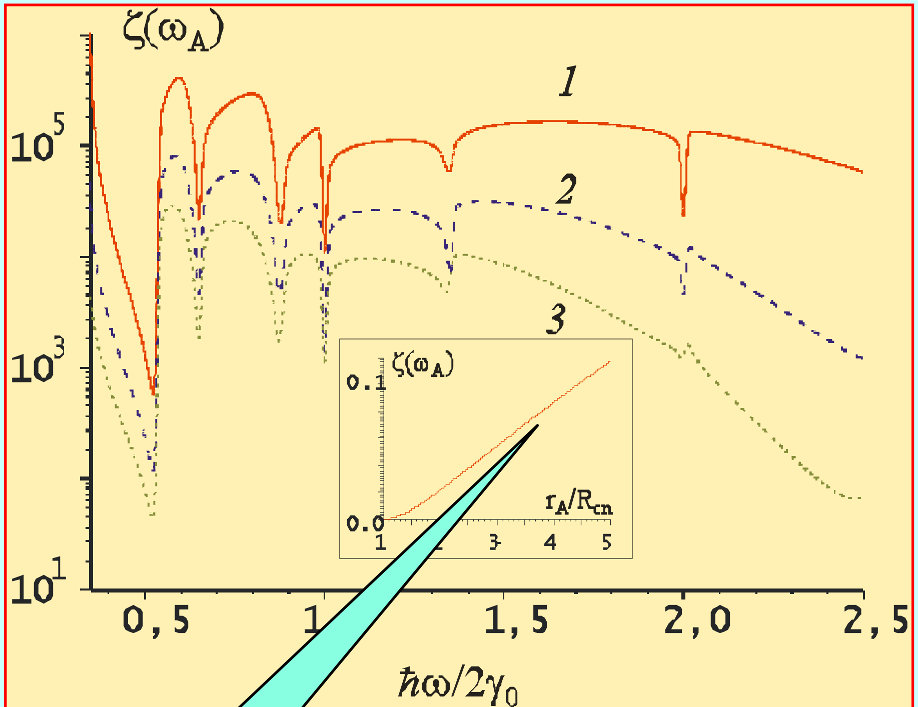
G.Gabrielse, et al. Phys. Rev. Lett. 55, 67, 1985

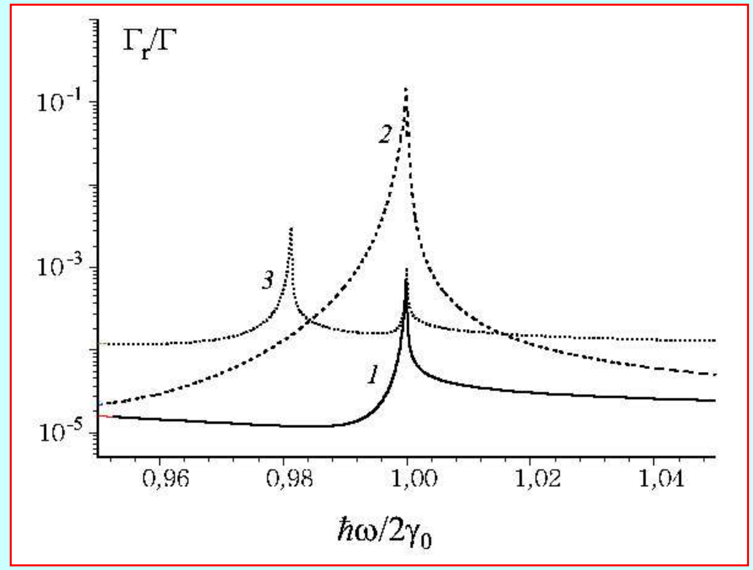
Electromagnetic effects in nanotubes

Decay rate ratio

Phys. Rev. Lett. 89, 115504, 2002

At different distances outside the (9,0) zigzag CNT





Contribution of the radiative channel

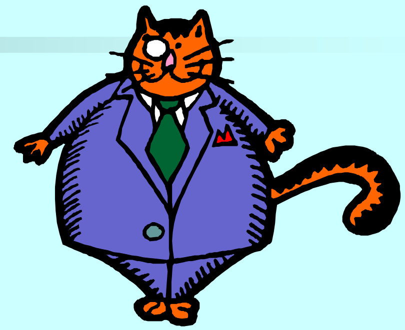
$\Gamma/\Gamma_0 \sim 10^5$

perfectly conducting cylinder

$$\xi(\omega_A) = \frac{\Gamma}{\Gamma_0} = 1 + \frac{3R_{cn}}{2\pi k_A^3} \operatorname{Im} \sum_{p=-\infty}^{\infty} \int_C \frac{\beta_A v_A^4 I_p^2(v_A R_{cn}) K_p^2(v_A r_A)\, dh}{1 + R_{cn}\beta_A v_A^2 I_p(v_A R_{cn}) K_p(v_A R_{cn})}$$

Electromagnetic effects in nanotubes

NEXT STEPS



- Antenna (finite-length) effects
- monomolecular travelling wave tube (ampl and genert)
- x-ray transportation and control
- CNT-based composites
  - •Finite length of a single CNT
  - •Interaction of electronic subsystems
- Evolution in CNT ensembles of femtosecond pulses
  - •To incorporate relaxation in the theory
  - •To develop homogenization procedure

Instabilities in CNTs

- Interaction with quantum states of light

good luck !

Electromagnetic effects in nanotubes

Acknowledgment: Collaboration in ED of nanostructures

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NanoModeling

(AM221) www.spie.org

SPIE's 49th Annual Meeting, 2-6 August 2004 Denver, USA

Chairs: **Akhlesh Lakhtakia**, The Pennsylvania State Univ.;
Sergey A. Maksimenko, Belarus State Univ.

Invited speakers:

M. C. Demirel (Biodetection and biomolecules), PennState

T. G. Mackay (Unusual metamaterials), Univ. of Edinburgh

T. S. Rahman (Atomistic modeling of thin films), Kansas Univ.

V. Shchukin (Semiconductor diode lasers in photonic bandgap crystals), Nanosemiconductor GmbH and Ioffe Institute

V. B. Shenoy (Nanomechanics), Indian Institute of Science, Bangalore