METALLIC NANOPARTICLE CHAINS (PLASMONIC CHAINS) AS OPTICAL WAVEGUIDES AND MORE

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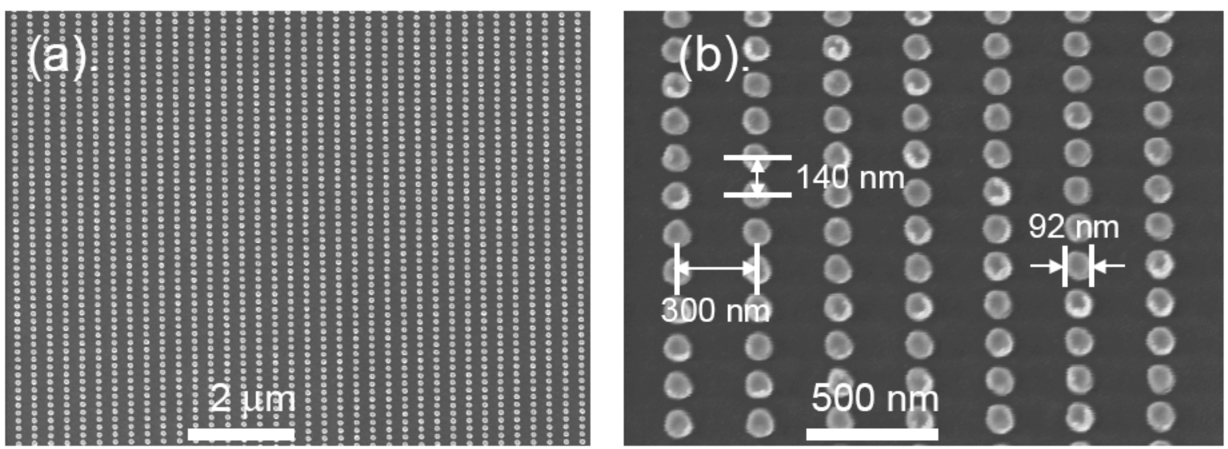
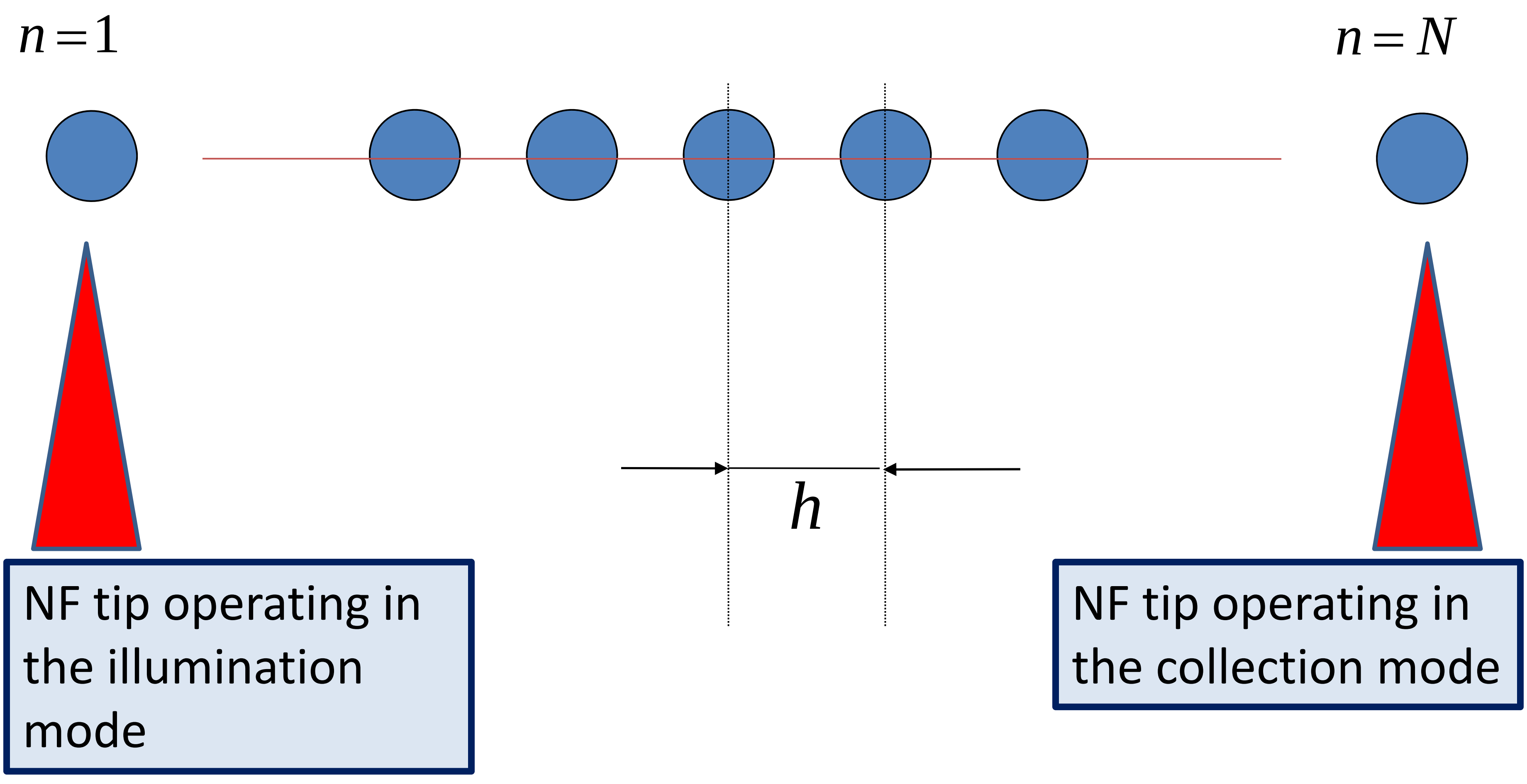
Why plasmonic chains are of interest?

- Spectroscopy and sensing
- Waveguiding and optical elements
- Advances in manufacturing



From K.B.Crozier, E.Togan, E.Simsek, T.Yang, Opt.Ex. 15, 17482 (2007)

Physical model (a waveguiding application)



A few easy choices

chains Partcles	Regular, linear, Infinite, quasistatic interactions	Substrate, host medium	More complex compositio, disorder	Far zone interaction, interference effects, localization	Non-receprical effects (e.g., in H-field)
Spheres	XXXXXXXXX	XXX	X	XXXXXX	X
More complicated shape	XXXXXXXXX	X	X	XX	
Dipole approx	XXXXXXXXX	XXXX	XX	X	
Beyond DA	XXX	X		X	
Quantum FS effects	Single nanoparticle				

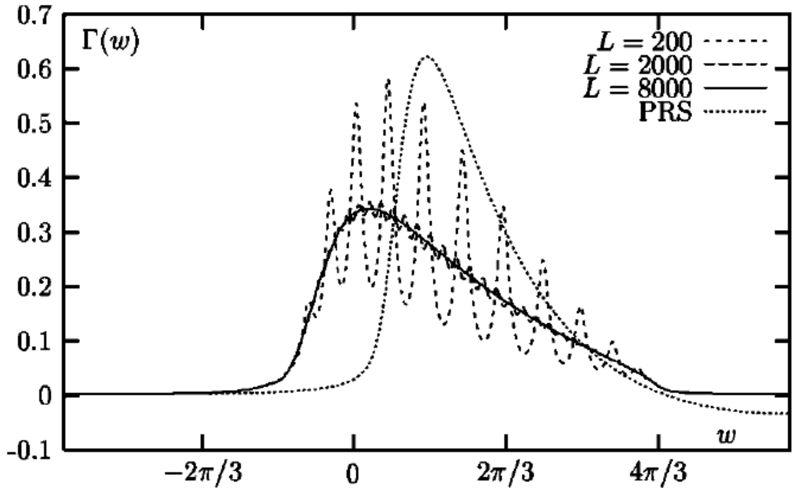
Is the dipole approximation adequate?

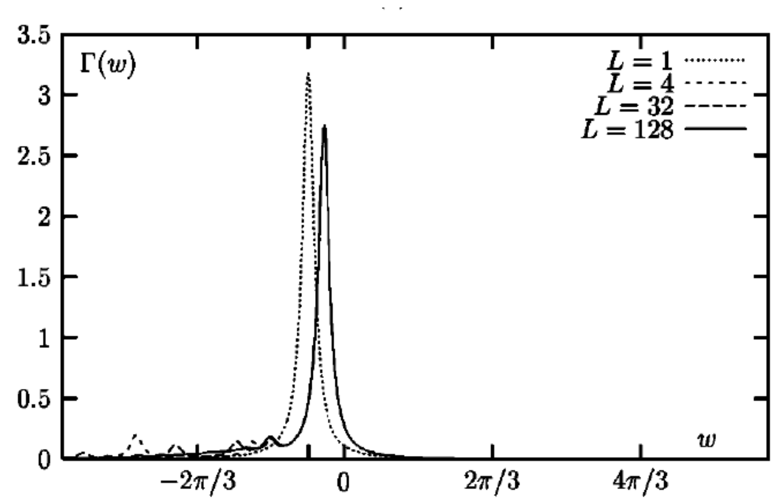
PRO:

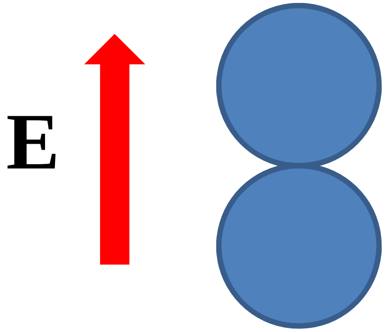
1. It's simple!
2. It allows physical insight.
3. Captures interference phenomena well (DA is not the same as quasistatics).
4. Allows to deal with complex shapes as long as the polarizability is known.
5. Is not that inaccurate for moderate separations, especially for TE polarization.
6. After all, the mathematical shapes we use are approximations, often rough.

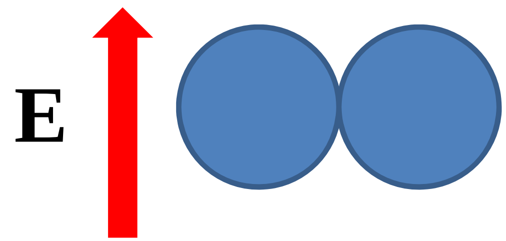
CONTRA:

DA does break when the particles are close to touching, especially for polarization the TM polarization







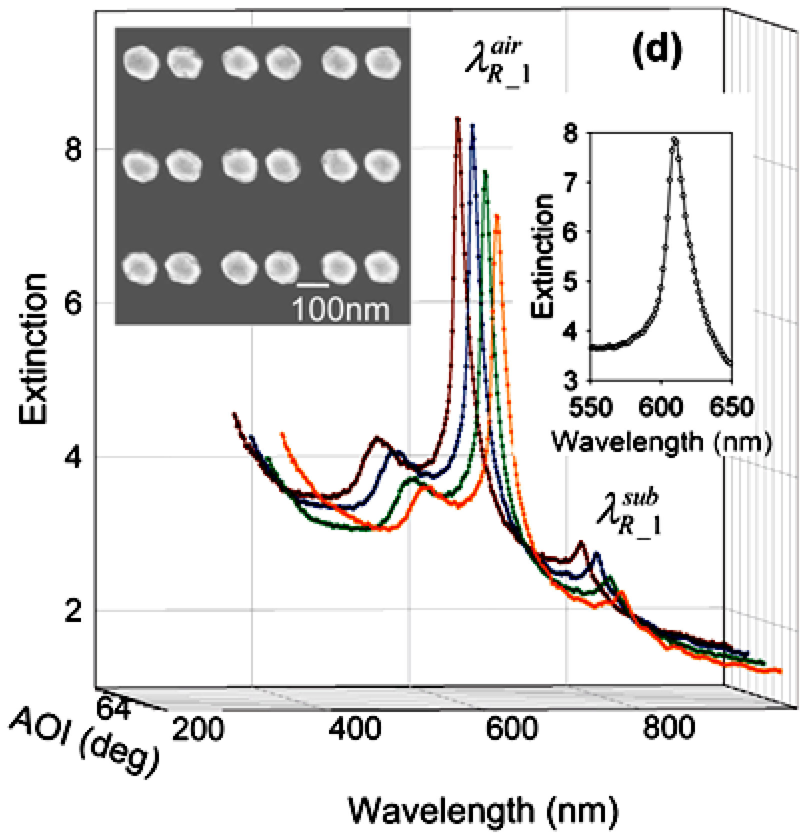


$$\sigma_e = 4\pi kV \,\mathrm{Im} \int \frac{\Gamma(w)dw}{z - w}$$

$$z = \frac{4\pi}{3}\frac{\varepsilon + 2}{\varepsilon - 1} \xrightarrow[\text{Drudean metals}]{} \frac{4\pi}{3}\frac{\omega_F^2 - \omega^2 - i\gamma\omega}{\omega_F^2}$$

From V.A.Markel, V.N.Pustovit, S.V.Karpov et al., PRB 70, 054202 (2004)

1. SPECTROSCOPY (NARROW NON-LORENTZIAN RESONANCES)



From V.G.Kravets, F.Schedin, A.N.Grigorenko, PRL 101, 087403 (2008)

The dipole approximation

$$\mathbf{d}_n = \alpha_n \left[\mathbf{E}_n + \sum_{m \neq n} \hat{G}(k; x_n, x_m) \mathbf{d}_\mathbf{m} \right]$$

The coupled-dipole equations in the frequency domain

$k = \dfrac{\omega}{c}$

$x_n = hn$

θ - angle of incidence

α_n - Polarizability of the *n*-th particle

$\hat{G}(k; x_n, x_m)$ - Frequency-domain Green's function

$$\mathbf{E}_n \propto \begin{cases} \delta_{n1} & \text{Near-field tip} \\ \exp\left(ik \cos\theta \cdot x_n\right) & \text{Plane wave} \end{cases}$$

* Incident plane wave
* Infinite, homogeneous periodic chain ($\alpha_n = \alpha$)

$$d_n = E_0 \frac{\exp\left(iqx_n\right)}{1/\alpha - h^{-3}S}$$

$$q = k\cos\theta = \frac{\omega}{c}\cos\theta$$

The propagation constant

$$S = h^3 \sum_{m \neq n} G(k; x_n, x_m)\exp(iqx_n)$$

The dipole sum ("self-energy")

$$S = S(k,q)$$

If $kh = \xi,\ qh = \eta,$ then

$$S_{\perp} = 2\xi^3 \sum_{n>0} \left[\frac{1}{\xi n} + \frac{i}{(\xi n)^2} - \frac{1}{(\xi n)^3} \right] \exp(in\xi)\cos(n\eta)$$

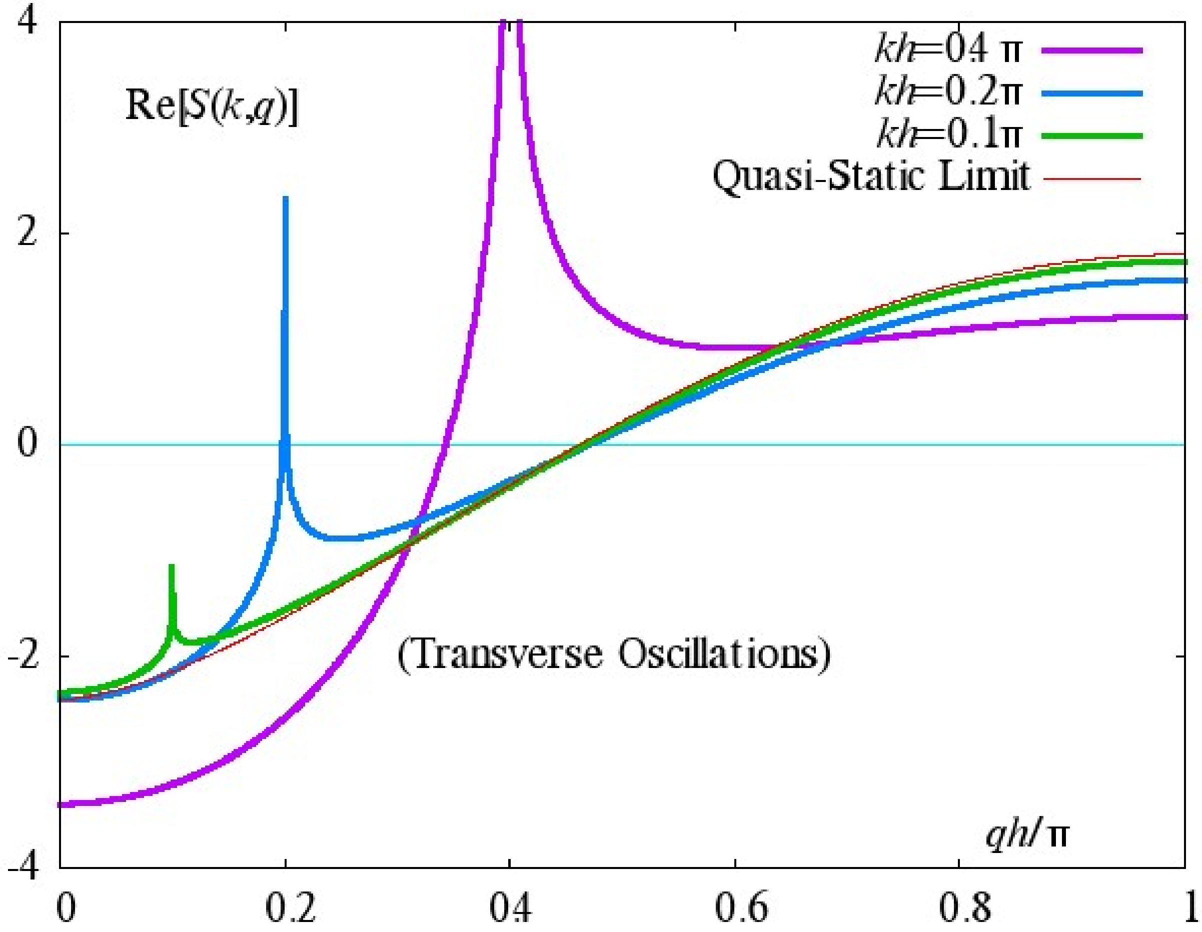
$$S_{\parallel} = 4\xi^3 \sum_{n>0} \left[-\frac{i}{(\xi n)^2} + \frac{1}{(\xi n)^3} \right] \exp(in\xi)\cos(n\eta)$$

The "$\perp$" dipole sum diverges logarithmically if

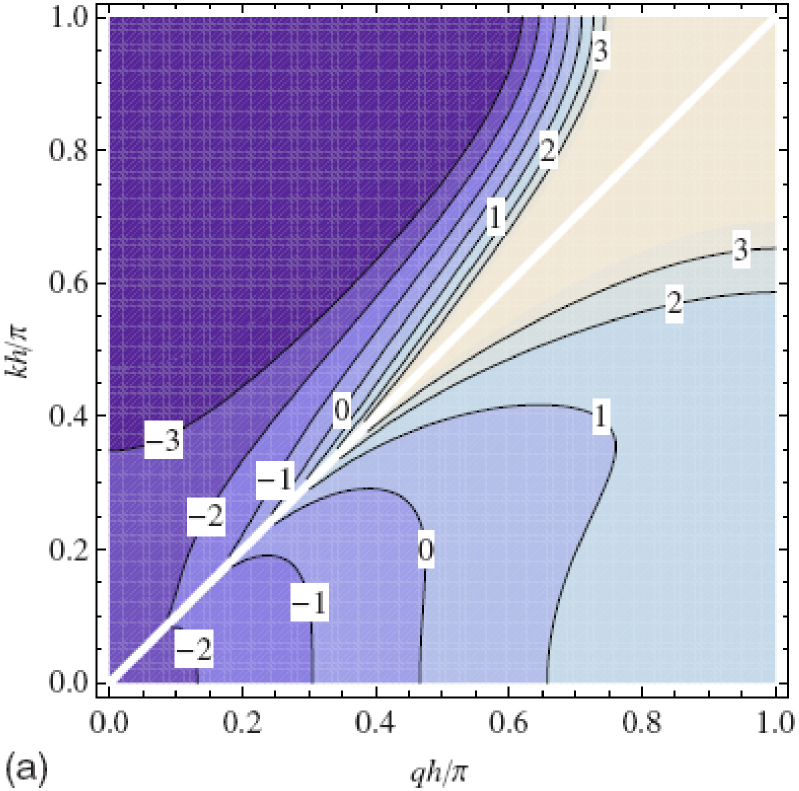
$$\xi \pm \eta = (k \pm q)h = 2\pi L$$

L being an integer

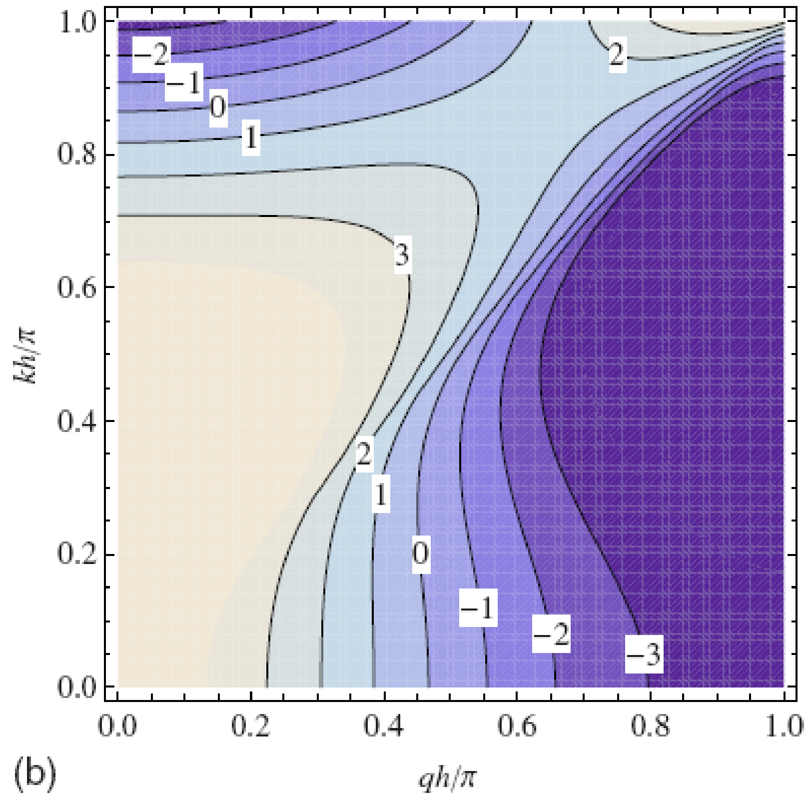
Re $S_{\perp}$ for some fixed values of frequency



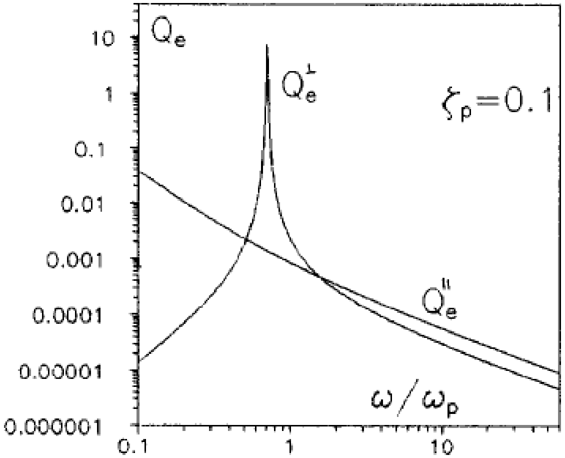
$\mathrm{Re}[S(kh, qh)]$



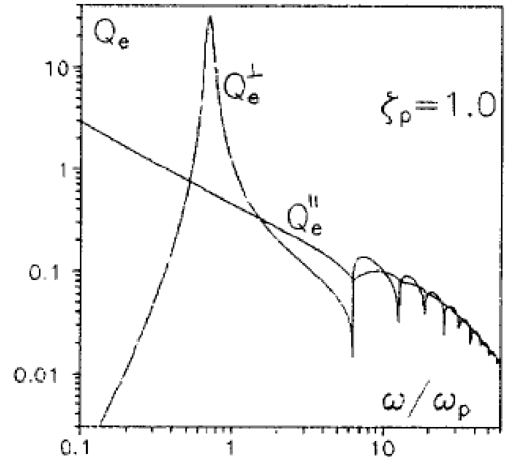
Orthogonal (TE) bpolarization



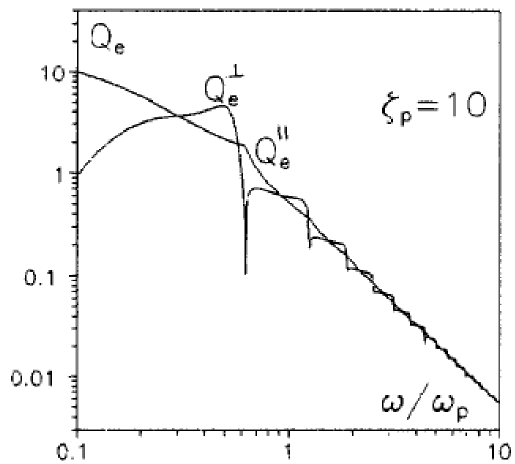
Parallel (TM) polarization



(a)



(b)



(c)

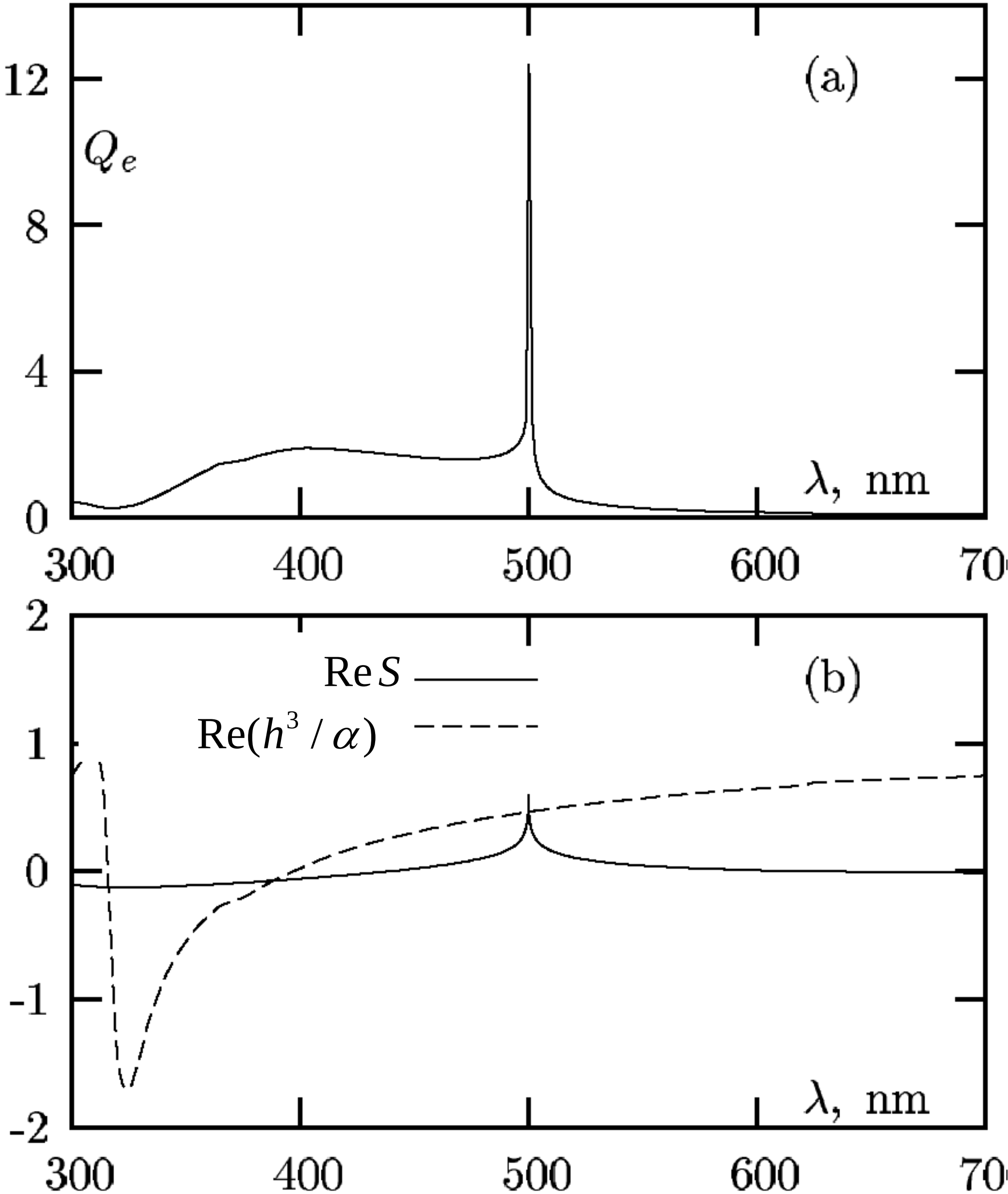
Narrow spectral features in extinction spectra of an infinite chain of Drudean spheres.

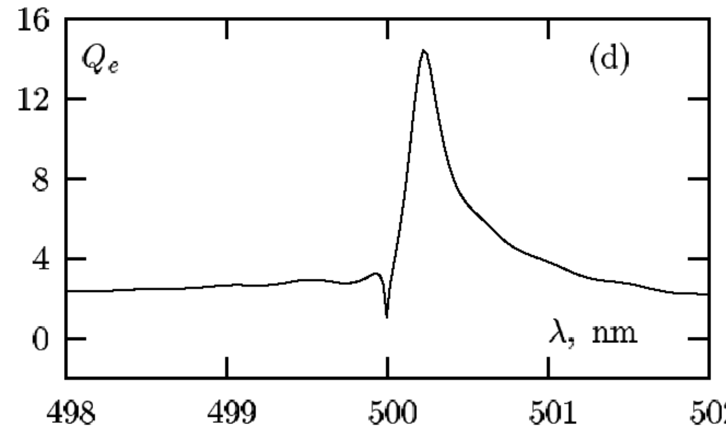
$\frac{\omega_P h}{c} = 0.1$ (a), $\frac{\omega_P h}{c} = 1$ (b), and $\frac{\omega_P h}{c} = 10$ (c).

" $\perp$ " - polarization orthogonal to the chain

" $\|$ "- polarization parallel to the chain

From V.A.Markel, J. Mod. Opt. 40, 2281 (1993).





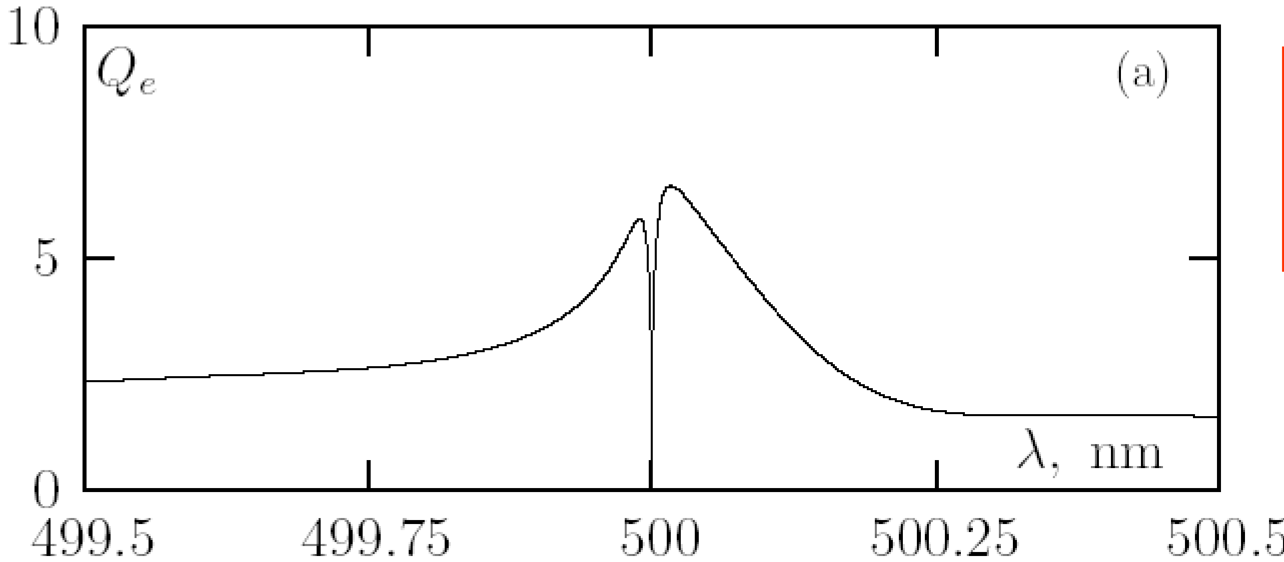
Experimental parameters for silver

$a = 50\text{nm}, \quad h = 500\text{nm}$

$$\Delta\lambda = \frac{h}{2\pi}\exp\left[-\frac{C}{2(2\pi)^2}\left(\frac{h}{a}\right)^3\right]$$

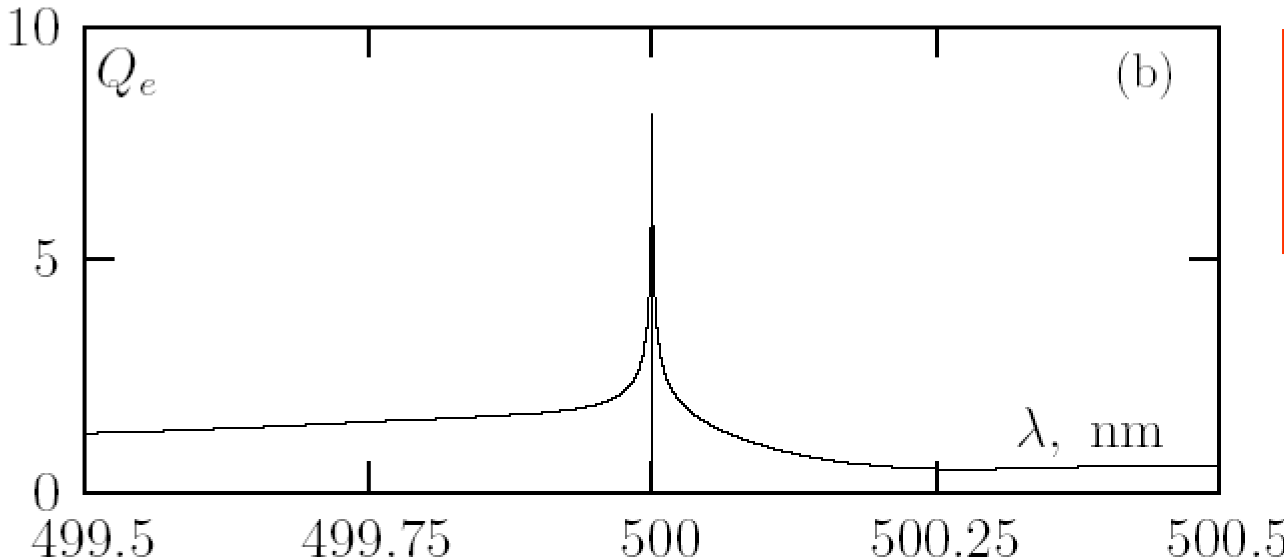
$C \sim 1$

From V.A.Markel, J.Phys.B 38, L347 (2005)



$a = 45\text{nm}$

$h = 500\text{nm}$



$a = 40\text{nm}$

$h = 500\text{nm}$

Why is the resonance non-Lorentzian ?

$$\Lambda(\omega) = \frac{f_n}{\omega - \omega_0 - \Sigma(\omega)}$$

$$\Sigma(\omega) \approx \Sigma(\omega_0) + \left.\frac{\partial \Sigma}{\partial \omega}\right|_{\omega=\omega_0} (\omega - \omega_0)$$

(Qasiparticle pole approximation)

But $\frac{\partial S}{\partial \omega}$ does not exist at $\omega = \omega_0$!

Sensitivity to local environment

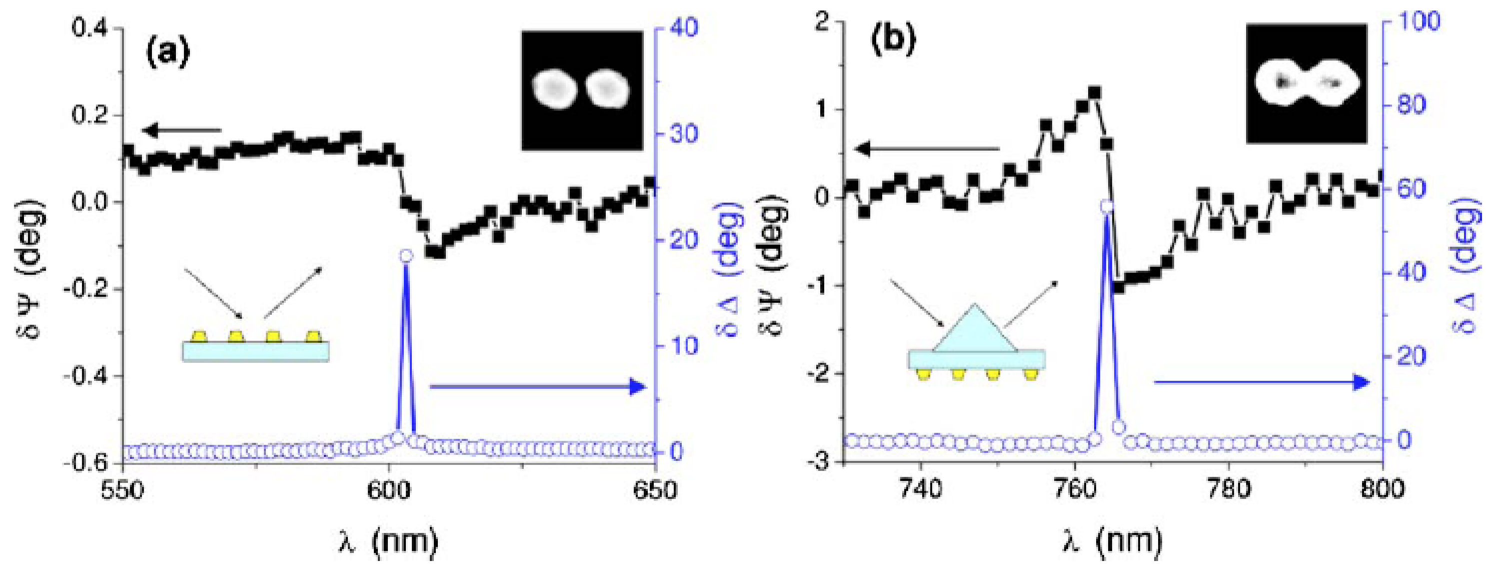


Fig. 2. (Color online) A typical CPR response to a change in local index of refraction. (a) The difference amplitude signal $\delta\Psi$ and phase signal $\delta\Delta$ for CPR of Figs. 1(c) and 1(d) registered for a change in gas RI $\Delta n = 4 \times 10^{-5}$. (b) $\delta\Psi$ and $\delta\Delta$ for CPR of Figs. 1(e) and 1(f) registered for a change in liquid RI $\Delta n = 6 \times 10^{-4}$.

From V.G.Kravets, F.Schedin, A.V.Kabashin A.N.Grigorenko, Opt.Lett. 35, 956 (2010)

2. DISPERSION RELATIONS

$$h^3 / \alpha(k) - S(k,q) = 0 \quad \Rightarrow \quad \omega = f(q) \qquad \text{[Recall that } k = \omega / c\text{]}$$

Is it possible to find a solution such that $k, q \in \mathbb{R}$?

Only if: (i) $q > k$

(ii) $\mathrm{Im}(1/\alpha) = -2k^3 / 3$

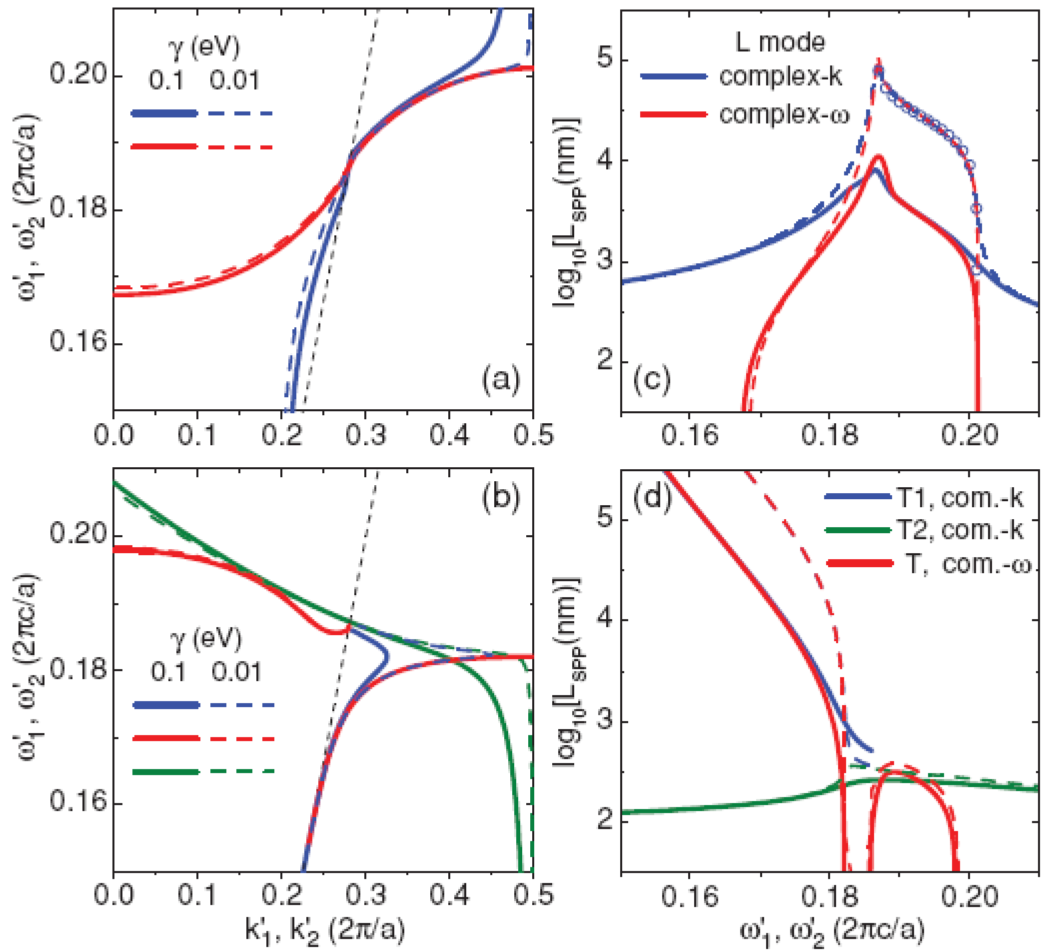
Modes with $q > k$ are called Surface Plasmon Polaritons (SPP)

What is the role of losses (both radiative and absorptive)?

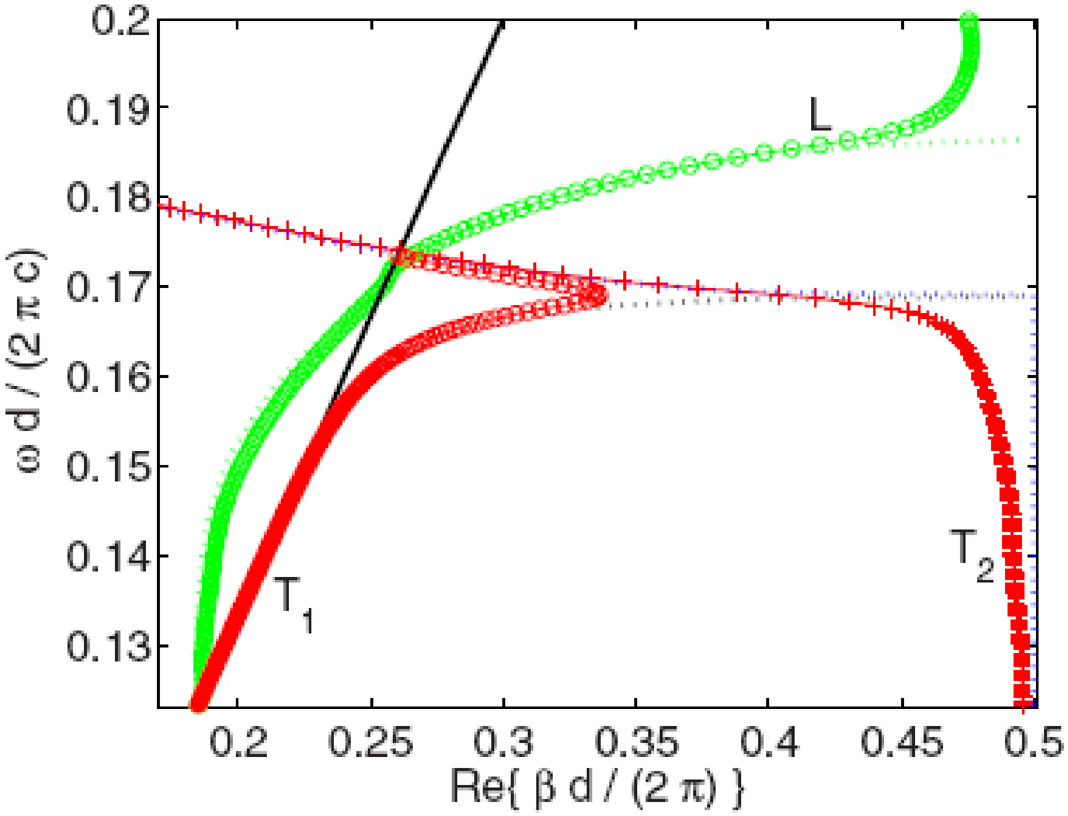
A few possible approaches:

* Take real ω and seek complex q
* Take real q and seek complex ω
* Solve eqn $\mathrm{Re}\left[h^3/\alpha - S(k,q)\right] = 0$ for $k, q \in \mathbb{R}$

When losses are small, there is not much of a difference...



From I.B. Udagedara, I.D.Rukhlenko, M. Premaratne, PRB 83, 115451 (2011)



From M.Conforti M.Guasoni, JOSA B 27, 1576 (2010)

What is the role of particle nonsphericity?

- Nanoparticle chains have been studied almost exclusively for the case of spherical particles
- However, nonsphericity can be expected to provide a useful additional parameter to control:
  - SPP dispersion curves
  - SPP bandwidth
  - Propagation distance

Model for the polarizability, α

$$\frac{1}{\alpha} = \frac{4\pi}{\varepsilon_h V}\left(\nu + \frac{\varepsilon_h}{\varepsilon_m - \varepsilon_h}\right) - i\frac{2k^3}{3}$$

ε_h is the permittivity of the host medium (a transparent dielectric or vacuum)

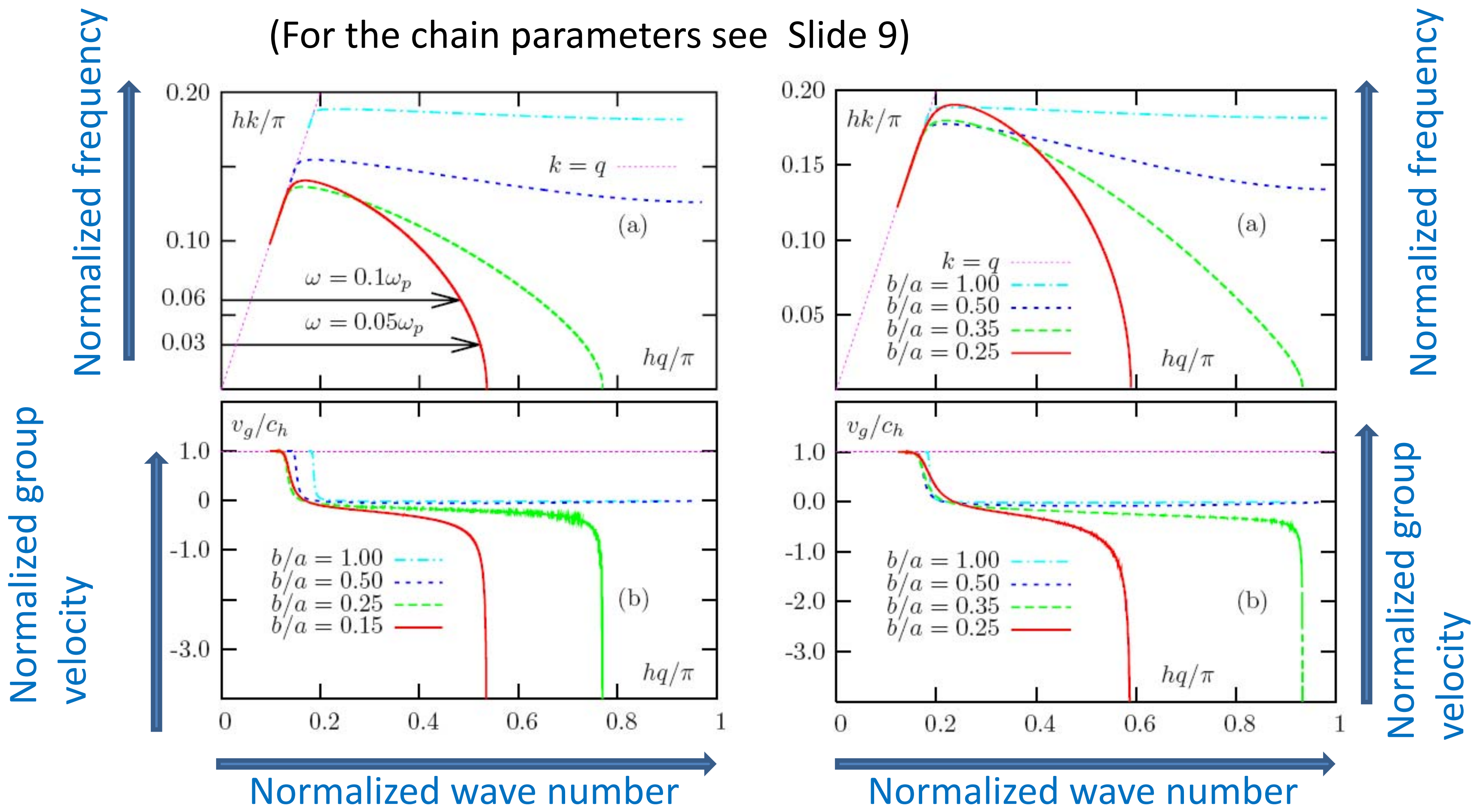
$\varepsilon_m = \varepsilon_0 - \dfrac{\omega_p^2}{\omega(\omega + i\gamma)}$ is the permittivity of metal (given by the Drude formula)

$V = \dfrac{4\pi abc}{3}$ is the volume of spheroid; $k = \dfrac{\omega}{c}$

ν is the depolarization factor

Dispersion curves and group velocities for transversely polarized SPPs and different aspect ratios *a/b* of spheroids

(For the chain parameters see Slide 9)

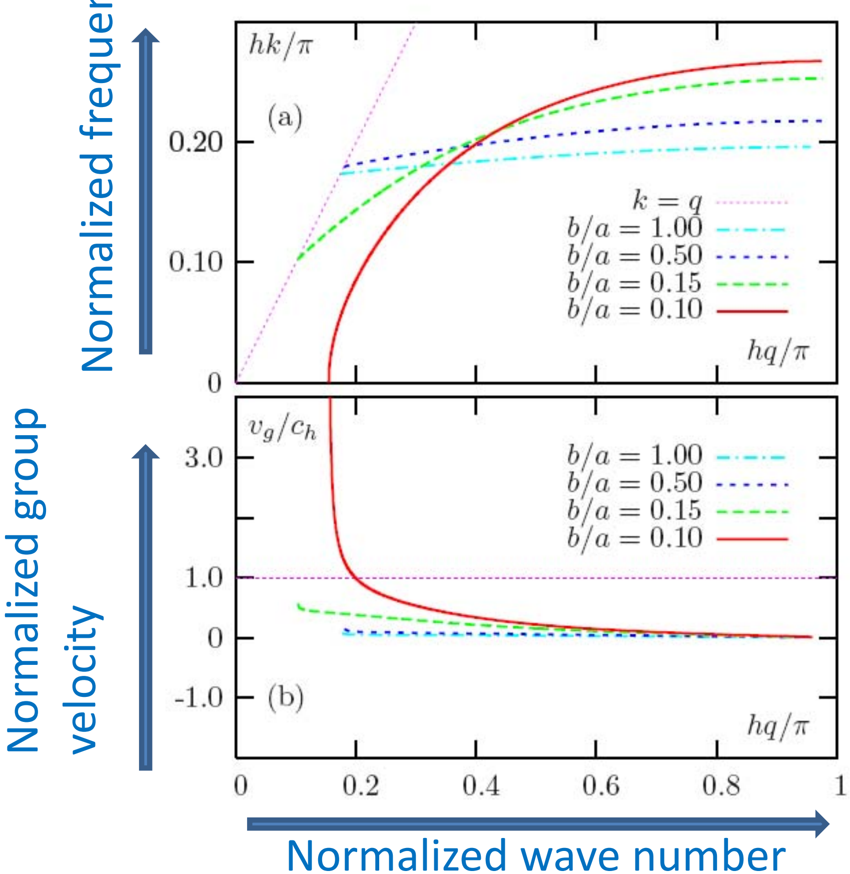


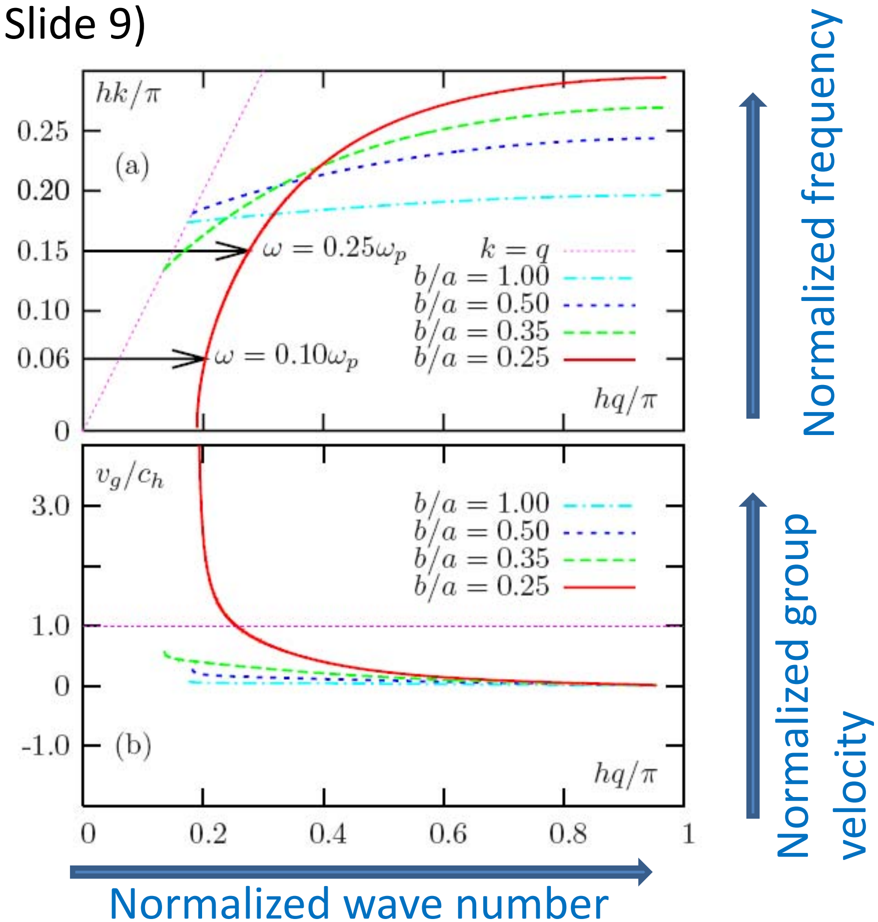
Prolate spheroids whose axis of symmetry is perpendicular to the chain

Oblate spheroids whose axis of symmetry is parallel to the chain

Dispersion curves and group velocities for longitudinally polarized SPPs and different aspect ratios *a/b* of spheroids

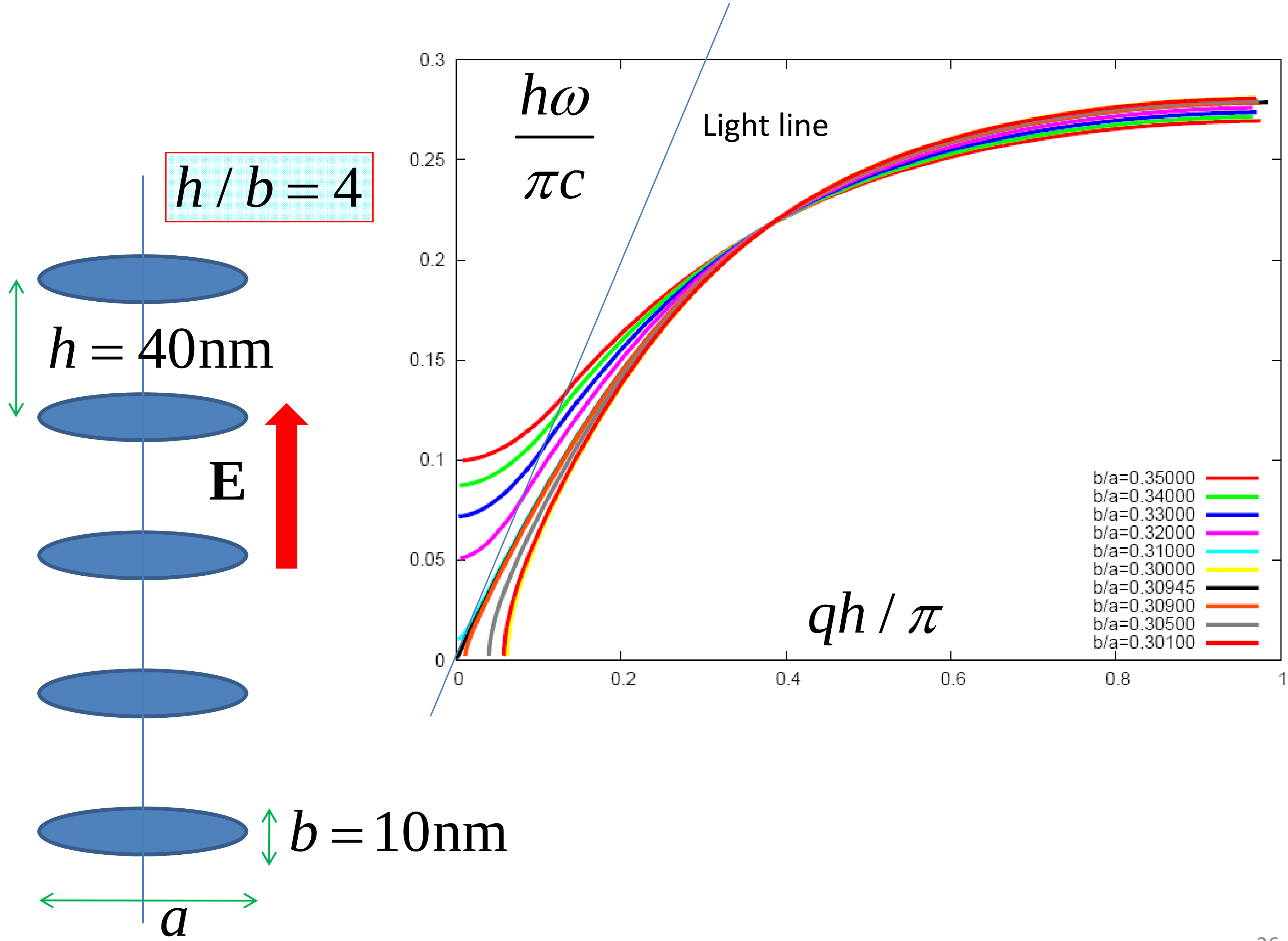
(For the chain parameters see Slide 9)



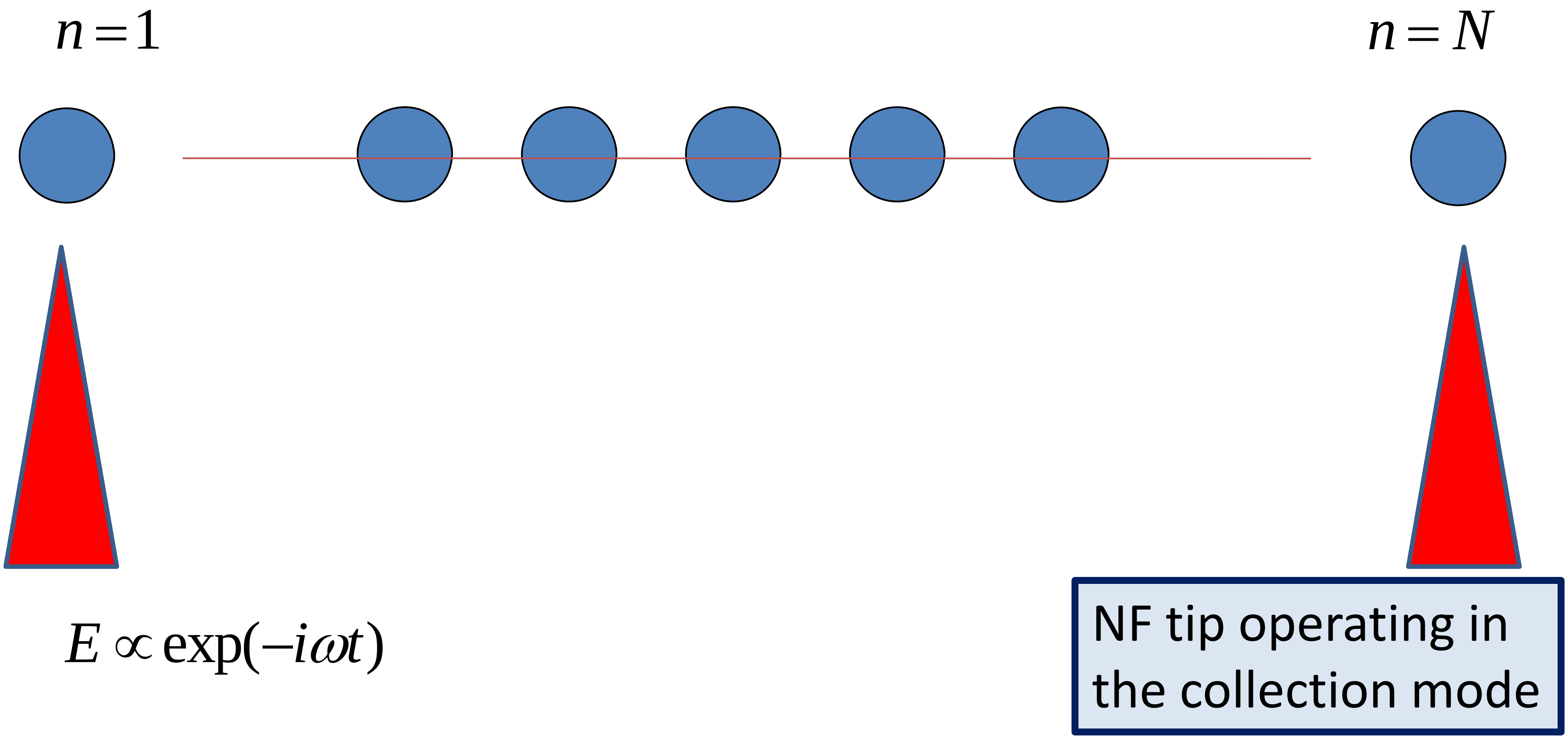


Prolate spheroids whose axis of symmetry is perpendicular to the chain

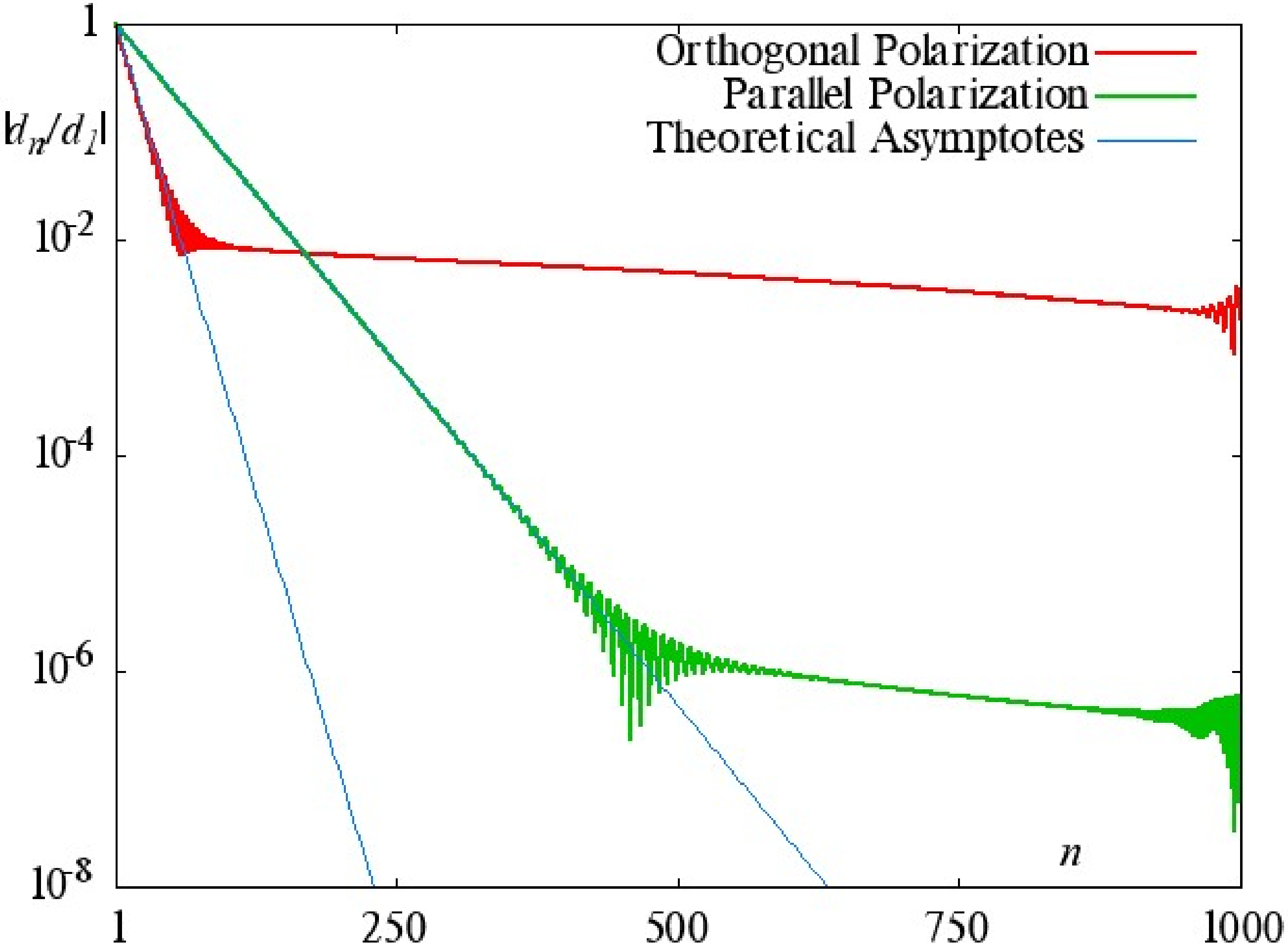
Oblate spheroids whose axis of symmetry is parallel to the chain


$h/b=4$
$h=40\text{nm}$
E
$b=10\text{nm}$
a
$\frac{h\omega}{\pi c}$
Light line
qh/π
0.3
0.25
0.2
0.15
0.1
0.05
0
0
0.2
0.4
0.6
0.8
1
b/a=0.35000
b/a=0.34000
b/a=0.33000
b/a=0.32000
b/a=0.31000
b/a=0.30000
b/a=0.30945
b/a=0.30900
b/a=0.30500
b/a=0.30100

3. PROPAGATION. STEADY STATE



Simulation for a finite chain of *N*=1000 identical nanospheres



Parameters:

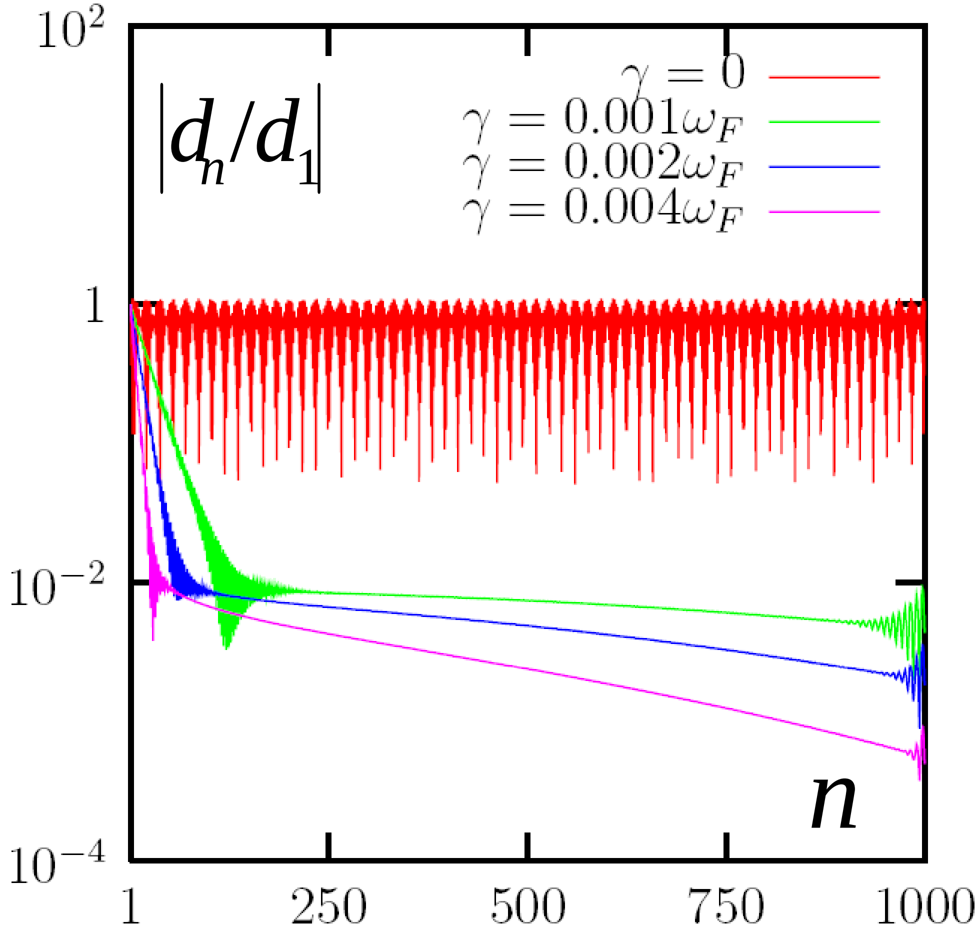
$\omega = \omega_F$

$\frac{\gamma}{\omega_F} = 0.002$

$\lambda = \frac{2\pi c}{\omega} = 10h$

$h = 4a$

Effect of Ohmic Losses



Parameters:

$\omega = \omega_F$

$\frac{\gamma}{\omega_F}$ varies

$\lambda = \frac{2\pi c}{\omega} = 10h$

$h = 4a$

Effects of disorder

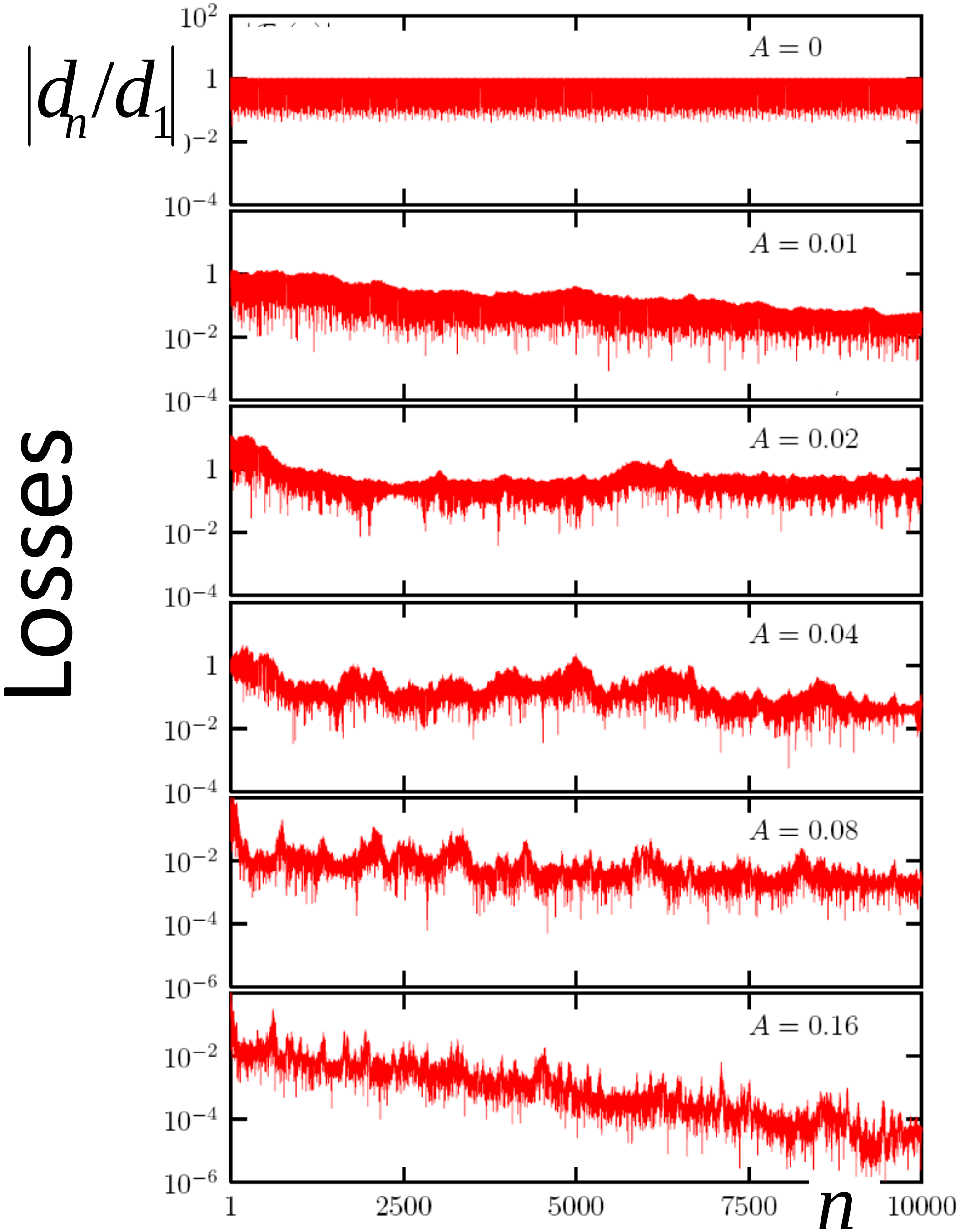
- Off-diagonal disorder (disorder in the nanoparticle positions)

  We assume here that the position of the *n*-th particle is evenly distributed in the interval $[h(n-A), h(n+A)]$, $A \ll 1$

- Diagonal disorder

  [A more subtle effect, not considered in this talk; see Phys.Rev.B **75**, 085426 (2007)]

Off-Diagonal Disorder in the Absence of Ohmic Losses



Parameters:

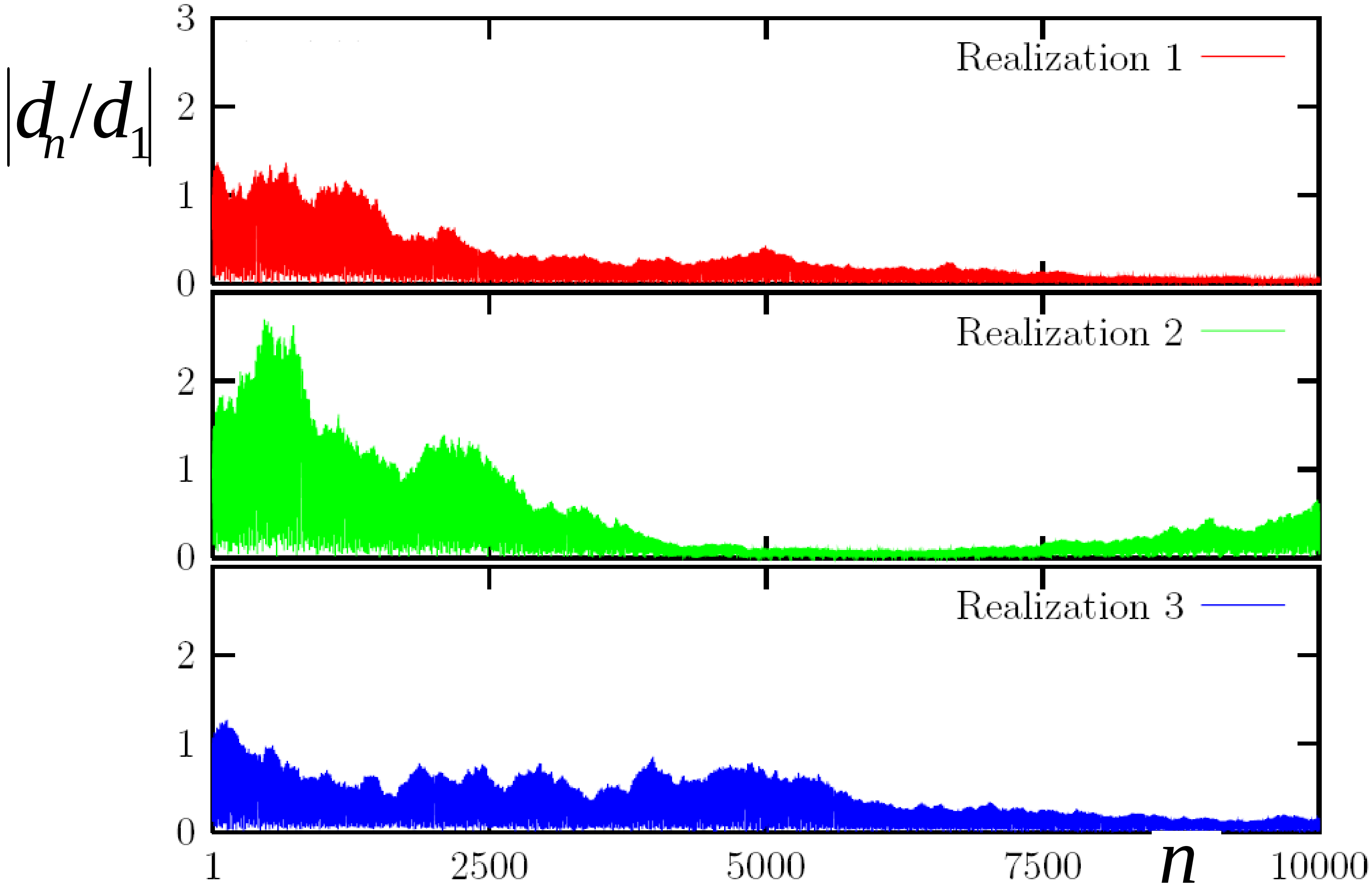
$\omega = \omega_F$

$\frac{\gamma}{\omega_F} = 0$

$\lambda = \frac{2\pi c}{\omega} = 10h$

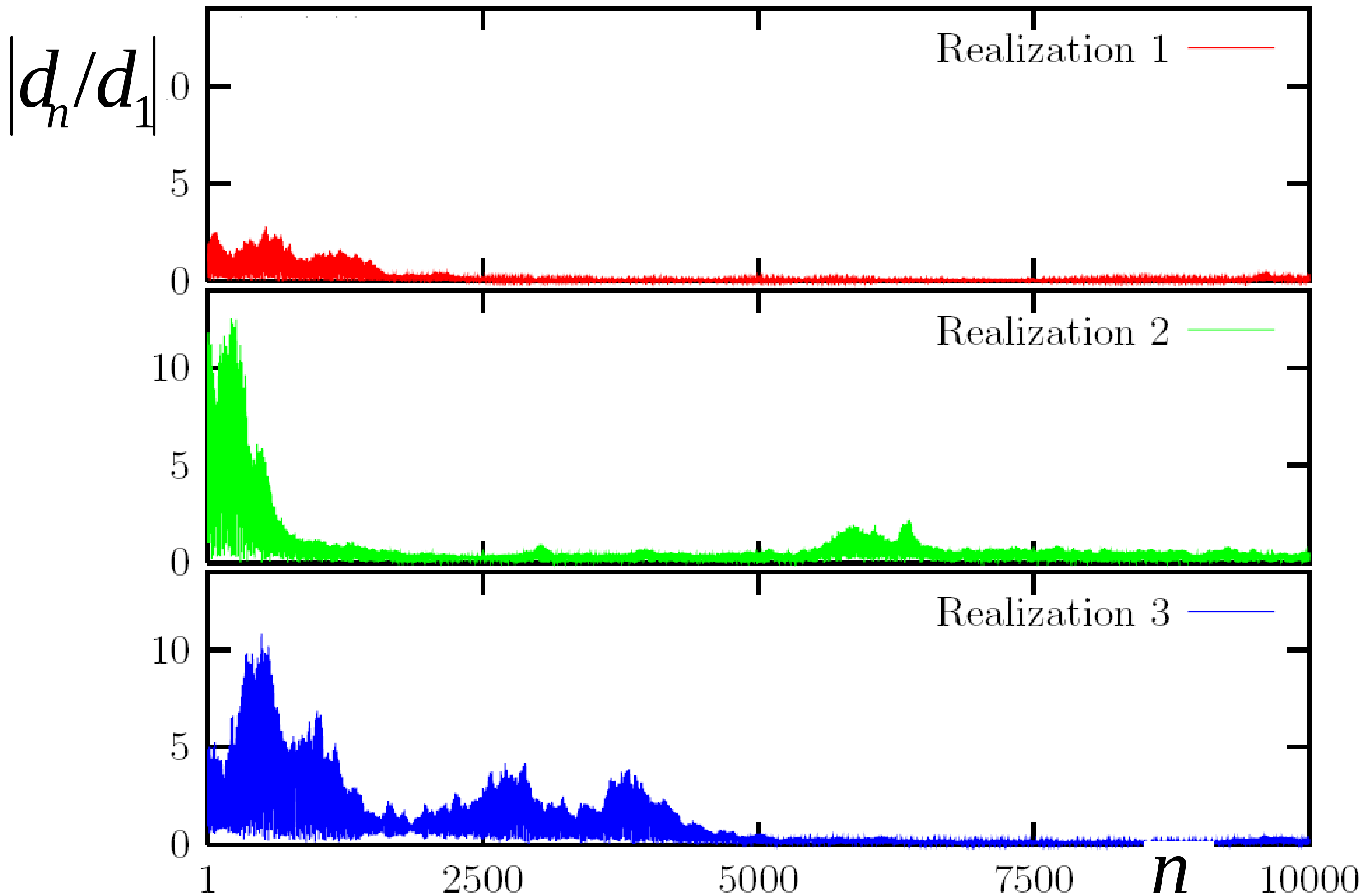
$h = 4a$

Different Realization of Disorder at the Level A=0.01



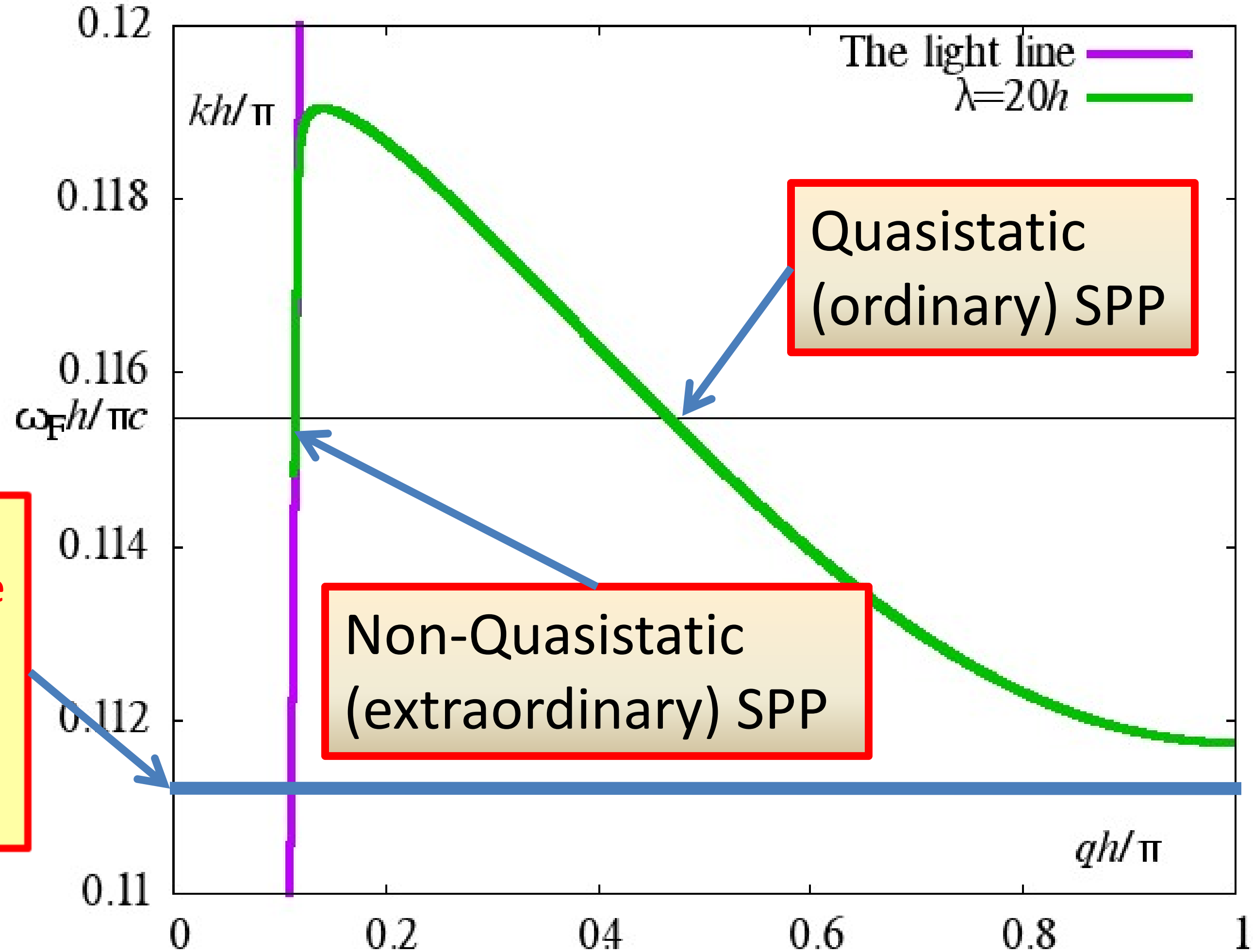
$\omega = \omega_F$

Different realization of disorder at the level A=0.02



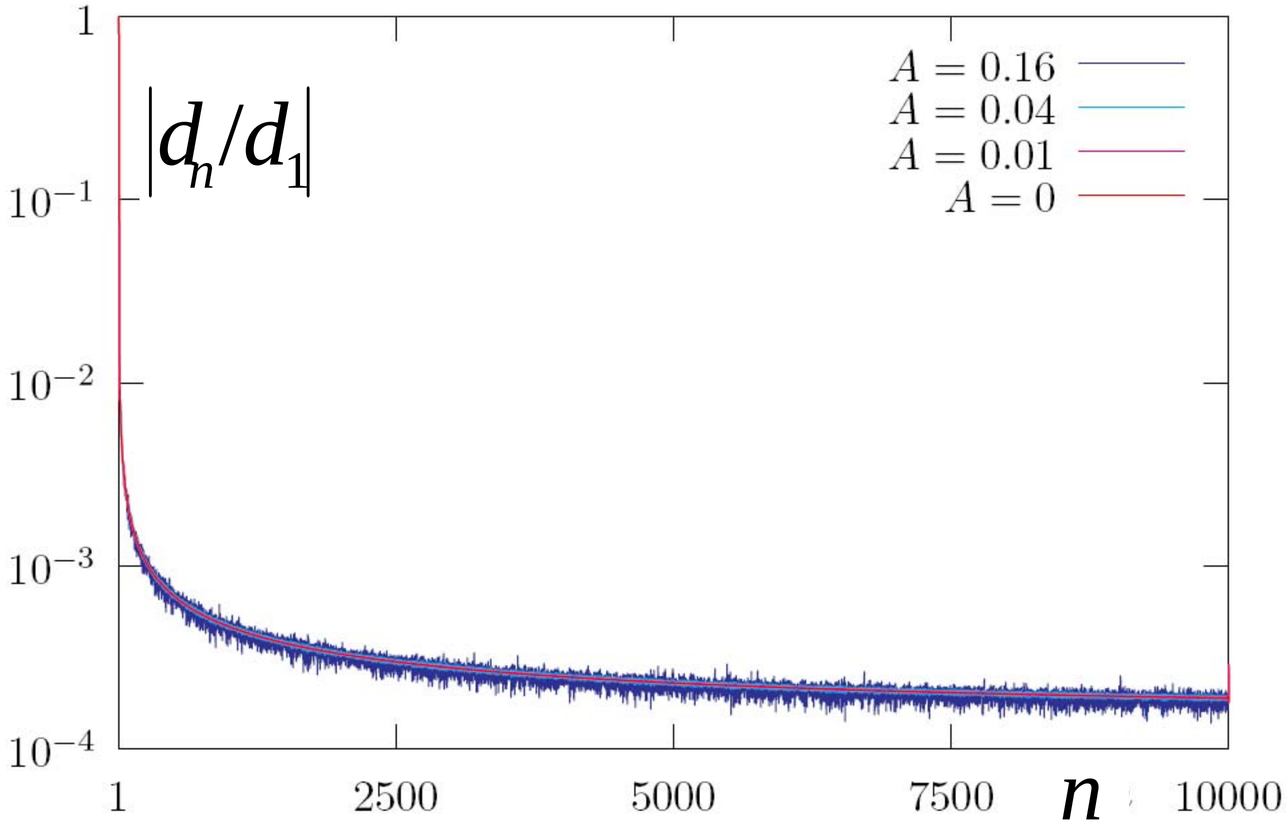
$\omega=\omega_F$

Non-quasistatic SPP at different levels of disorder



Let the frequesncy be small enough so that the ordinary SPP is not excited

Non-quasistatic SPP at different levels of disorder (continued)

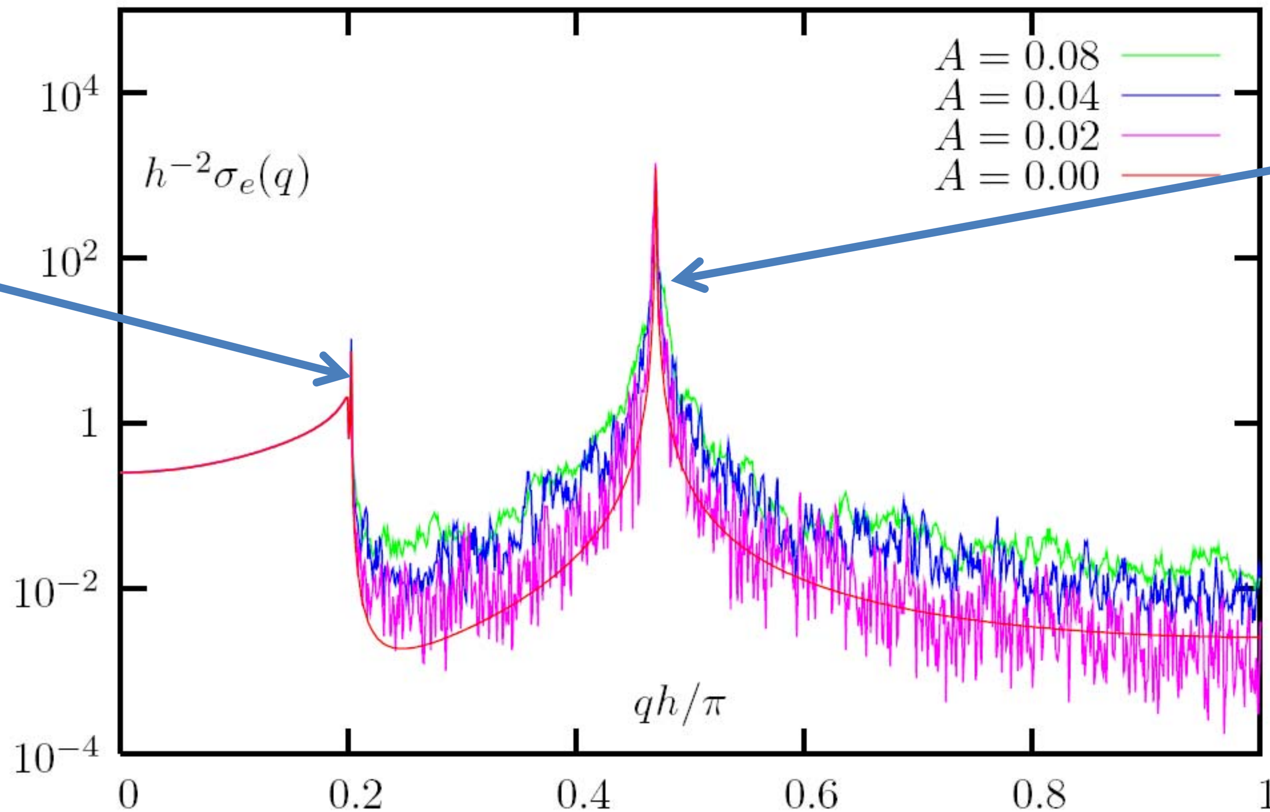


$\frac{\omega}{\omega_F}$=0.984

(small enough so that the ordinary SP is not excited)

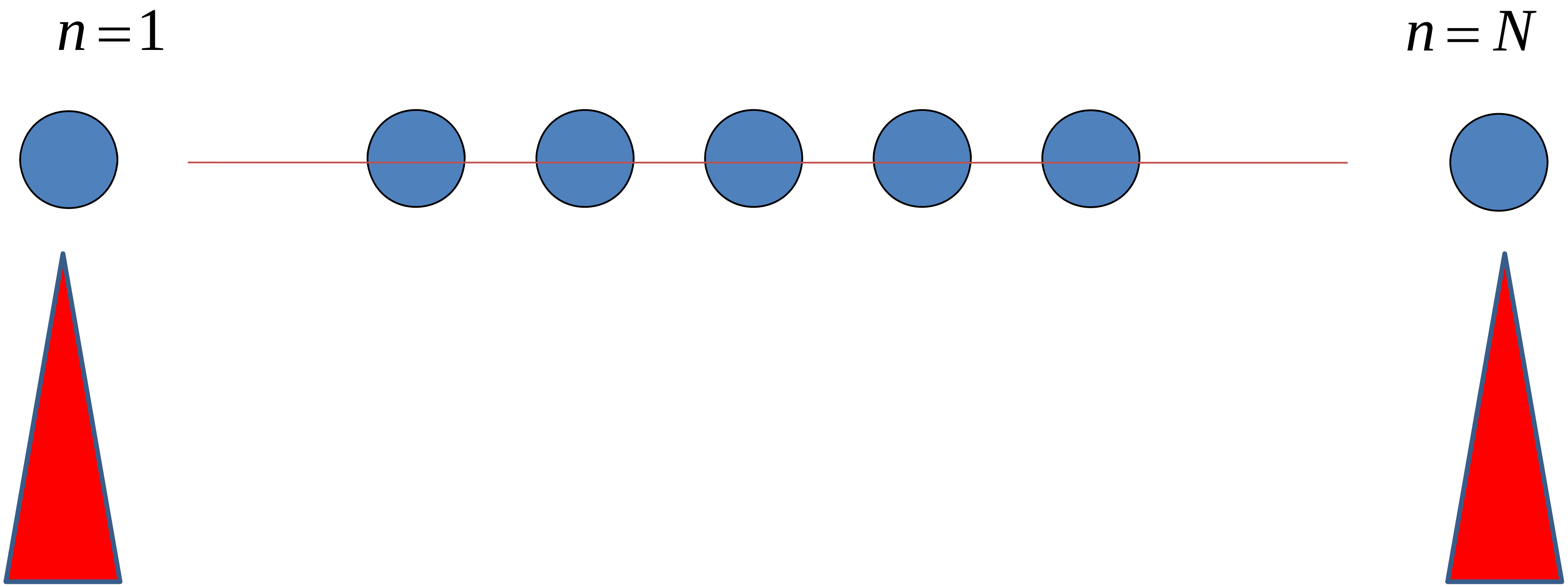
Specific extinction for excitation by a plane wave exp(iqx)
(e.g., created by the TIR, q can be larger than k)

Extraor-dinary (non-quasi-static) SPP



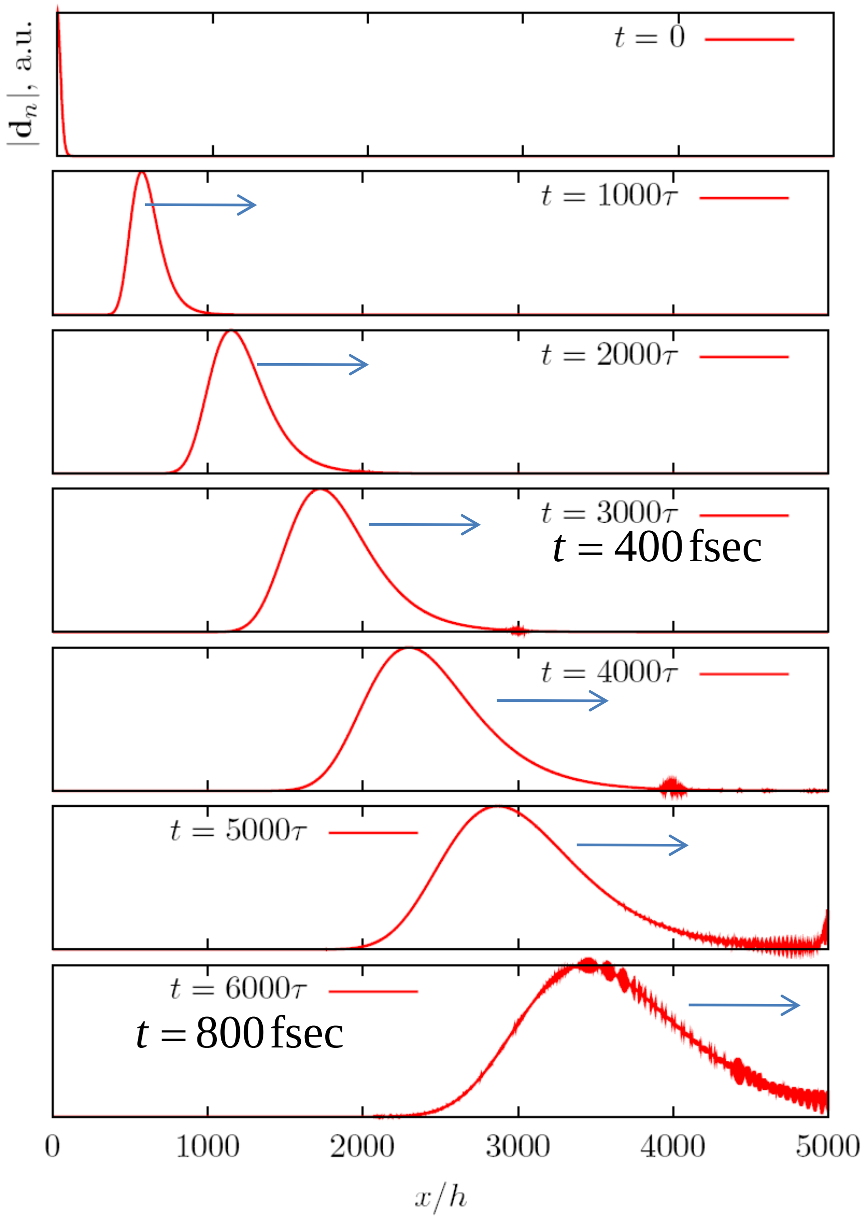
Ordinary (quasistatic) SPP

4. PROPAGATION. TRANSIENT PHENOMENA



$$E \propto \exp\left[\left(\frac{t}{\tau}\right)^2\right]\exp(-i\omega t)$$

NF tip operating in the collection mode



$v_g \approx 0.58c$

$L = 200\,\mu\text{m}$

Chain Parameters:

$h = 40\,\text{nm}$

$b = 10\,\text{nm}$

$\xi = \dfrac{b}{a} = 0.15$

$N = 5000$

$\tau = \dfrac{h}{c} = 0.133\,\text{fsec}$

Metal Parameters (Ag)

$\varepsilon = \varepsilon_0 - \dfrac{\omega_p^2}{\omega(\omega + i\gamma)}$

$\lambda_p = \dfrac{2\pi c}{\omega_p} = 136\,\text{nm}$

$\gamma/\omega_p = 0.002$

$\varepsilon_0 = 5$

Host Medium:

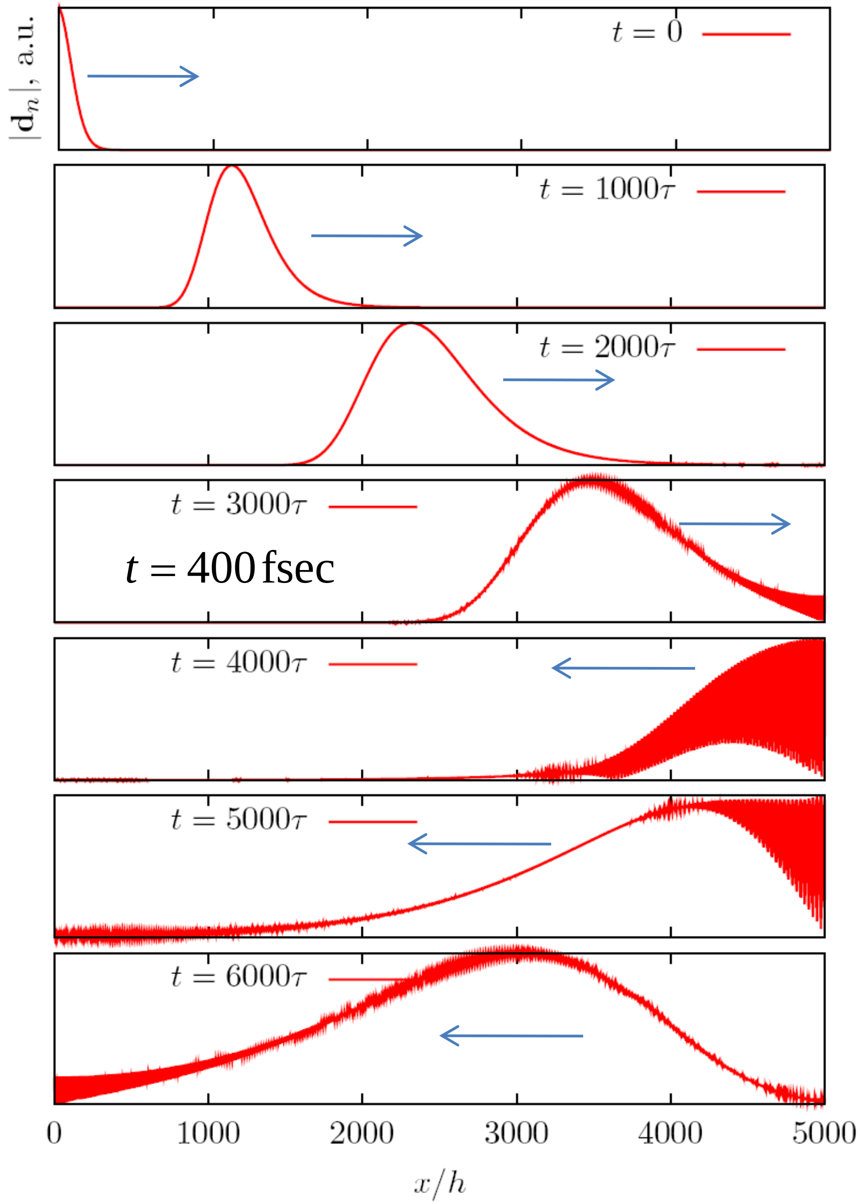
$\varepsilon_h = 2.5$

Pulse Parameters:

$\omega_0 = 0.1\omega_p\ [\lambda_0 = 1.36\,\mu\text{m}]$

$\Delta t = 7.2\,\text{fsec}$

$\Delta\omega/\omega_0 = 0.2$



$t = 400\,\text{fsec}$

$t = 800\,\text{fsec}$

Pulse Parameters Different from the Previous Graph:

$\omega_0 = 0.05\omega_p\ [\lambda_0 = 2.72\,\mu\text{m}]$

$\Delta t = 14.2\,\text{fsec}$

$\Delta\omega/\omega_0 = 0.2$ [same as before]

(But note that special relativity Is not violated. You can ask me why.)

$v_g \approx 1.17c$

5. PROPAGATION DISTANCES

Frequency	Prolate spheroid chain, transverse SPP *b*/*a*=0.15 *a*=40nm, *h*=25nm	Cylindrical wire R=25nm $\gamma \ll \omega \ll \omega_p$
$\omega = 0.1\omega_p$	7microns	3microns
$\omega = 0.05\omega_p$	15microns	3microns

Propagation distances in chains and in wires are generally comparable, but it seems that in the special case of prolate spheroids, the propagation distance can be increase by a factor of 2 – 5, depending on the working frequency.

CONSCLUSIONS

- Plasmonic chains have promissing applications in spectroscopy, sensing and waveguiding
- Theory and simulations are needed to guide the experiments and optimize design
- We need to look beyond the “traditional” EM boundary value solvers.