Non-Lorentzian Electromagnetic Resonances in One-Dimensional Chanins of Nanoparticles

Vadim A. Markel

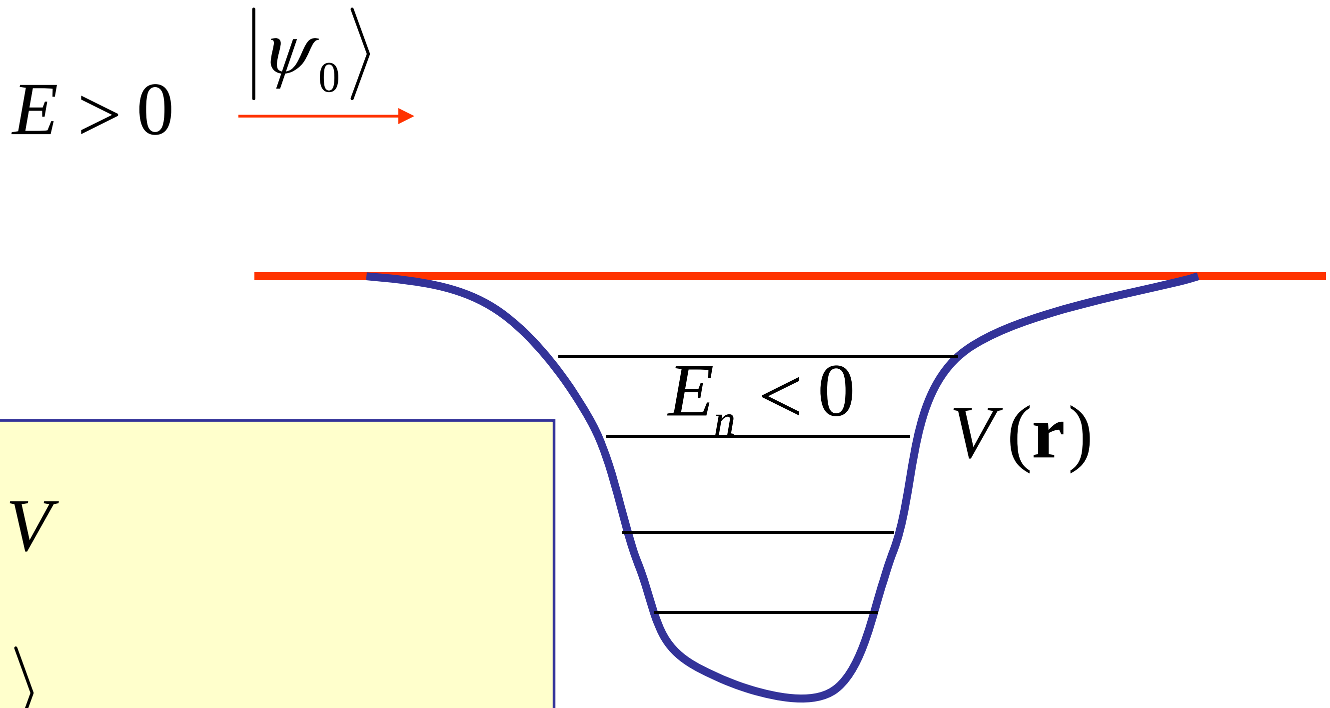
Radiology/Bioengineering

UPenn, Philadelphia

vmarkel@mail.med.upenn.edu

http://whale.seas.upenn.edu/vmarkel

Scattering Resonances in QM



$$H = -\frac{\hbar^2}{2m}\nabla^2 + V$$

$$|\psi\rangle = |\psi_0\rangle + |\psi_s\rangle$$

$$|\psi_s\rangle = [1 - G_0(E)V]^{-1} G_0(E)V|\psi_0\rangle$$

$$G_0(E) = [E + \frac{\hbar^2}{2m}\nabla^2]^{-1}$$

Resonances $\Leftrightarrow$ singularities of

$$[1 - G_0(E)V]^{-1}$$

(viewed as a function of the complex variable E)

To find the singularities of $[1 - G_0(E)V]^{-1}$, we must consider spectral properties of the linear operator

$$W = G_0(E)V$$

Generally, neither symmetric, nor Hermitian

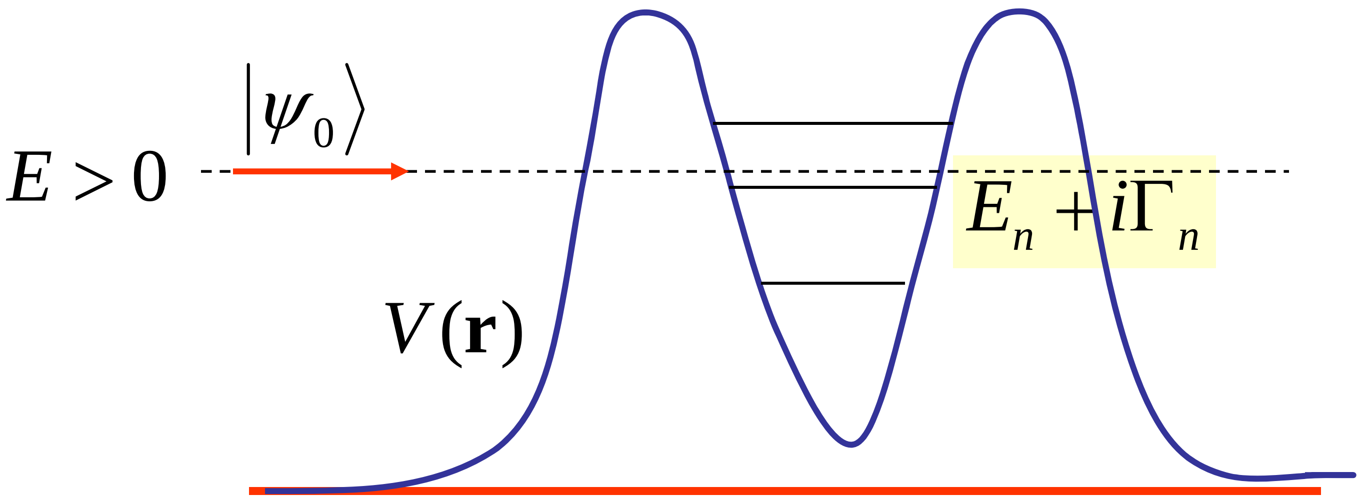
Depends on energy

Energy-independent

$$W(E)\left|\psi_n(E)\right\rangle = \lambda_n(E)\left|\psi_n(E)\right\rangle$$

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + \frac{1}{\lambda_n(E)}V\right)\left|\psi_n(E)\right\rangle = E\left|\psi_n(E)\right\rangle$$

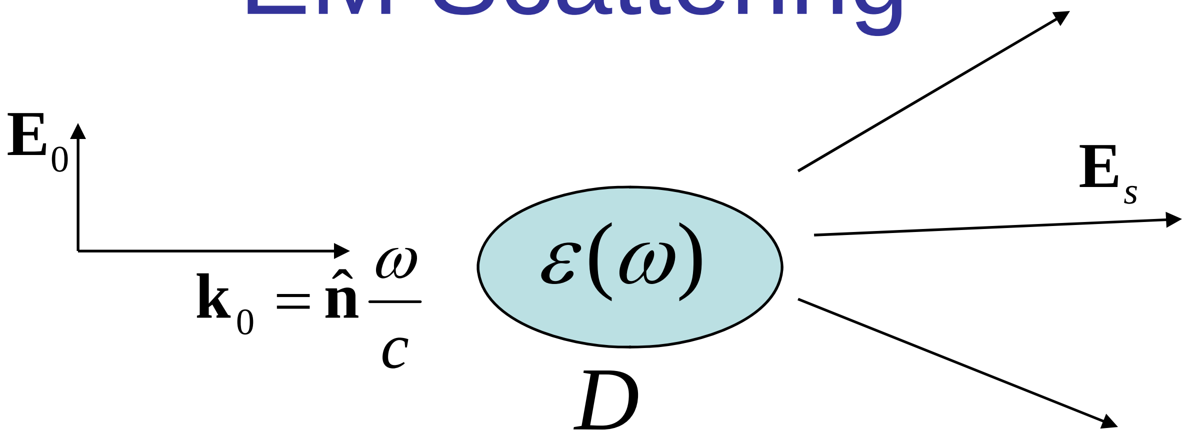
Lorentzian Resonances on Quasi-Stationary States



$$|\psi_s\rangle \approx \sum_n \frac{|n\rangle\langle n|V|\psi_0\rangle}{E - E_n - i\Gamma_n}$$

Classical Lorentzian resonances

EM Scattering



$$\mathbf{E}_s(\mathbf{r}) = [1 - G_0(\omega)V(\omega)]^{-1} G_0(\omega)V(\omega)\,\mathbf{E}_0(\mathbf{r})$$

Both operators are frequency-dependent

$$\langle \mathbf{r} | V(\omega) | \mathbf{r}' \rangle = \frac{\varepsilon(\omega) - 1}{4\pi}\Theta(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')$$

$$\Theta(\mathbf{r}) = \begin{cases} 1, \text{ if } \mathbf{r} \in D \\ 0, \text{ othervise} \end{cases}$$

Scalar parameter

$$G_0(\omega)V(\omega) = \chi(\omega)W(\omega)$$

$$\chi(\omega) = \frac{\varepsilon(\omega)-1}{4\pi}$$

$$W(\omega) = G_0(\omega)\Theta$$

Symmetric (but, generally, non-Hermitian)

Depends only on the scatterer shape

$$[1-G_0(\omega)V(\omega)]^{-1} = z(\omega)[z(\omega)-W(\omega)]^{-1}$$

$$z(\omega) = \frac{1}{\chi(\omega)}$$ - spectral parameter of the theory

Quasistatic Limit

$$G_0(\omega) \rightarrow G_0(\omega = 0) = G_0^{(\mathrm{QS})}$$

$$W(\omega) \rightarrow W(\omega = 0) = W^{(\mathrm{QS})}$$

This operator is Hermitian within the quasistatics (because we have neglected retardation)

$$[z(\omega) - W^{(\mathrm{QS})}]^{-1} = \sum_n \frac{|n\rangle\langle n|}{z(\omega) - w_n}$$

Purely real (quasisatic) eigenvalues

Lorentzian Resonances in the Quasistatics

$$z(\omega) = X(\omega) - i\delta(\omega)$$

$$[z(\omega) - W^{(\mathrm{QS})}]^{-1} \approx \sum_n \frac{|n\rangle\langle n|}{X(\omega) - w_n - i\Gamma_n} ,$$

where $\Gamma_n = \delta(\omega_n)$

and ω_n is the solution to

$$X(\omega) = w_n$$

(Qasiparticle pole approximation)

EXAMPLE:

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

$$X(\omega) = -4\pi \frac{\omega^2}{\omega_P^2} ,$$

$$\delta(\omega) = 4\pi \frac{\omega\gamma}{\omega_P^2} ,$$

$$\Gamma_n = \sqrt{4\pi w_n} \frac{\gamma}{\omega_P}$$

Resonances Beyond Quasistatics

$$w_n = w_n(\omega)$$

$$[z(\omega) - W]^{-1} \approx \sum_n \frac{F_n}{X(\omega) - \mathrm{Re}[w_n(\omega_n)] - i\Gamma_n},$$

where $F_n = \dfrac{|n\rangle\langle\overline{n}|}{\langle\overline{n}|n\rangle}$, $\quad \sum_n F_n = 1$

$$\Gamma_n = \delta(\omega_n) - \mathrm{Im}[w_n(\omega_n)]$$

and ω_n is the solution to

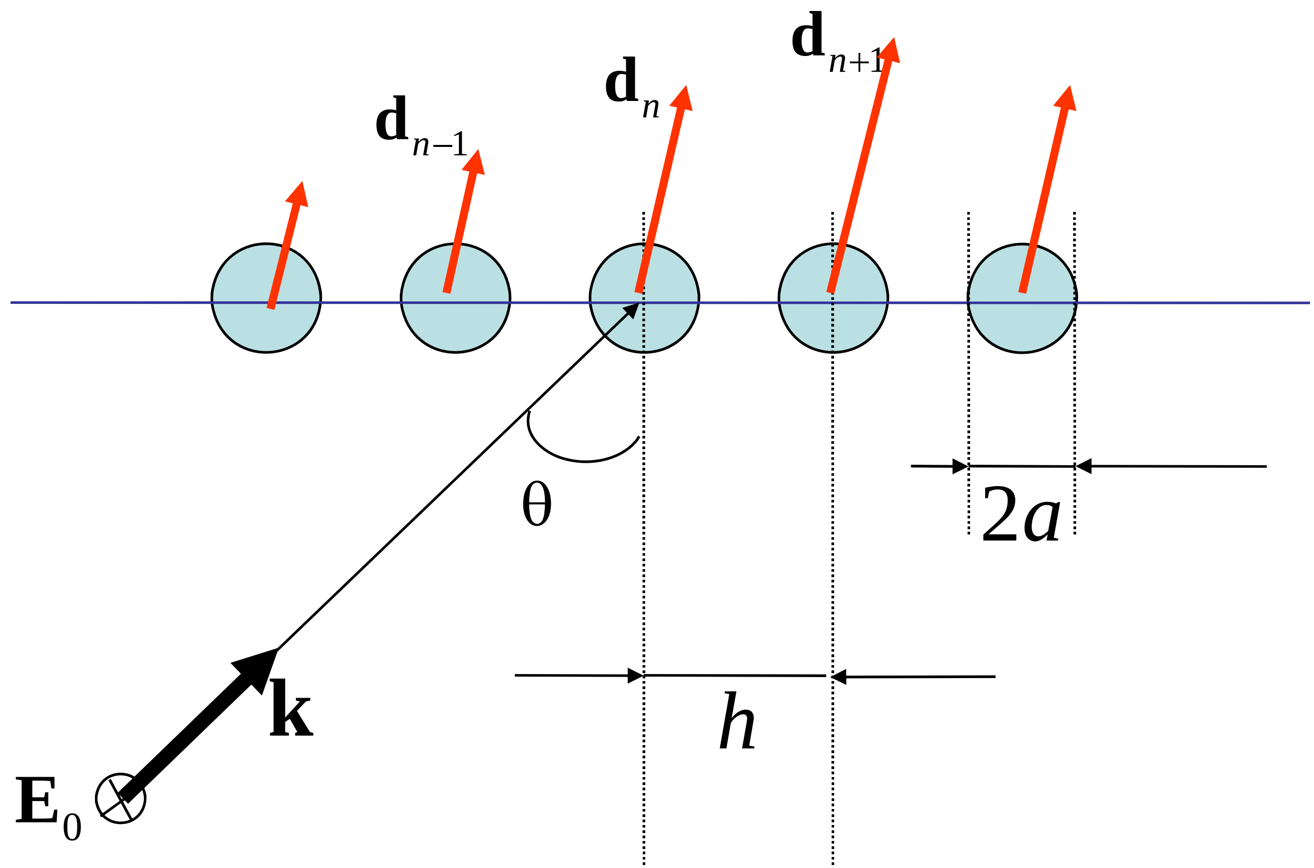
$$X(\omega) = \mathrm{Re}[w_n(\omega)]$$

Origin of the Non-Lorentzian Resonances

What if $w_n(\omega)$ is a much faster function than $X(\omega)$?

The quasiparticle pole approximation will not be valid in this case.

Physical Model



Dipole Approximation

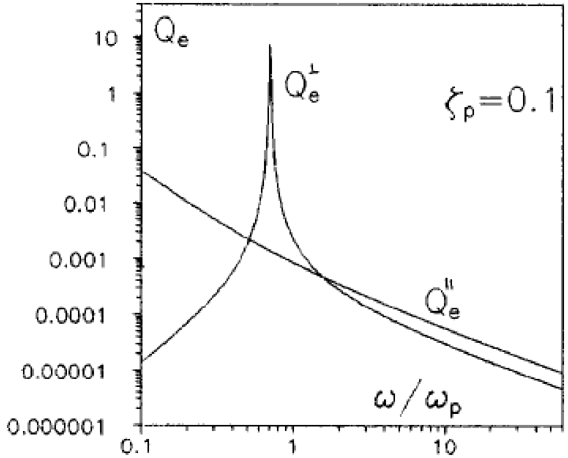
$$d_n^{\perp} = \alpha\left[E_0 e^{ikn\sin\theta} + \sum_{n'\neq n} W_{n-n'}(kh)\, d_{n'}^{\perp}\right]$$

$$W_n(x) = k^3\left[\frac{1}{|xn|} + \frac{i}{|xn|^2} - \frac{1}{|xn|^3}\right] e^{i|xn|}$$

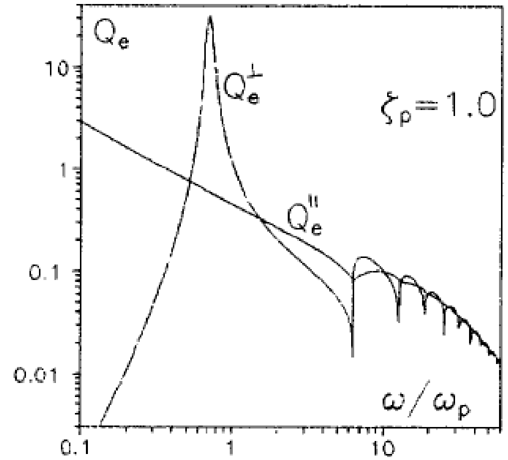
$$d_n^{\perp} = \frac{a^3 E_0 e^{ikhn\sin\theta}}{\underbrace{a^3/\alpha}_{z(\omega)} - \underbrace{(ka)^3 S(kh)}_{w(\omega)}}$$

$$S(x) = 2\sum_{n>0}\left[\frac{1}{xn} + \frac{i}{(xn)^2} - \frac{1}{(xn)^3}\right] e^{inx}\cos[nx\sin\theta]$$

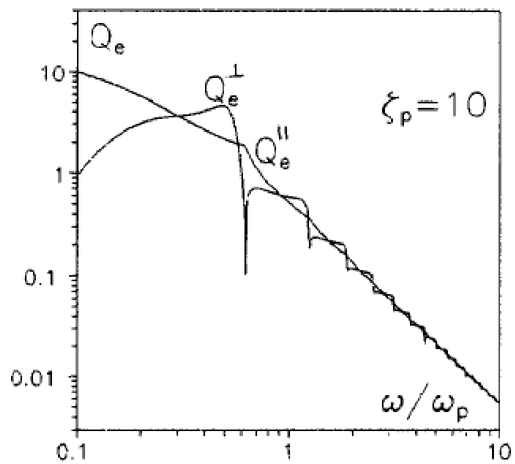
Diverges if $(1 \pm \cos\theta)\,kh = 2\pi l$



(*a*)



(*b*)



(*c*)

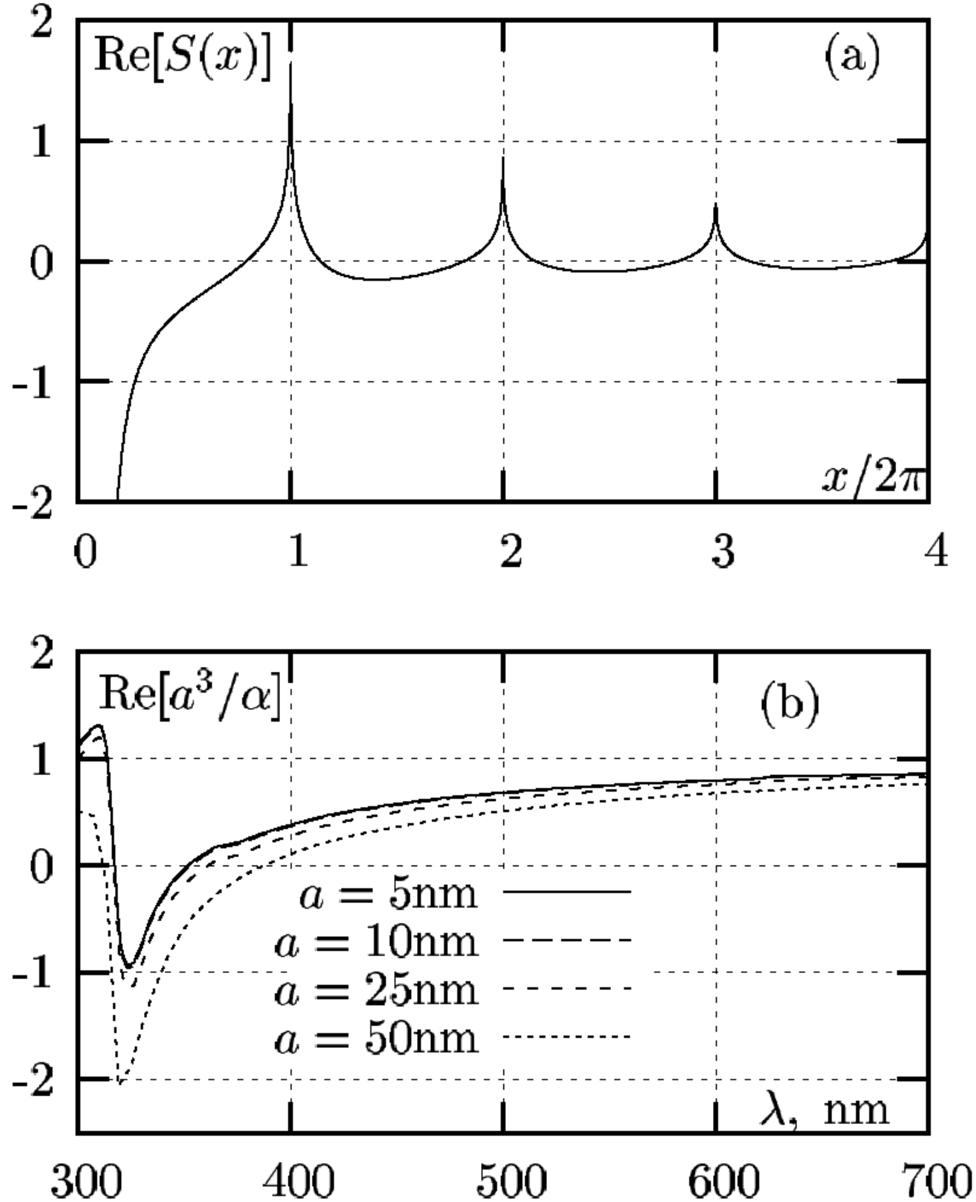
Narrow spectral features in extinction spectra of an infinite chain of Drudean spheres.

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

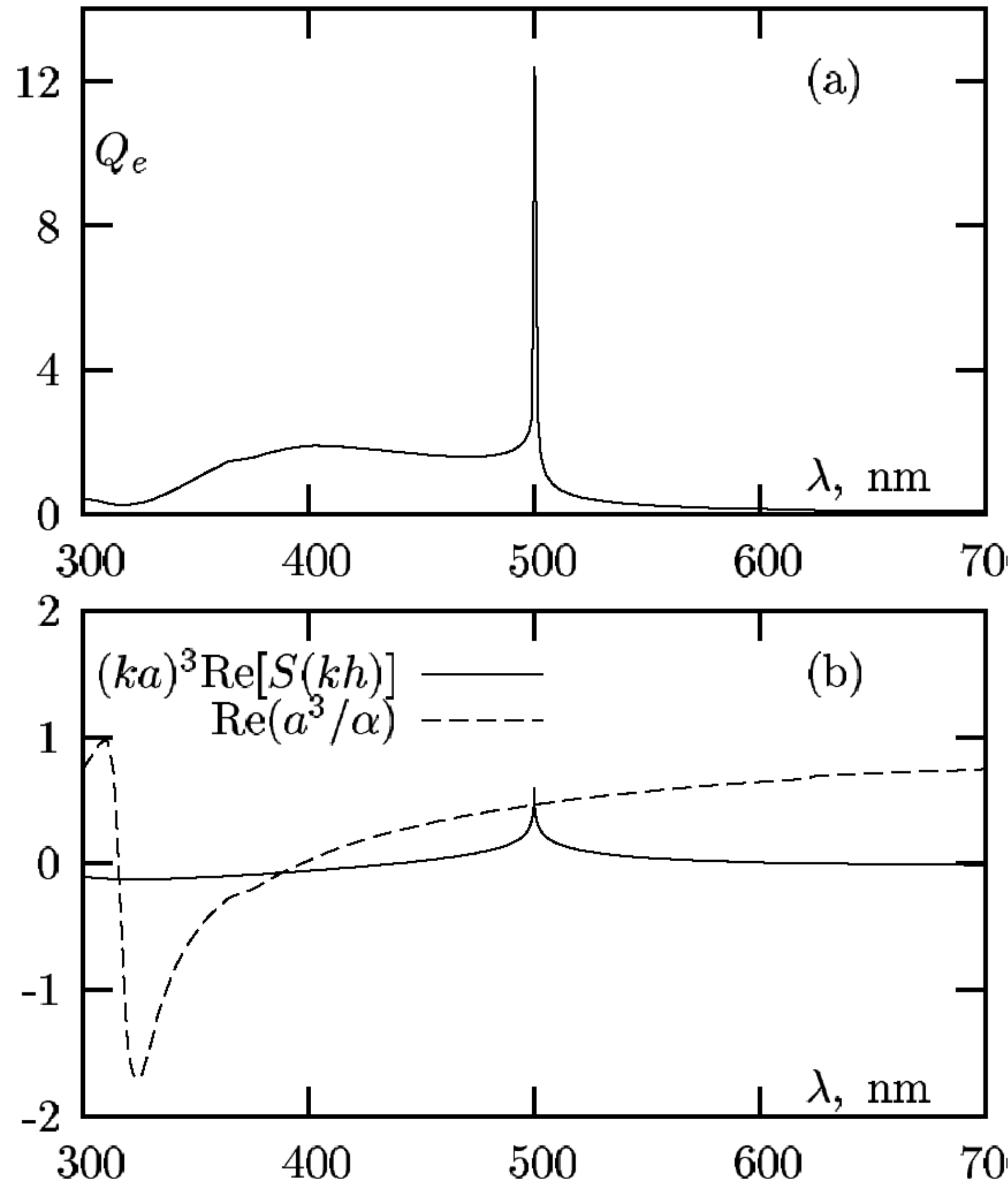
" $\perp$ " - polarization orthogonal to the chain

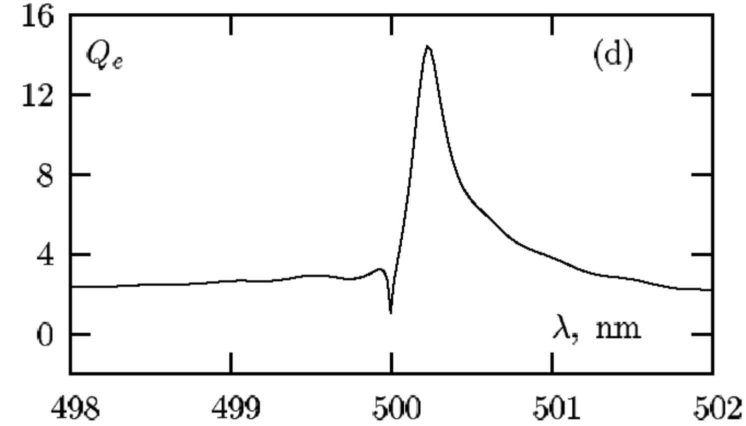
" $\|$ "- polarization parallel to the chain

(from V.A.Markel, *J. Mod. Opt*., 1993 **40**(11), 2281-2291)



Re[S(x)]
(a)
x/2π
2
1
0
-1
-2
0
1
2
3
4
Re[a³/α]
(b)
a = 5nm
a = 10nm
a = 25nm
a = 50nm
λ, nm
2
1
0
-1
-2
300
400
500
600
700

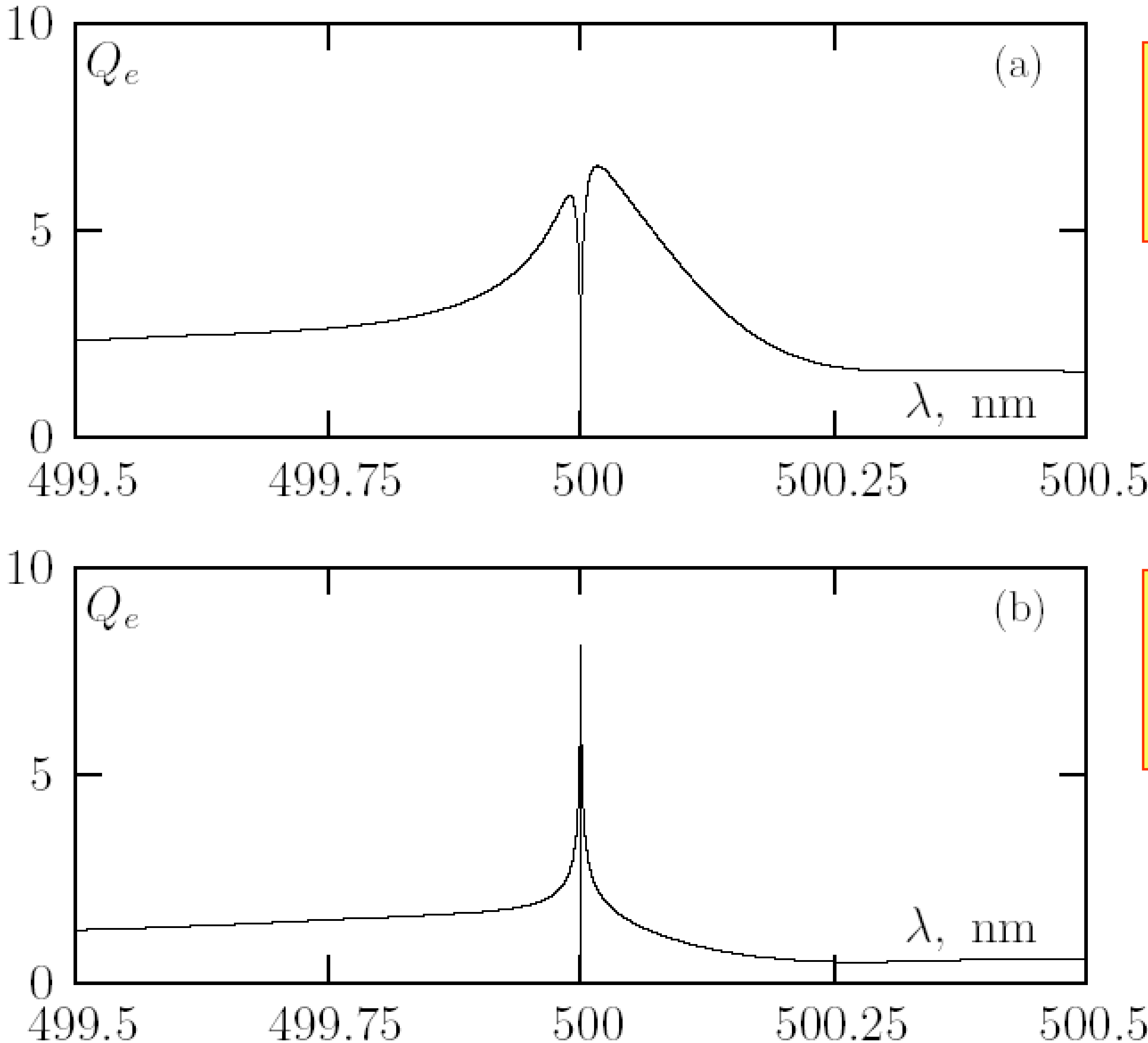




$a = 50\mathrm{nm}, \quad h = 500\mathrm{nm}$

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

$C \sim 1$



$a = 45$nm

$h = 500$nm

$a = 40$nm

$h = 500$nm

Unusual Properties of the Non-Lorentzian Resonances in 1D Dipole Chains

- Transverse dipole oscillations are shifted to the RED (normally, they would be shifted to the BLUE) from the plasmon frequency.
- Have negligibly small integral weight.
- Width is not controlled by relaxation.
- Can exists in a chain where the interparticle distance is much larger than the particle diameter.
- Can not be excited in the near field (i.e., by a near-field probe).
- Spectral lines consist of two sharp peaks; extinction in a poit between the peaks is exactly zero (in infinite chains).
- In principle, can be arbitrarily narrow (but this would require exponentially long chains).

Limitations and Potential Problems

Cause of inaccuracy

Expected effect on the resonance lineshapes (authors intelligent guess)

Finite-size and quantum effects	Minor effect
Dipole approximation	Minor; would not broaden the resonances
Short-range disorder	Can broaden resonances
Long-range disorder	Can eliminate resonances
Nonlinearity	?

Publications

V.A.Markel, “Coupled-dipole approach to scattering of light from a one-dimensional periodic dipole structure,” *J.Mod.Opt.* **40**(11) 2281-2291, (1993).

V.A.Markel, “Divergence of dipole sums and the nature of non-Lorentzian exponentially narrow resonances in one-dimensional periodic arrays of Nanospheres,” J.Phys.B **38,** L115–L121 (2005).

Available electronically from http://whale.seas.upenn.edu/vmarkel/papers.html