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

Vadim A. Markel
Radiology/Bioengineering
UPenn, Philadelphia

vmarkel@mail.med.upenn.edu
http://whale.seas.upenn.edu/vmarkel
Scattering Resonances in QM

\[ E > 0 \quad |\psi_0\rangle \]

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

\[ |\psi\rangle = |\psi_0\rangle + |\psi_s\rangle \]

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

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

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

Energy-independent

Depends on energy

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

\[ \left( -\frac{\hbar^2}{2m} \nabla^2 + \frac{1}{\lambda_n(E)} V \right)\left|\psi_n(E)\right\rangle = E\left|\psi_n(E)\right\rangle \]
Lorentzian Resonances on Quasi-Stationary States

\[ E > 0 \]

\[ |\psi_0\rangle \]

\[ V(r) \]

\[ E_n + i\Gamma_n \]

\[ |\psi_s\rangle \approx \sum_n \left| n \rightangle \left\langle n | V | \psi_0 \right\rangle \frac{E - E_n - i\Gamma_n}{E - E_n - i\Gamma_n} \]

Classical Lorentzian resonances
EM Scattering

\[ k_0 = \hat{n} \frac{\omega}{c} \]

\[ \varepsilon(\omega) \]

\[ D \]

\[ E_s (\mathbf{r}) = \left[ 1 - G_0(\omega)V(\omega) \right]^{-1} G_0(\omega)V(\omega) E_0 (\mathbf{r}) \]

Both operators are frequency-dependent

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

\[ \Theta(\mathbf{r}) = \begin{cases} 
1, & \text{if } \mathbf{r} \in D \\
0, & \text{otherwise} 
\end{cases} \]
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

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

\[ z(\omega) = \frac{1}{\chi(\omega)} \quad \text{- spectral parameter of the theory} \]
Quasistatic Limit

\[ G_0(\omega) \rightarrow G_0(\omega = 0) = G_0^{(QS)} \]
\[ W(\omega) \rightarrow W(\omega = 0) = W^{(QS)} \]

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

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

Purely real (quasistatic) eigenvalues
Lorentzian Resonances in the Quasistatics

\[ z(\omega) = X(\omega) - i\delta(\omega) \]

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

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

and \( \omega_n \) 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

\[ w_n = w_n(\omega) \]

\[ [z(\omega) - W]^{-1} \approx \sum_{n} \frac{F_n}{X(\omega) - \text{Re}[w_n(\omega_n)] - i\Gamma_n}, \]

where

\[ F_n = \frac{|n\rangle\langle\bar{n}|}{\langle\bar{n}|n\rangle}, \quad \sum_{n} F_n = 1 \]

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

and \( \omega_n \) is the solution to

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

\[ n - d + \theta = h \]

\[ E_0 \rightarrow k \]

\[ d_{n-1}, d_n, d_{n+1} \]

\[ 2a, h \]
Dipole Approximation

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

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

\[ d_n^\perp = \frac{a^3 E_0 e^{ik\sin \theta}}{a^3 / \alpha - (ka)^3 S(kh)} \]

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

Diverges if \((1 \pm \cos \theta)kh = 2\pi l\)
Narrow spectral features in extinction spectra of an infinite chain of Drudean spheres.

\[
\frac{\omega_p h}{c} = 0.1 \quad (a), \quad \frac{\omega_p h}{c} = 1 \quad (b), \quad \text{and} \quad \frac{\omega_p h}{c} = 10 \quad (c).
\]

"⊥" - polarization orthogonal to the chain

"||" - polarization parallel to the chain

(from V.A. Markel, *J. Mod. Opt.*, 1993 40(11), 2281-2291)
\[ a = 50 \text{nm}, \quad h = 500 \text{nm} \]

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

\[ C \sim 1 \]
\[\begin{align*}
\text{(a)} \\
\lambda, \text{ nm} \\
\end{align*}\]

\[\begin{align*}
Q_e \\
\lambda, \text{ nm} \\
\end{align*}\]

\[\begin{align*}
\text{(b)} \\
\lambda, \text{ nm} \\
\end{align*}\]

\[\begin{align*}
a &= 45\text{nm} \\
h &= 500\text{nm} \\
\end{align*}\]

\[\begin{align*}
a &= 40\text{nm} \\
h &= 500\text{nm} \\
\end{align*}\]
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**

<table>
<thead>
<tr>
<th>Limitation</th>
<th>Expected effect on the resonance lineshapes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Finite-size and quantum effects</td>
<td>Minor effect</td>
</tr>
<tr>
<td>Dipole approximation</td>
<td>Minor; would not broaden the resonances</td>
</tr>
<tr>
<td>Short-range disorder</td>
<td>Can broaden resonances</td>
</tr>
<tr>
<td>Long-range disorder</td>
<td>Can eliminate resonances</td>
</tr>
<tr>
<td>Nonlinearity</td>
<td>?</td>
</tr>
</tbody>
</table>
Publications


Available electronically from [http://whale.seas.upenn.edu/vmarkel/papers.html](http://whale.seas.upenn.edu/vmarkel/papers.html)