Adjoint-Based Photonic Design: Optimization for Applications from Super-Scattering to Enhanced Light Extraction

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Post-doc, MIT Applied Math

PI: Steven Johnson

Collaborators: *Homer Reid (Math), Chia Wei Hsu (Harvard Physics), Wenjun Qiu (Physics), Brendan DeLacy (US Army ECBC), Marin Soljacic (Physics), John Joannopoulos (Physics)*

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Super-Scattering Particle Design

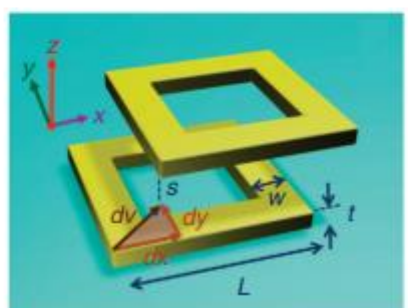
- *Fundamental Question*: How can we design nano-particles to maximally extinguish light, per unit of material volume/weight?

Extinction = Absorption + Scattering

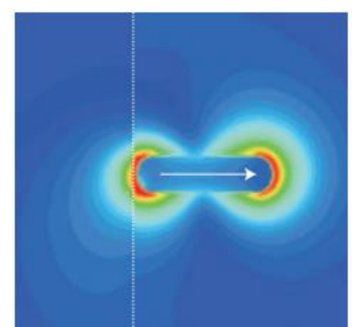
- *Application*: Obscurance (i.e. smokescreens)

Troops concealed by smokescreens

- Nano-particle absorbers/scatterers have potential applications in
  - Imaging
  - Biomedicine
  - Optical antennas
  - Metamaterials



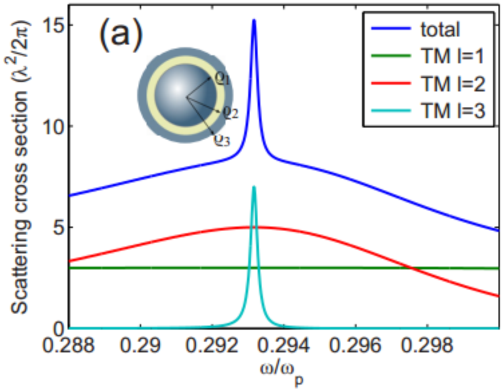
Zhang et al
Nat. Comm. 3, 1180 (2012)



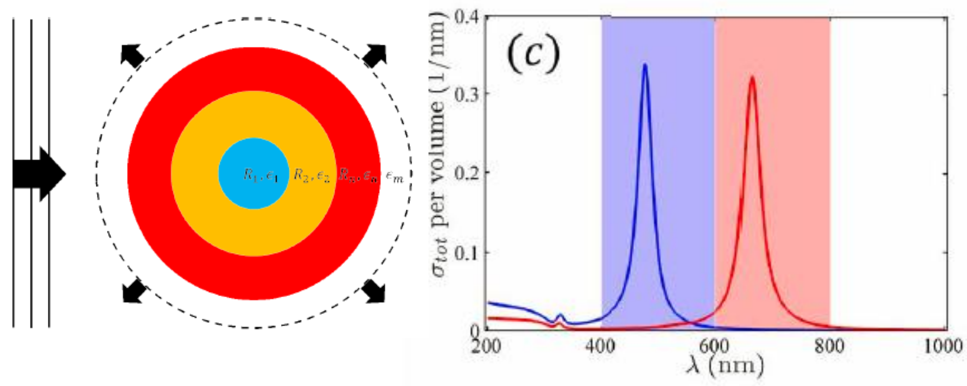
Van Hulst et al
Nat. Photon. 2, 234 (2008)

Previous work

- Primarily spherically-symmetric structures



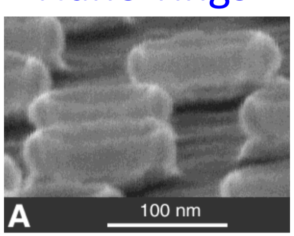
Fan et. al. APL 98, 043101 (2011)



Soljacic et. al. Opt. Exp. 21, 1465 (2012)

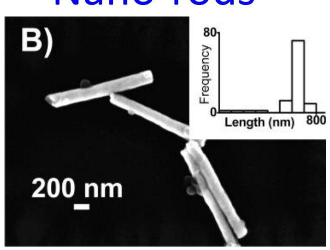
- Some exploration of non-spherical particles: not systematic, or at different frequencies / different metrics

Nano-rings



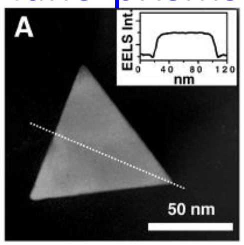
Aizpurua et. al. PRL 90, 057401 (2003)

Nano-rods



Payne et. al. JCB 110, 2150 (2006)

Nano-prisms



Jun et. al. Science 294, 1901 (2001)
Kelly et. al. JPCB 107, 668 (2003)

The Computational Challenge

Goal: maximize **extinction / volume,** $\boldsymbol{\frac{\sigma}{V}}$

- Need to explore large design space of non-spherical, three-dimensional structures

- For every structure, many frequencies (broadband performance)

- For every frequency, many incidence angles (random orientation)

The Computational Challenge

Goal: maximize **extinction / volume,** $\frac{\sigma}{V}$

Our Approach

- Need to explore large design space of non-spherical, three-dimensional structures

  Adjoint-based shape derivatives, within sophisticated optimizer

ab-initio.mit.edu/NLopt

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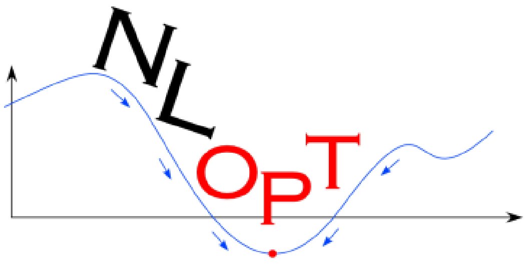

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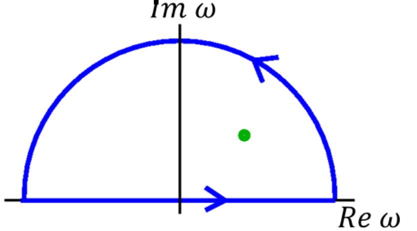
- For every structure, many frequencies (broadband performance)
  Complex-frequency transformation



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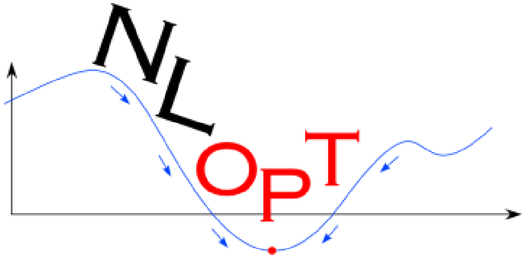

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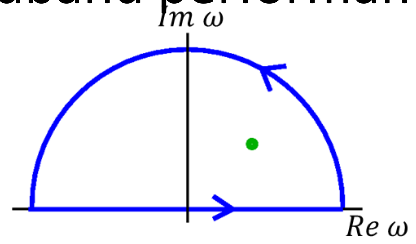
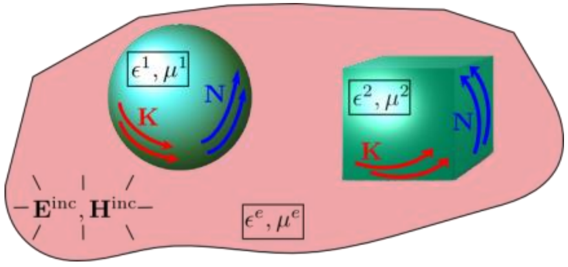


- For every frequency, many incidence angles (random orientation)
  Boundary-element method
  - Discretize surface only, not volume
  - all angles essentially free



homerreid.ath.cx/scuff-EM/

Complex Frequency Transformation

- By optical theorem, extinction equals:

$$\sigma = Im[f]$$

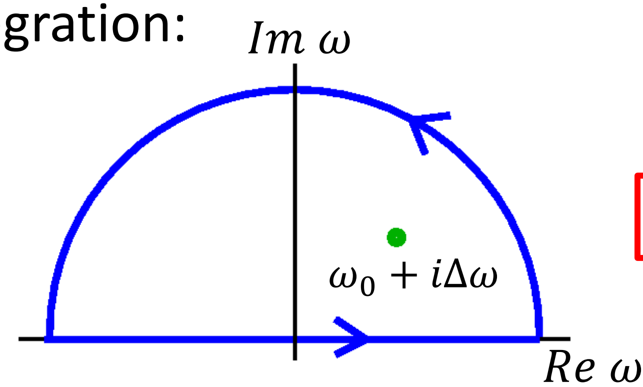
f = forward scattering amplitude
→ analytic in upper-half place (causality)

- Suppose we want to measure broadband performance:

$$\sigma_{avg} = Im \int \underbrace{f(\omega)}_{\text{analytic (no poles)}} \underbrace{\frac{\Delta\omega/\pi}{(\omega-\omega_0)^2+\Delta\omega^2}}_{\text{one pole at } \omega_0 + i\Delta\omega} d\omega$$

Lorentzian "window" function

- Contour integration:



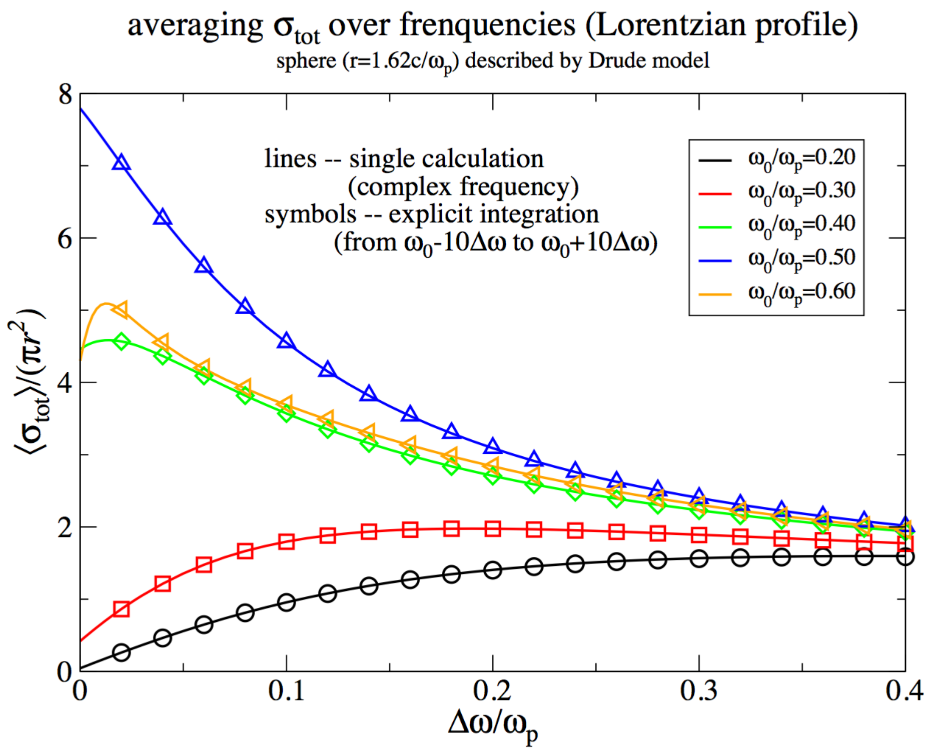
$$\boxed{\sigma_{avg} = Im\, f(\omega_0 + i\Delta\omega)}$$

Many real ω to One complex ω!

Verification: many-to-one frequency transform

Complex frequency = *Complex materials*

$$\omega_0 \to \omega_0 + i\Delta\omega \iff \varepsilon, \mu \to \varepsilon, \mu(\omega_0 + i\Delta\omega) \cdot \left(1 + i\frac{\Delta\omega}{\omega_0}\right)$$

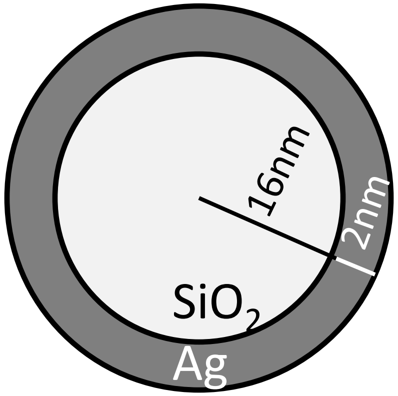


Can be solved with existing FEM/BEM codes!

Beyond Spheres: Ellipsoids

Optimizing $\frac{\sigma}{V}$ over $\lambda = [600{,}800]nm$, random orientation

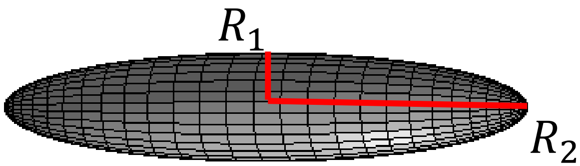
- Among all multi-coated SiO_2/Ag spheres, global optimum always



Very small (quasi-static)
Silica sphere with
single Silver coating

$$\frac{\sigma_{avg}}{V} = \int \frac{\sigma(\omega)}{V} \frac{\Delta\omega/\pi}{(\omega-\omega_0)^2 + \Delta\omega^2} d\omega$$
$$= 0.09nm^{-1}$$

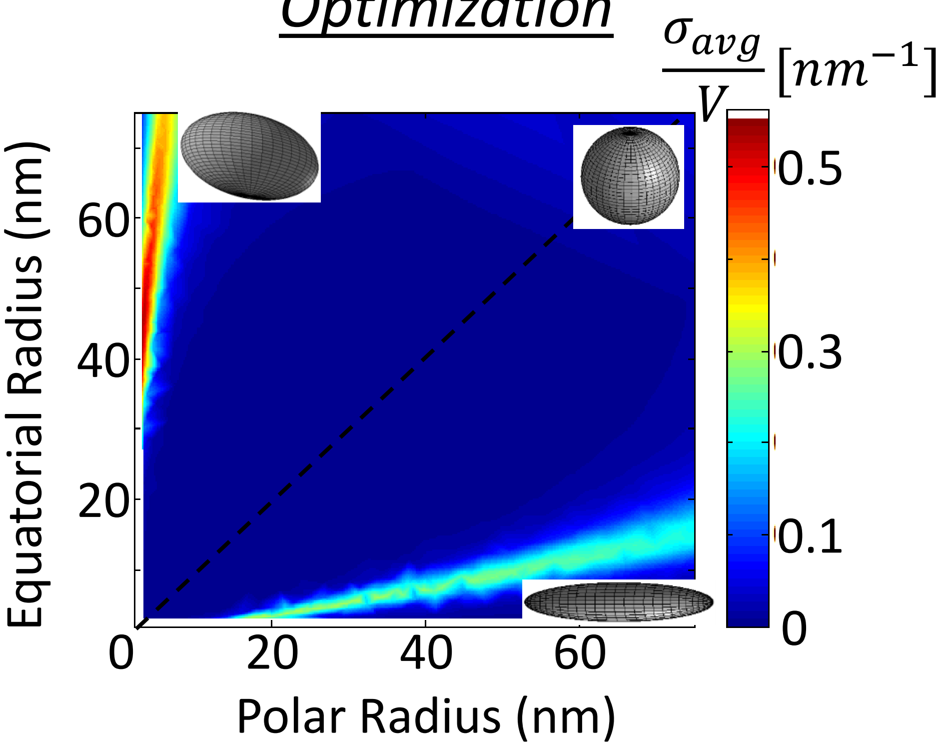
- Extending optimization to ellipsoids, how well can we do?



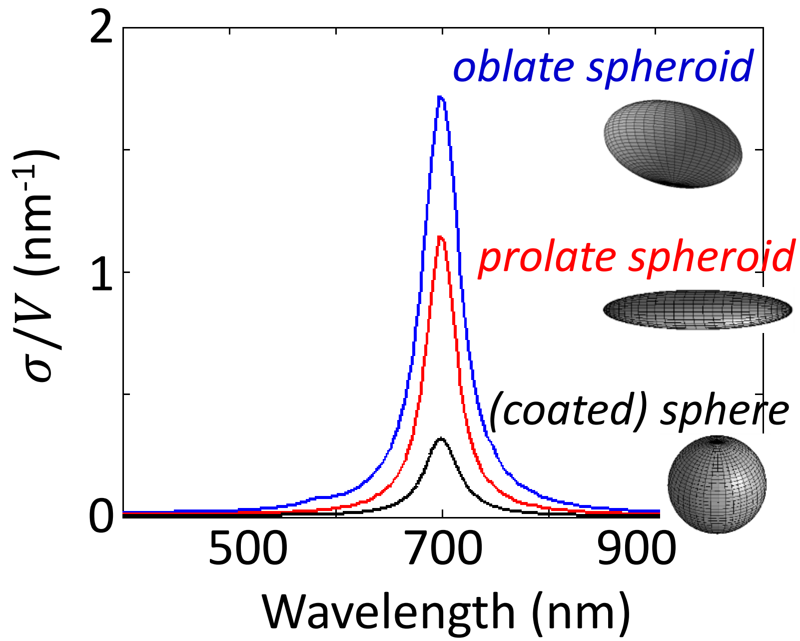
Assume surface of revolution
(i.e. spheroids, 2 degrees of freedom)

Optimal Un-Coated, Ag Ellipsoids

Optimization



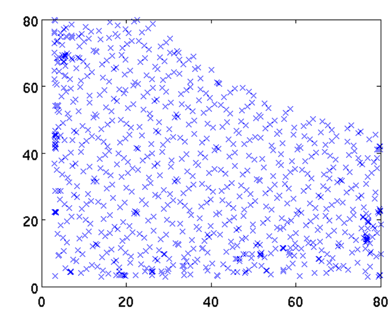
Optimal Shapes



Optimal Ellipsoid

- $r_1 = 3nm, r_2 = r_3 = 45nm$ (r_1 at lower bound)
- $\frac{\sigma_{avg}}{V} = 0.53nm^{-1}$, ~6x better than optimal sphere
- Oblate (disk) > Prolate (needle)

Actual sampling

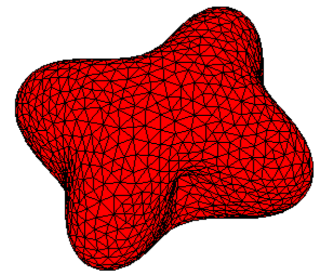


Beyond Ellipsoids: Star-Shaped Structures

- Use spherical harmonics as basis functions for shapes

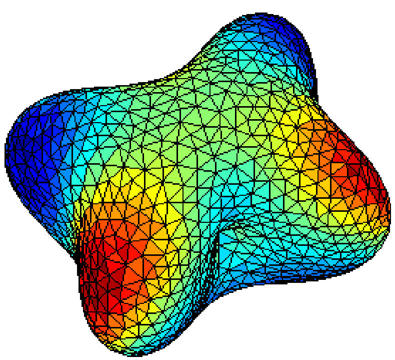
$$r(\theta,\phi) = \sum_{l,m} c_{lm} Y_{lm}(\theta,\phi)$$

- Adjoint shape derivatives: reciprocity in action



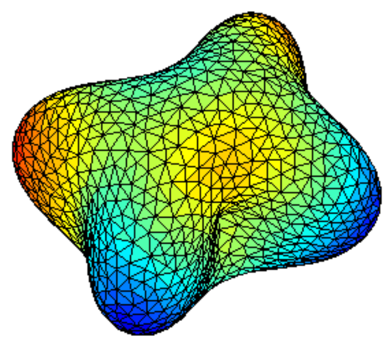
Example Structure

Direct Simulation



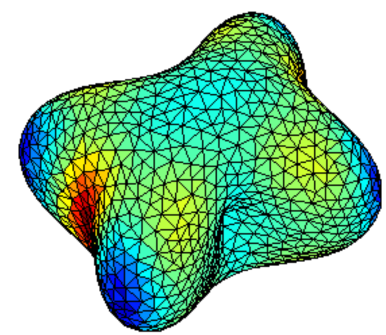
$\otimes$

Adjoint Simulation



$=$

Gradient



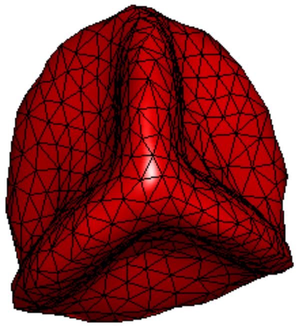
$$\frac{\delta F}{\delta x_n} = (\epsilon_2 - \epsilon_1)\vec{E}_{\parallel} \cdot \vec{E}^{A}_{\parallel} + \left(\frac{1}{\epsilon_1} - \frac{1}{\epsilon_2}\right)\vec{D}_{\perp} \cdot \vec{D}^{A}_{\perp}$$

With only two simulations → Derivative at every surface point!

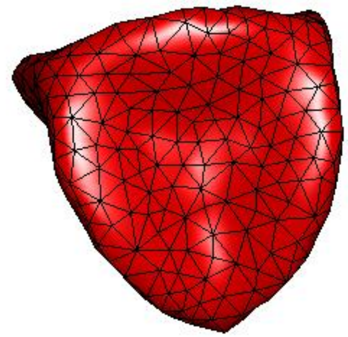
Optimization #1: Ag near λ=400nm

Optimal Structure (<150 iterations)

"Top" View



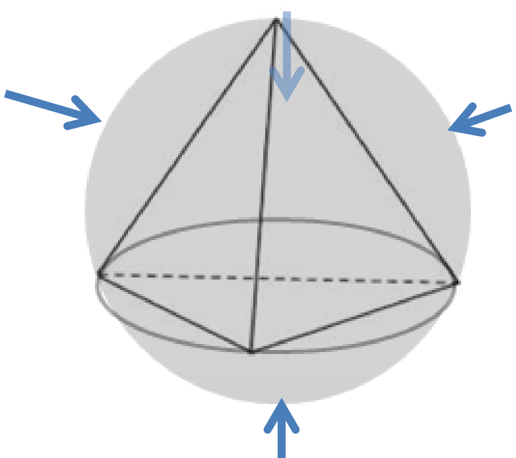
"Side" View



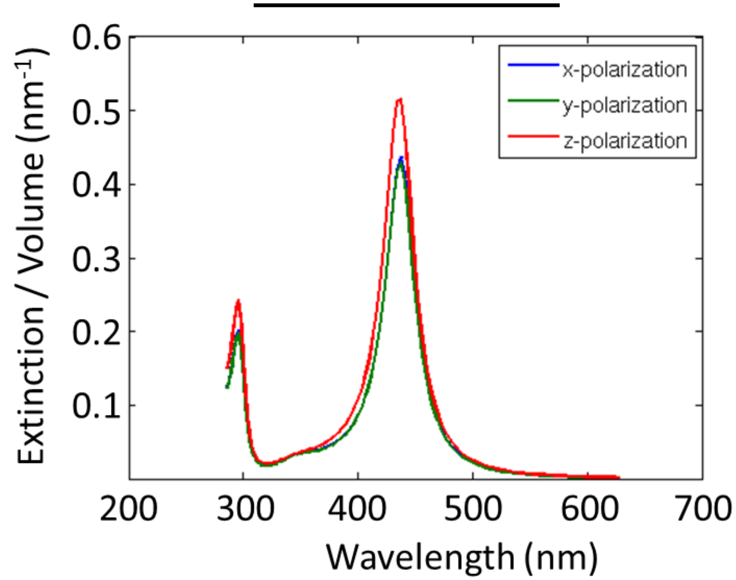
Dimensions ~5nm

How to think about structure?

- Inscribe tetrahedron in sphere
- "Push in" at centroids



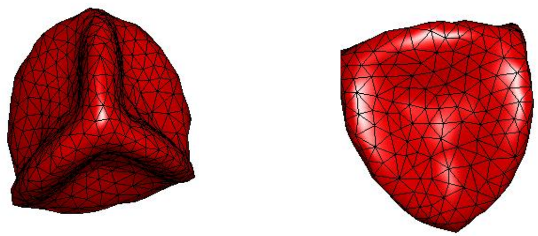
Performance

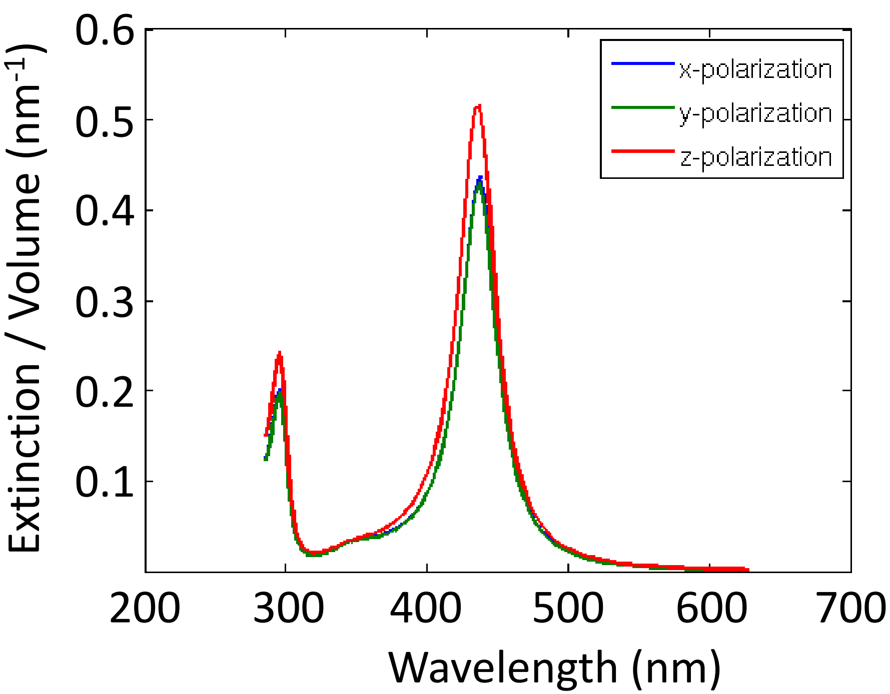


≈ equal for all 3 polarizations

Comparison of Optimal Structures

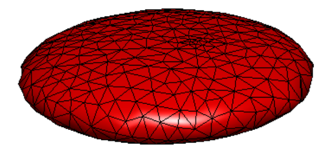
Optimal General Shape



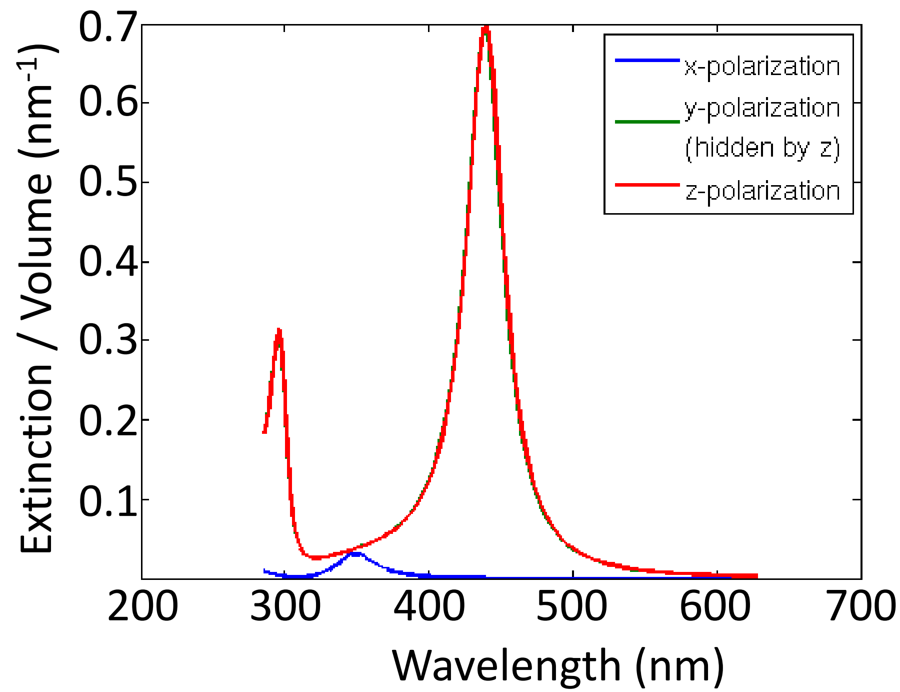


Roughly equal for all three polarizations

Optimal Ellipsoid

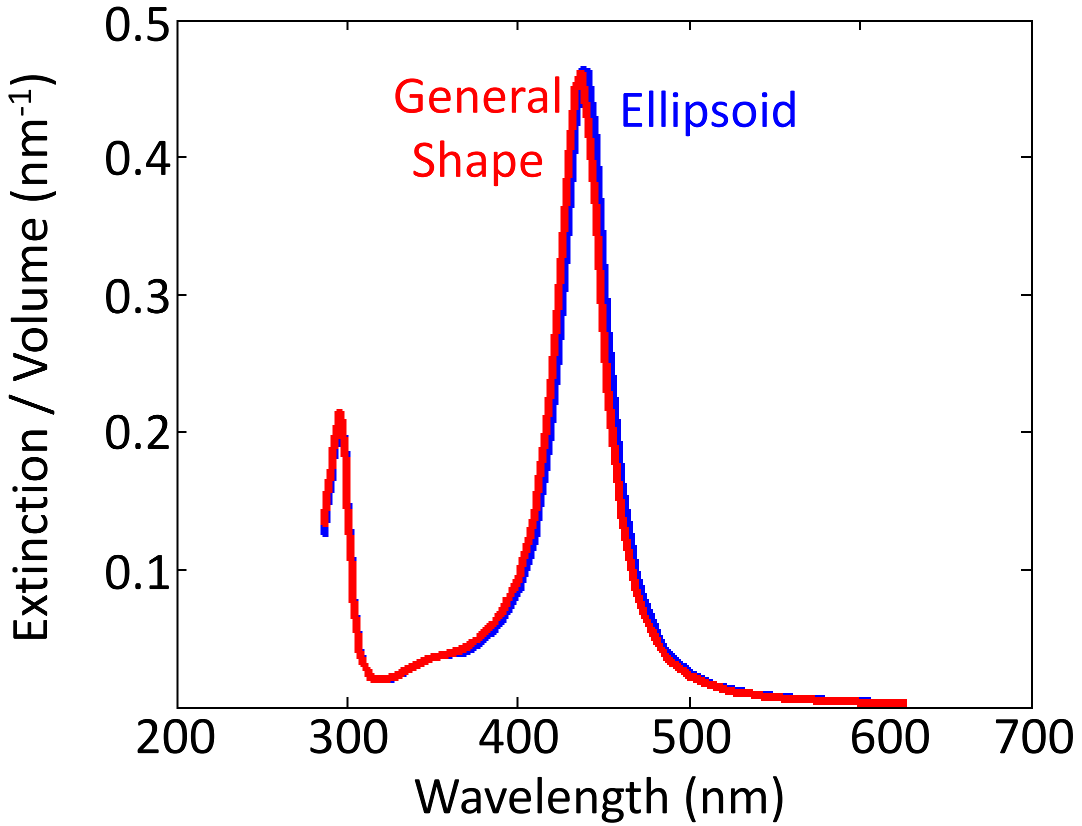


$r_1 = r_2 = 2$nm, $r_3 = 10$nm



y-, z-pol: very strong response
x-pol: very weak

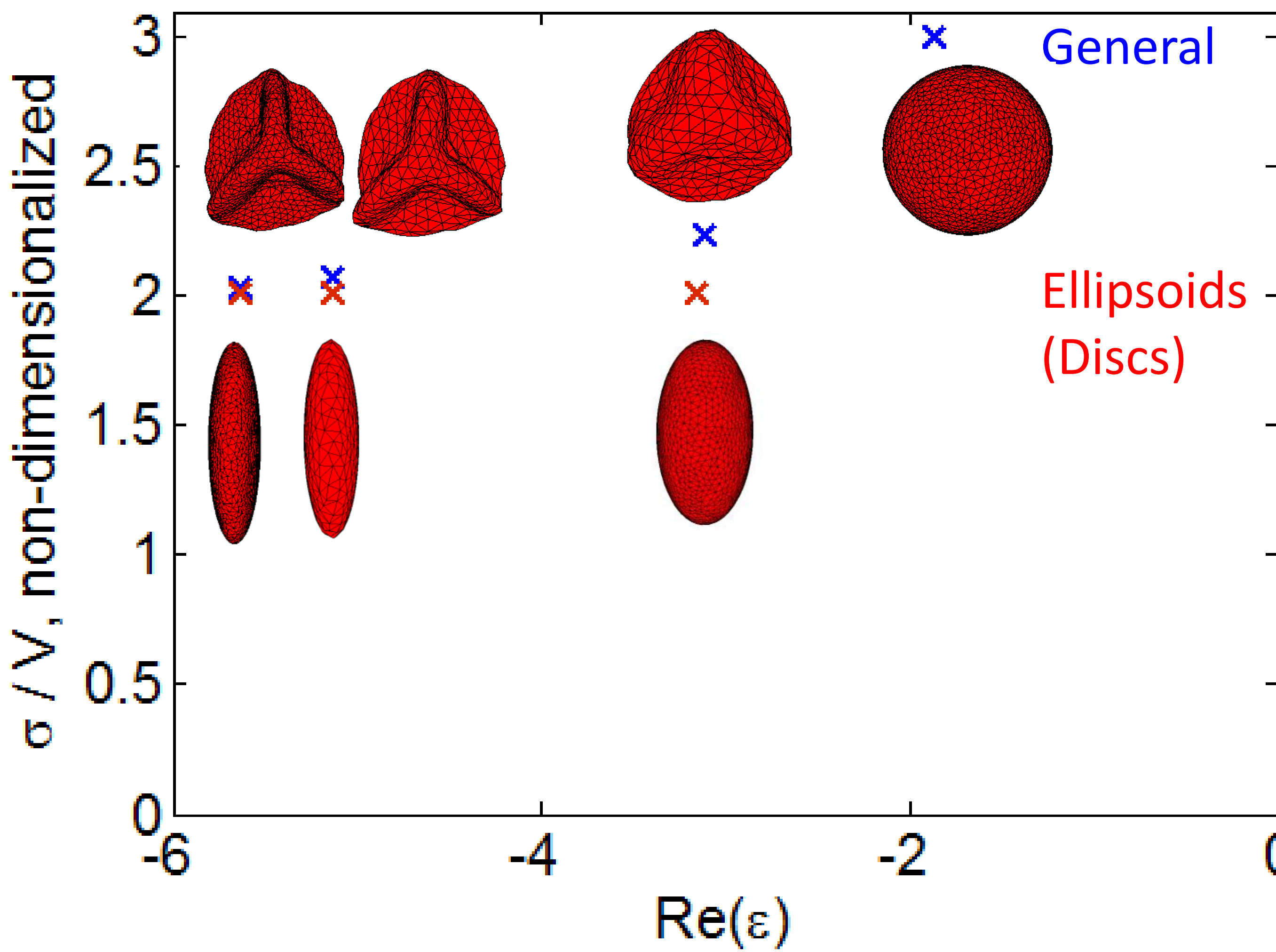
Optimal Structures Comparison: Total Response



Almost exactly the same?!

General shape optimum roughly 3% better than ellipsoidal optimum…

More Optimizations

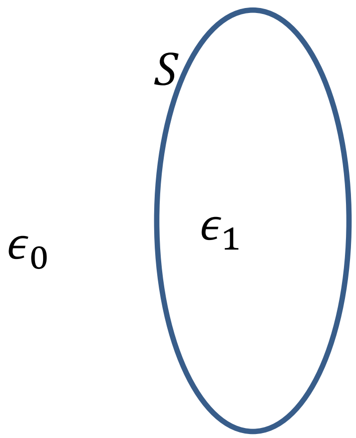


Quasi-Static Resonances

Solving Gauss' Law: $\nabla \cdot \epsilon E = \rho$

- Resonant with respect to what? There is no frequency.
- Resonance with respect to **permittivity**. For a given structure, there are specific (negative & real-valued) permittivities for which a surface charge can exist, *without an incident field*

- Mathematical formulation:



$$\nabla \cdot \epsilon(x) \nabla \phi(x) = -\rho_{ext}(x)$$

$$2\pi \frac{\epsilon_1 + \epsilon_0}{\epsilon_1 - \epsilon_0} \sigma(x) = n \cdot \nabla \phi^{\infty}(x) + \int_S F(x,y)\sigma(y)dy$$

$$F(x,y) = -\frac{n(x) \cdot (x-y)}{|x-y|^3}$$

for $\phi^{\infty} = 0$:

$$\boxed{\frac{\epsilon_n + 1}{\epsilon_n - 1} \sigma_n(x) = \frac{1}{2\pi} \int_S F(x,y)\sigma_n(y)dy}$$

Quasi-Static Resonances

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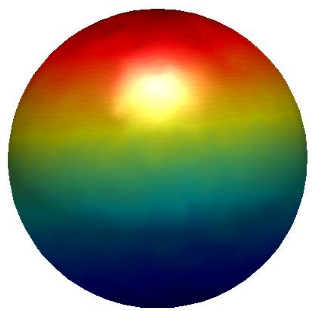
<u>*Spheres*</u>

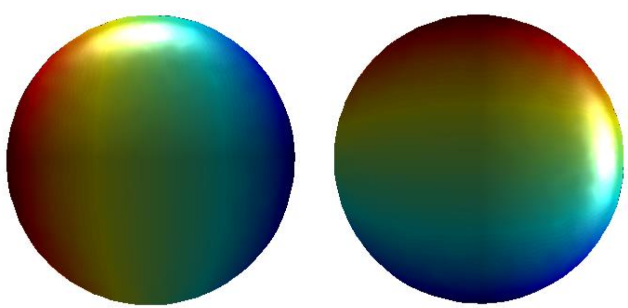
Three modes at
$\epsilon = -2$



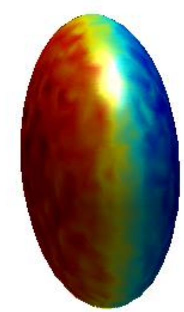
Each mode contributes to σ/V through dipole strength p_n and coupling $c_n = \left(\frac{1}{1-\epsilon_n} - \frac{1}{1-\epsilon}\right)^{-1}$

<u>*Ellipsoids*</u>

Two modes at
$\epsilon \approx -\frac{\pi}{4}\frac{r_{max}}{r_{min}}$



One mode at
$\epsilon \approx -\frac{8}{\pi}\frac{r_{min}}{r_{max}}$

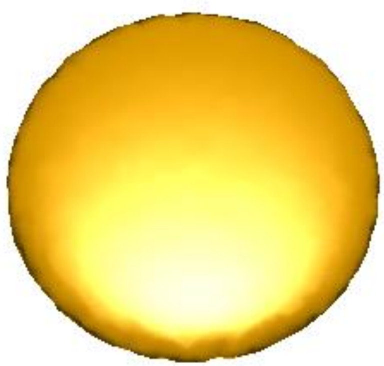


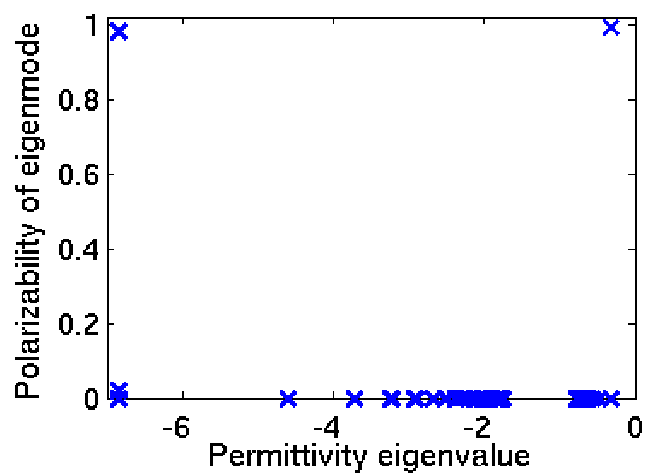
Images are surface charge densities of respective modes

Quasi-static particle design

- Suppose we want to **design** particle for maximum extinction at $\epsilon(\omega_0)$. How?
  (1) Want at least one resonance $\epsilon_n \approx Re[\epsilon(\omega_0)]$
  (2) Want polarizability concentrated at ϵ_n
  (i.e. should not be "wasted" at other permittivities)

- How does an oblate spheroid look, from this perspective?





Two dipole modes at
$\epsilon_1 = \epsilon_2 = -6.9$

One dipole mode at
$\epsilon_3 = -0.3$

So we have two strong, degenerate resonances.
But we're losing a significant part of polarizability at -0.3, right? Not quite.

Sum rules

- Fuchs (1975-1976), for collections of homogeneous particles:
  - #1: sum rule on polarizabilities

$$\sum_{modes\, n} p_{n,\alpha} = 1 \qquad \alpha = x, y, z$$

  - #2: weighted sum of resonant permittivities

$$\sum_{modes\, n} \frac{1}{1-\epsilon_n}\overline{p}_n = \frac{1}{3}$$

$\overline{p}_n$ = avg. $p_{n,\alpha}$ for all x,y,z

This is the crucial sum rule *(barely noticed in literature).*
Very roughly speaking, weighted average of ϵ_k has to $= -2$

- The primary upper bound discussed in the literature has been the *integrated extinction per volume*:

$$\int \frac{\sigma_{ext}(\lambda)}{V} = \pi^2 Tr[\gamma]$$

$\gamma = static\ polarizability$

But this is **shape-dependent**, does not provide a general limit

Quasi-static upper bound

- Constrained optimization:

Maximize $$\frac{\sigma(\omega_0)}{V} = \frac{1}{3}\frac{\omega_0}{c}\sum_n \left(\frac{1}{1-\epsilon_n} - \frac{1}{1-\epsilon(\omega_0)}\right)^{-1} p_n$$

Subject to (1-3) $$\sum_{modes\ n} p_{n,\alpha} = 1 \qquad \forall\alpha \in [x,y,z]$$ Sum Rule #1

(4) $$\sum_{modes\ n} \frac{1}{1-\epsilon_n}\overline{p}_n = \frac{1}{3}$$ Sum Rule #2

(5) $\epsilon_n < 0$ Uniqueness Theorem

Quasi-static upper bound

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(4) $\sum_{modes\ n} \frac{1}{1-\epsilon_n}\bar{p}_n = \frac{1}{3}$ Sum Rule #2

(5) $\epsilon_n < 0$ Uniqueness Theorem

- For $\epsilon(\omega_0) < -2$:

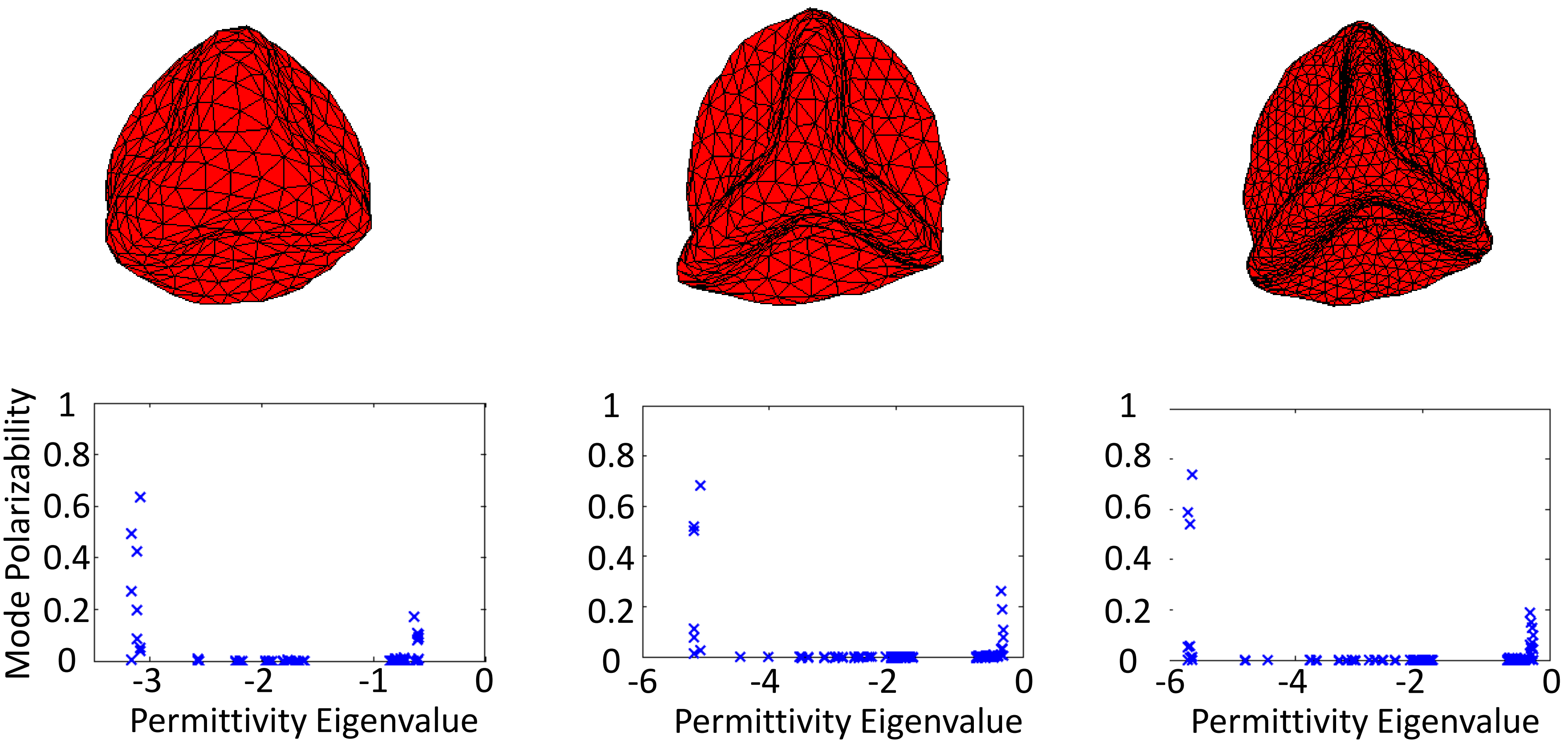
For a material permittivity $\epsilon = \epsilon_r + i\epsilon_i$ and susceptibility $\chi = \epsilon - 1$:

$$\frac{\sigma_{ext}(\omega_0)}{V} \leq \frac{2}{3}\frac{\omega_0}{c}\frac{|\chi|^4}{(|\chi|^2 + \chi_r)\chi_i}$$

$$\approx \frac{2}{3}\frac{\omega_0}{c}\frac{\epsilon_r^2}{\epsilon_i} \qquad (|\epsilon_r| \gg |\epsilon_i|, 1)$$

Given material and frequency, this bounds extinction for ***any possible shape***!

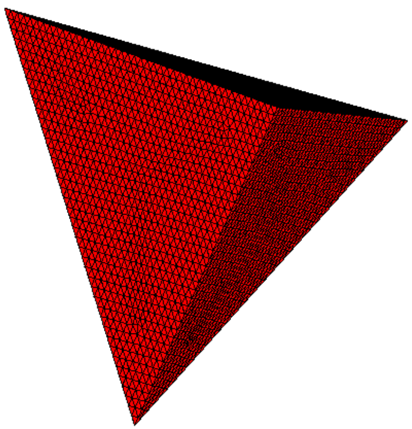
What do the optimal structures have in common?



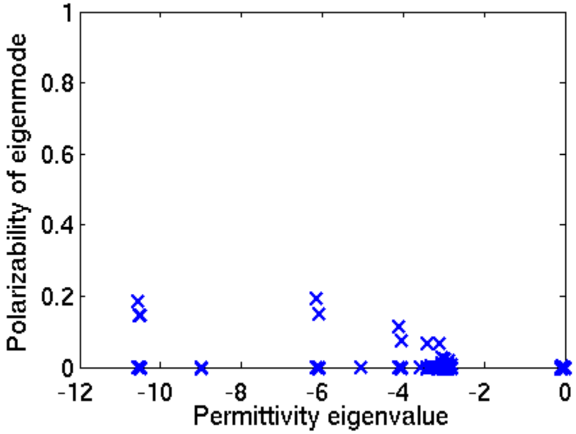
They all have roughly degenerate eigenmodes at the desired permittivity; all other modes have *zero dipole moment*, except as required by sum rules

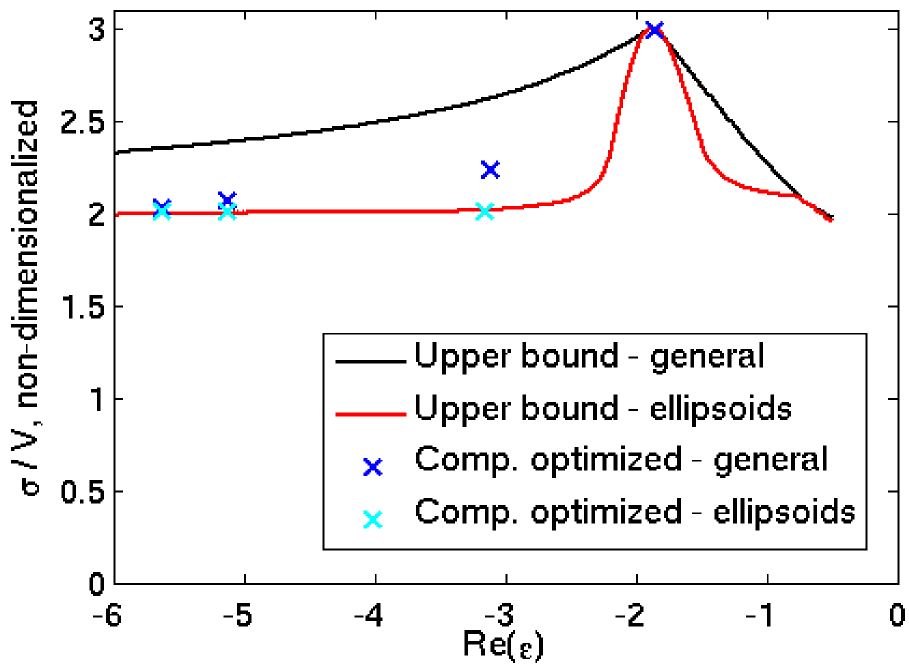
All mode calculations courtesy of excellent MNPBEM Toolbox: http://physik.uni-graz.at/~uxh/mnpbem/mnpbem.html

whereas
e.g. a tetrahedron



has resonances:





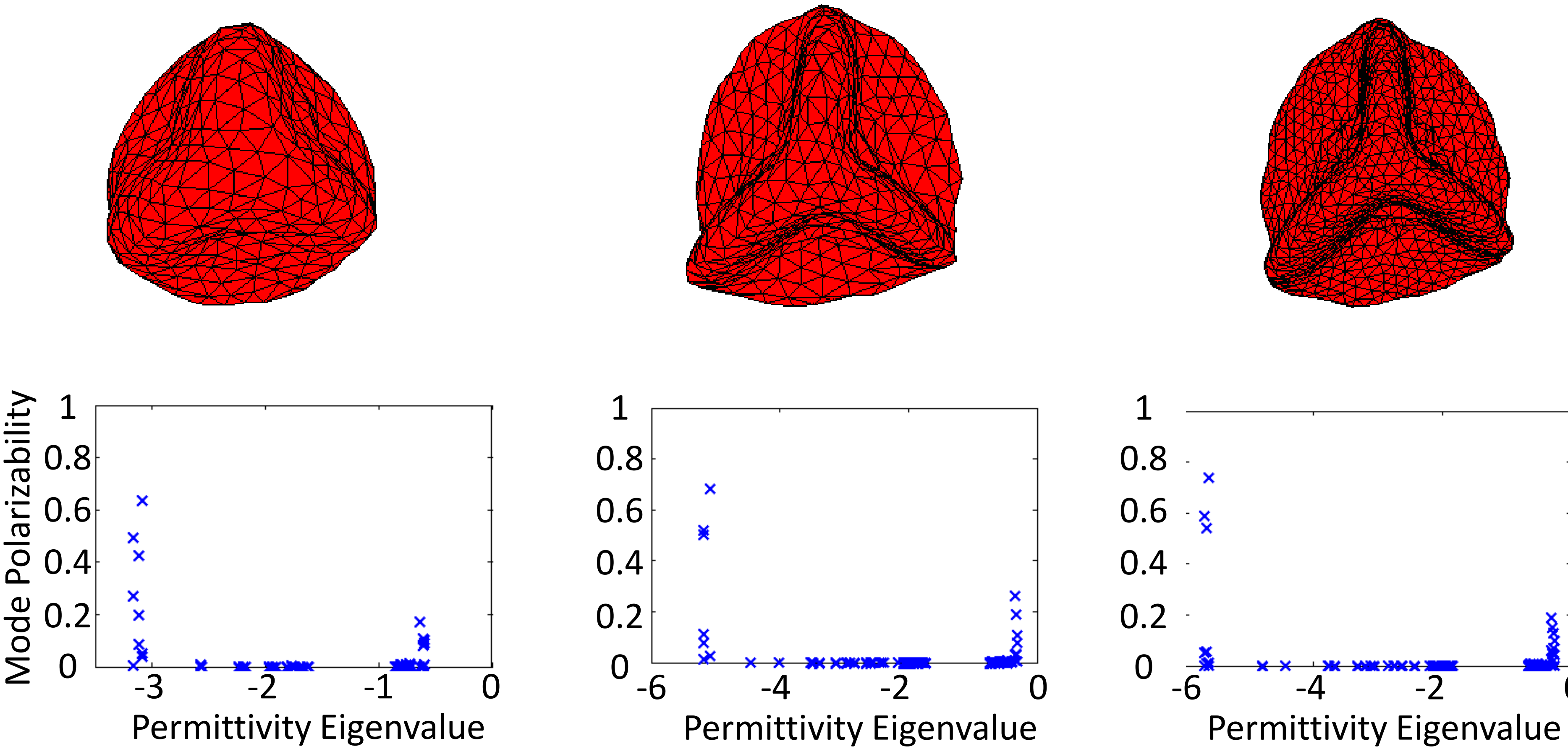
The left-hand side can roughly be interpreted as:

of "full-strength" resonances (ideal max. 3, for 3 polarizations)

About the general upper bound:

- Ellipsoids are nearly optimal; they even hit the upper bound in three limits ($\epsilon \to -\infty$, $\epsilon = -2$, $\epsilon \to 0$)
- There are structures that do better than ellipsoids; can possibly take manufacturability into account
- From the optimizations, perhaps the "true" upper bound is even closer to the ellipsoid than the one derived here

What do the optimal structures have in common?

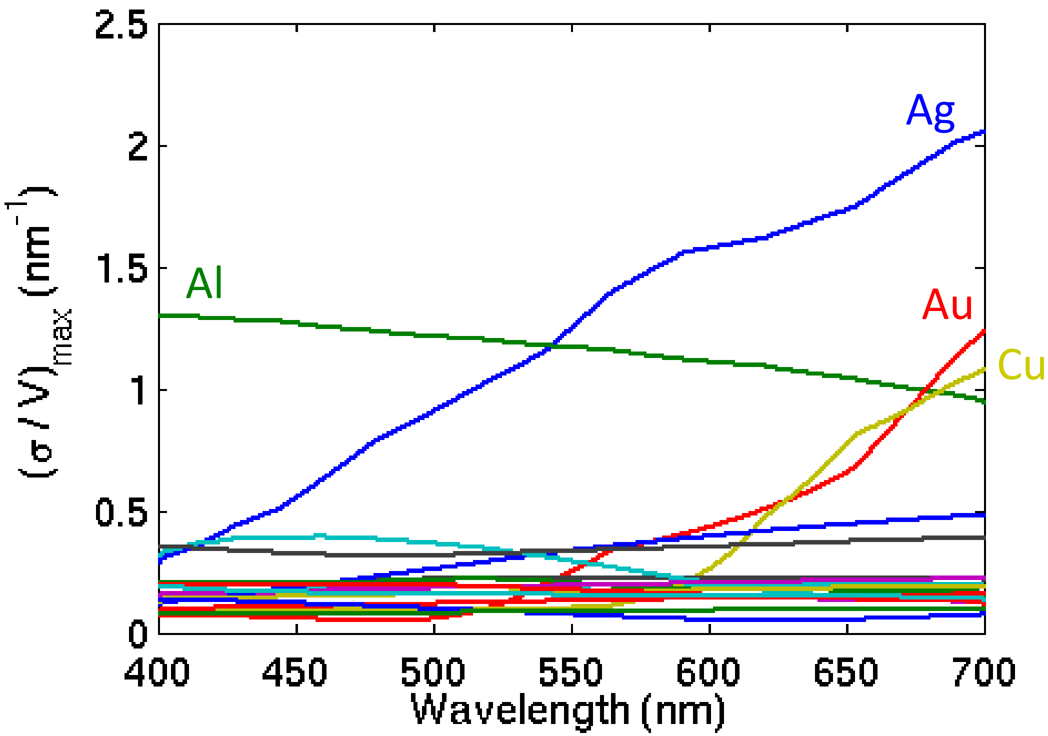


They all have roughly degenerate eigenmodes at the desired permittivity; all other modes have *zero dipole moment*, except as required by sum rules

All mode calculations courtesy of excellent MNPBEM Toolbox: http://physik.uni-graz.at/~uxh/mnpbem/mnpbem.html

Ideal Materials

Upper bound allows *shape-independent* calculation of ideal materials

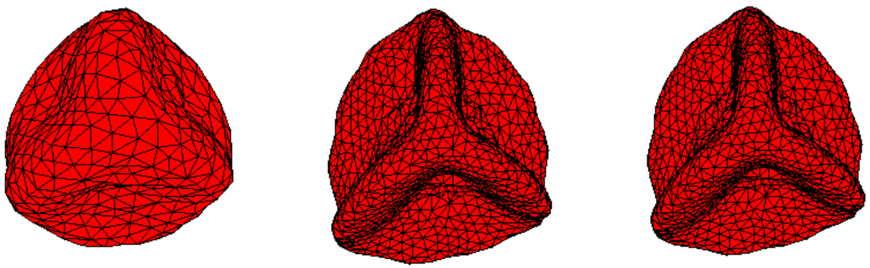


The lines that are not labeled:
Cr, Co, Ir, Li, Mo, Ni, Os, Pd, Pt, Rh, Ta, Ti, W, V

material data: refractiveindex.info (Palik, Rakic, Sopra-SA)

Key Points

- Moving from spheres to ellipsoids: ~**6x** improvement
- Moving from ellipsoids to arbitrary shapes: **<1.34x** improvement even theoretically possible
- There are shapes superior to ellipsoids; can be found through computational shape optimization



New, Quasi-Static Extinction Limit

$$\frac{\sigma(\omega_0)}{V} \leq \frac{2}{3}\frac{\omega_0}{c}\frac{|\chi|^4}{(|\chi|^2 + \chi_r)\chi_i}$$

Ideal Materials for the Visible