3-Dimensional Eigenmodal Analysis of Plasmonic Nano-Structures: with a Focus on Molecular Nanosensors

Hua Guo, ETH Zürich
Benedikt Oswald, PSI
Peter Arbenz, ETH Zürich
Goal: 3-dimensional eigenmodal analysis of nano-structures.

Numerical code: Femaxx.

Parallelized for distributed memory clusters.

Finite element method (FEM) in tetrahedral mesh.
- arbitrary geometry
- arbitrary material arrangement
- arbitrary dispersion property

Eigensolvers
- Jacobi-Davidson QZ (JDQZ) method
- Nonlinear Jacobi-Davidson (NLJD) method

Post-processing
- resonance
- electric/magnetic field plots
- stored energy
- radiation
- dissipation
- quality factor
Numerical formulation

- $\Omega_1$: nanostructure.
- $\Omega_2$: substrate.
- $\Omega_3$: environment.
- $\Omega = \Omega_1 \cup \Omega_2 \cup \Omega_3$: computational domain.
- $\Gamma$: boundary.
- To reduce error, let $\Gamma$ be sphere.

**Time-harmonic electric field curl-curl equation**

$$\nabla \times (\mu_r^{-1} \nabla \times E(x)) - k_0^2 \varepsilon_r E(x) = 0, \quad \nabla \cdot (\varepsilon_r E(x)) = 0, \quad x \in \Omega$$

**subjected to the 1st order absorbing boundary condition**

$$n \times (\nabla \times E(x)) = -i k_0 \sqrt{\mu_r \varepsilon_r} n \times (n \times E(x)), \quad x \in \Gamma$$

$k_0$: complex wavenumber  
$\mu_r$: magnetic relative permeability  
$\varepsilon_r$: electric relative permittivity
Numerical formulation

$\varepsilon_r$ of nanostructure

- complex-valued
- varies w.r.t. frequency (dispersion)

Figure: $\varepsilon_r$ for gold.

**Numerical formulation**

### Constrained nonlinear eigenvalue problem

Discretized by 3-D FEM yields

\[
T(\lambda)x = Ax + \lambda Rx - \lambda^2 Mx = 0 \\
C^T x = 0.
\]

- \(\lambda\) is the eigenvalue; \(x\) is the eigenvector.
- \(A, R\) are constant matrices.
- \(M, C\) depend on \(\text{Re}(\lambda)\).

### Two-loops eigensolver

- outer loop: construct a sequence of quadratic eigenproblem (QEP).
- inner loop: solve each QEP by JDQZ/NLJD.

Numerical formulation

1. resonance: $\omega = \text{Re}(\lambda) c$

2. Q factor: $Q = \frac{\text{Re}(\lambda)}{2 \text{Im}(\lambda)}$

3. electric/magnetic field

4. radiation power: $P_r = \frac{1}{2} \int_\Gamma \text{Re}(\mathbf{E}(x) \times \mathbf{H}(x)^*) \cdot d\mathbf{x}$

5. dissipation energy: $U_d = \frac{\varepsilon_0}{2} \int_{\Omega_1} \text{Im}(\varepsilon_r) |\mathbf{E}(x)|^2 d\mathbf{x}$

6. radiative quantum yield: $\eta = \frac{P_r}{(P_r + \omega U_d)}$

7. stored energy for dispersive medium (under debate):
   
   $U_s = \frac{\varepsilon_0}{4} \int_{\Omega} \left(\text{Re}(\varepsilon_r) + \omega \frac{d(\text{Re}(\varepsilon_r))}{d\omega}\right) |\mathbf{E}(x)|^2 d\mathbf{x} + \frac{\mu_0}{4} \int_{\Omega} |\mathbf{H}(x)|^2 d\mathbf{x}$
Example 1: spherical nanoparticle

(a) dipolar mode:
\[ \lambda_{\text{res}} = 408.9 \text{ nm}, \quad Q = 6.0. \]

(b) quadrupolar mode:
\[ \lambda_{\text{res}} = 356.0 \text{ nm}, \quad Q = 8.1. \]

**Figure:** electric field distributions of a silver sphere with radius= 60 nm.
Example 2: optical nano-antennas

(a) geometry

(b) $|\mathbf{E}|$ in gap:
\[ \lambda_{\text{res}} = 1140 \text{ nm} \]

(c) $|\mathbf{E}|$ in gap:
\[ \lambda_{\text{res}} = 989 \text{ nm} \]


(d) geometry (top view)

(e) $|\mathbf{E}|$: $\lambda_{\text{res}} = 734 \text{ nm}$
Example 3: Fano resonance nanostructures

Fano resonance arises from the interference between radiative/non-radiative modes.

(f) gold dolmen nanostructure: 
\( \lambda_{res} = 1221 \text{ nm} \)

(g) heptamers of silver spheres: 
\( \lambda_{res} = 502 \text{ nm} \)
Molecular nanosensor: a dipole nano-antenna

(h) geometry

(i) charge profiles
(j) dark mode: gap = 10 nm, resonance at 557.4 nm

(k) bright mode: gap = 20 nm, resonance at 665.7 nm

(l) $|E|$ along x-axis: gap = 5 nm
Table: Dark modes vs. bright modes. The dipole antenna is in vacuum.

<table>
<thead>
<tr>
<th>gap width (nm)</th>
<th>dark mode</th>
<th>bright mode</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\lambda_{\text{res}}$ (nm)</td>
<td>$\eta$</td>
</tr>
<tr>
<td>20</td>
<td>582.0</td>
<td>$2.4 \times 10^{-3}$</td>
</tr>
<tr>
<td>10</td>
<td>557.4</td>
<td>$7.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>5</td>
<td>532.1</td>
<td>$2.3 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

Remark
- With decreasing gap, **bright mode** shifts to red, **dark mode** shifts to blue.
- **Dark mode** radiates scant energy to far field (small $\eta$).
- **Dark mode** has lower Q factor, due to higher loss in dissipative dipoles.
Figure: Sketch of the molecular nanosensor with top view (left) and the cross section (right) view. Yellow area: the dipole antenna (gap width is 10 nm); Gray area: the silica substrate. Light gray area: a molecule layer.
Molecular nanosensor: sensitivity

Figure: The resonance $\lambda_{res}$ (bright mode) vs. the refractive index $n$ of the molecule layer. The layer thickness is 5 nm.

Remark

- Bright mode is suitable for sensing.
- Sensitivity: 172 nm per unit of refractive index (RIU).
Molecular nanosensor: sensitivity

(a) The sensitivity (nm/RIU) of the nanosensor vs. the layer thickness $t$.

(b) Electric field plot. $t = 40$ nm.

Remark

- Sensitivity improves as $t$ increases.
- e.g., if $t = 40$ nm, the sensitivity is nearly 300 nm/RIU.
- If surrounding environment is all filled with molecules ($t \to \infty$), the sensitivity is 404 nm/RIU.
- Higher than many single nanoparticles.
1. Electromagnetic eigenmodal tool Femaxx.

2. Examples of several nanostructures (nano-sphere, nano-antennas, fano resonance structures).

3. Simulation of a dipole antenna for molecular nanosensing.
Acknowledgement

1. Suggestions from Prof. Olivier Martin.
2. Swiss National Supercomputing Centre (CSCS).
3. Gmsh group.
4. Project (ID:200021-117978) is sponsored by Swiss National Science Foundation.