On the numerical modeling of sharp metallic tips

International Conference from Nanoparticles and Nanomaterials to Nanodevices and Nanosystems (IC4N)
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Overview

‣ Illustrate some computational approaches ...
  ‣ Multiple Multipole Program
  ‣ Finite Element Analysis of the (Quasi-)Static Formulation
  ‣ Discontinuous Galerkin Discretization in the Time-Domain

‣ ... and Applications
  ‣ Nanotips for SNOM or TERS
  ‣ Nanoparticles as Probes for Sample Scanning
  ‣ Near-Field Scanning Microwave Microscope
  ‣ Laser induced Electron Emission from Nanotips
Multiple Multipole Program

- MMP: Efficiency proven method for EM problems.
  - Scatterers mimicked by point sources that fulfill the BCs.
  - Multipoles can be of, e.g., point- or line-type

**Example** Scattering analysis by MMP

\[ \mathbf{k}_{\text{inc}} = k_{\text{inc}} \mathbf{e}_x + k_{\text{inc}} \mathbf{e}_y \]

+ Standard multipoles for fields in \( D_2 \)

\( x \) Standard multipoles for fields in \( D_1 \)

\( \cdots \) Matching points

\[ \text{Field}_i = \sum_{n=1}^{N} A_n^i E_n^i + \text{error} \]

Metallic wire tip modeling for a SNOM, TERS, etc.

Upper part
- Incident TM0: connection
- Reflected modes: connect.
- Remaining field: ring or complex origin multipoles

Fictitious boundary

Lower part
- Standard MMP modeling of axisymmetric structures using ring or complex origin multipoles
- Refinement towards tip

«Sommerfeld» TM0 mode has relatively low loss and provides nice focusing at tip
- Excitation requires discontinuity (may be away from tip)
Upper part
- Incident TM0: connection
- Reflected modes: connect.
- Remaining field: ring or complex origin multipoles

Fictitious boundary

Lower part
- Standard MMP modeling of axisymmetric structures using ring or complex origin multipoles
- Refinement towards tip

• Advantages of MMP over, e.g., Finite Differences Methods (FDTD)
• Boundaries are exact (no staircasing!)
• Built-in error estimate as mismatch of boundary condition
Model of a parabolic Mirror SNOM

Radially polarized beam is focused by a parabolic mirror on a metallic tip.

Model similar, except for excitation:
- Bessel expansion of the incident field

Example:
- Silver tip
- Diameter 3 µm
- Wavelength 633 nm
- Scatt field shows outgoing SPP
Model of a parabolic Mirror SNOM

Characteristics:
- Field is strongly confined below the tip
- Standing wave pattern -> excitation of counter propagating SPPs

Time averaged scattered field
(factor of sqrt(2) between adjacent contour lines)
Au tip @633 nm

Parameters:
Field 1nm below the tip
Tip radius: 5nm
Wire diameter: 3µm
Tip angle: 15°, 25°

Study of different tip materials:
- Plasmon resonance can lead to an increased enhancement (Ag)
- Field enhancement strongly geometry dependent
MMP and layered media Green’s functions

- MMP: Efficiency proven method for EM problems.
  - Scatterers mimicked by point sources that fulfill the BCs.

**Example** Scattering analysis by MMP

\[ \mathbf{k}_{\text{inc}} = k_{x,\text{inc}} \mathbf{e}_x + k_{y,\text{inc}} \mathbf{e}_y \]

- Layered media monopoles for fields in \( D_2 \) \( \ldots \) \( D_n \)
- Standard multipoles for fields in \( D_1 \)
- Matching points

\[ \text{Field}_i = \sum_{n=1}^{N} A_n^i E_n^i + \text{error} \]

Layered media Green’s functions

- The Sommerfeld integration is needed.
  - For a point source in z-direction where the layers change in y-direction the Green’s functions are obtained by:

<table>
<thead>
<tr>
<th>In 3D (most general):</th>
<th>Text</th>
</tr>
</thead>
<tbody>
<tr>
<td>( G_i(x, y, z) = \frac{1}{\delta} \int_{-\infty}^{\infty} dk_x dk_z e^{ik_x x} e^{ik_z z} \tilde{G}_i(k_x, k_z, y) )</td>
<td>Free-space wave nature: ( \tilde{G}_i(k_x, k_z) \propto A \frac{e^{ik_y y}}{2ik_y} )</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>In 2D:</th>
<th></th>
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</thead>
<tbody>
<tr>
<td>( G_i(x, y) = \frac{1}{\delta} \int_{-\infty}^{\infty} dk_x e^{ik_x x} \tilde{G}_i(k_x, y) )</td>
<td>( G_i(x, y) \propto AH_0^{(1)}(k\hat{n}) )</td>
</tr>
</tbody>
</table>

\( r \): distance between source and observation points
\( \rho \): distance between source and observation points

Analytic (exact) solutions used in the numerical description --> highly accurate solutions
Scattering analysis by MMP (3D)

- The scattering cross section of a particle in a layered geometry

\[ \int_S (E \times H^*) \times dS \]
Scattering analysis by MMP (3D)

- **Ag sphere and Ag spacer**
  - Free space
  - $t=0$ nm
  - $t=5$ nm
  - $t=10$ nm
  - $t=18$ nm
  - $t=50$ nm

- **Au sphere with Ag spacer**
  - Free space
  - $t=0$ nm
  - $t=5$ nm
  - $t=10$ nm
  - $t=18$ nm
  - $t=50$ nm

- **Au sphere with Au spacer**
  - Free space
  - $t=0$ nm
  - $t=5$ nm
  - $t=10$ nm
  - $t=18$ nm
  - $t=50$ nm

Graphs show variations in back scattering cross section with wavelength for different spacings ($t$) and materials (Ag and Au) between the sphere and the spacer.
Scattering analysis by MMP (3D)

- An example  Peak 1 @ 444nm

![Graph showing scattering analysis with peaks at 444nm]
Scattering analysis by MMP (3D)

- An example  Peak 2 @ 396.5nm
Scattering analysis by MMP (3D)

- An example: Peak 3 @ 372.5nm

The graph shows the backscattering cross section as a function of wavelength for different thicknesses of Ag spacer: 0nm, 5nm, 10nm, 18nm, and 50nm. The graph compares the results with free space and highlights the peak at 372.5nm.
Scattering analysis by MMP (3D)

- An example  Peak 1 @ 360nm
General Setup of a Near-Field Scanning Microwave Microscope

- Generate strong field concentration in small region

Metallic Tip

$r \ll \lambda$

Resonator

Measure detuning with VNA
Some Typical Measurements

Frequency Shift we want to model!
Modeling a Rounded Cone as Tip

- Wavelength much larger than the tip
- Solve as static problem using Finite Element Method (FEM)
- Modify material properties
Validation against Analytic Results (Sphere)
Results for a Conical Tip (Compared to Sphere)

![Graph showing relative shift vs gap size for different tip radii](image-url)
Laser induced Electron Emission from Nanotips

- Laser induced electron emission from nano-tips currently is a hot topic
- Goal is to be able to control the emission of electrons on the nanometer, femtosecond and sub-electronvolt level
- Such electrons could be used for investigating, e.g., intramolecular dynamics and other ultra-fast processes
- Collaboration with H. Yanagisawa (HY) from the Institute for Quantum Electronics at the Physics Dept., ETH Zurich
Background and Goals

- The **focus** of this talk is rather on giving an **introduction** into some **aspects of simulations techniques** than presenting the latest advances.

- Laser induced electron emission from nano-tips is a good example for this as it includes many aspects:
  - Static fields
  - **Time-dependent fields** (in particular focused laser beams)
  - Charged particle dynamics
    - emission
    - tracking in external and self fields
Experimental Setup (qualitative)

Pre amplifier and Position computer

Beam Shrinker and Beam expander

Electrode

Laser

Oscillator 800 nm 76 MHz, 55 fs

Laser Polarization

Magnifier for emission sites and energy spectroscopy

Resistive anode

MCP (Chevron)

Tip

Hy, PRL, 2009

PC

Lens

$f = 18$ mm

$\theta_p$

Heating

High voltage (negative)
Experimental Setup (qualitative)

Laser field influences electron dynamics depending on laser phase at emission
- Acceleration
- Deceleration
- Rescattering


Experimental Setup (qualitative)

In HY’s experiment these curves show differences and he assumes some physics to be going on...

Electron count rate vs. electron energy for various laser intensities
Subproblems

1) Modeling the static field
2) Modeling and simulating the laser
3) Modeling the electron emission and simulating the particle dynamics
1) Modeling the static field

Use mirror charges for representing the counter electrode.

Represent field by a set of monopoles (point charges).

Tip

$r = 100 \text{ nm}$

Counter electrode

$1500 \text{ V}$
1) Modeling the static field

Sanity check:
- Place two point charges
- Electrons should travel from one charge to the other
- and gain energy (color coding)
2) Modeling and simulating the laser

- **Focused laser beams** often modeled as **Gauss beams**
- Based on paraxial approximation and **no proper solution** of Maxwell’s equations
- **Problematic** to apply non-proper solutions in simulation tools

Gauss beam: $\lambda = 30 \, \mu m$, $w = 40 \, \mu m$
2) Modeling and simulating the laser

Instead derive a rigorous solution of Maxwell’s equations

\[
E_r = \frac{n(n + 1) \cos m \cos^{-1}\left(\frac{x - xs}{\sqrt{(x - xs)^2 + (y - ys)^2}}\right)}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}} \left[j_n \left(\frac{z - zs}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}\right) - k (y - ys)^2\right]
\]

\[
E_\varphi = \cos \left\{m \cos^{-1}\left(\frac{x - xs}{\sqrt{(x - xs)^2 + (y - ys)^2}}\right)\right\} \left[j_{n-1} \left(\frac{z - zs}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}\right) - n j_n \left(\frac{z - zs}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}\right)\right]
\]

\[
E_\theta = \frac{n(z - zs) P_n^m}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}} \left[(m + n) \sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2} P_{n-1}^m\right]
\]

\[
H_r = \frac{0(m, n, x, y, z)}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}
\]

\[
H_\varphi = \frac{i m \omega \epsilon \sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2} \sin \left\{m \cos^{-1}\left(\frac{x - xs}{\sqrt{(x - xs)^2 + (y - ys)^2}}\right)\right\} \left[j_n \left(\frac{z - zs}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}\right) - k (y - ys)^2\right]}{\sqrt{(x - xs)^2 + (y - ys)^2}}
\]

\[
H_\theta = \frac{i \omega \epsilon \cos \left\{m \cos^{-1}\left(\frac{x - xs}{\sqrt{(x - xs)^2 + (y - ys)^2}}\right)\right\} j_n \left(\frac{z - zs}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}\right)}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}}
\]

\[
H_z = \sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2} \left[\frac{n(z - zs)}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}} P_n^m\right]
\]

\[
\frac{1}{\sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2}} \left[(m + n) \sqrt{(x - xs)^2 + (y - ys)^2 + (z - zs)^2} P_{n-1}^m\right]
\]
2) Modeling and simulating the laser

- Instead derive a rigorous solution of Maxwell’s equations
- Real origin:

![Radiating point source](image-url)
2) Modeling and simulating the laser

- Instead derive a rigorous solution of Maxwell’s equations
- Complex origin (and complex arithmetic):

![Diagram comparing Complex Bessel Beam and Gauss Beam, showing difference larger for stronger focusing.](image-url)
2) Modeling and simulating the laser

In a time-domain simulation feed the laser field into the simulation domain at boundaries (port feeding). Construct pulse from laser spectrum.

Focused laser pulse

2D cut view but 3D simulation

Color code: Electric field magnitude
2) Modeling and simulating the laser

- In a **time-domain simulation** feed the laser field into the simulation domain at boundaries (**port feeding**)
- Simulation employs the **Discontinuous Galerkin Method** (a Finite Element-type method very suitable for time-domain simulations)

\[ U_i(x, t^n) = \sum_{p=0}^{P_i} c_i^p(t^n) \varphi_i^p(x) \]

**Much higher accuracy than FDTD**
3) Modeling/simulating electron emission and dynamics

- **Emission** is a **stochastic process** and depends on the laser **field strength** and local **work function**.

- **Crystal structure**
- **Electric (laser) field at surface**
- **Emission probability**
3) Modeling/simulating electron emission and dynamics

- Simulation of (relativistic) **electron dynamics**

**Time step Maxwell’s equations**

- Compute particle interaction
- Compute force from fields

**Emit and time step particles**
3) Modeling/simulating electron emission and dynamics

with $e^-e^-$ interaction

w/o $e^-e^-$ interaction

VELOCITY Magnitude

5.3e+8
5e+8
4e+8
3e+8
2e+8
1e+8
5e+7
Multiscale-character

- The Multiscale-problem of the setup
  - Tip radius 100 nm
  - Counter electrode distance 13 mm
  - Time step required for accurate particle dynamics ~50 asec --> 1 Million time steps
  - Laser center wavelength 830 nm
  - Laser beam waist 2 µm

- Restrict laser simulation to the tip area

- Employ adaptive time integration for the particles
  (time step not fixed but gets large when electrons move away from the singularity/strong field at the tip)
Putting it all together
Results

- First question: How many electrons were emitted in the experiment in one shot?
  - Perform simulations with a mean number of 2, 10, 100 electrons
  - To get statistics right
    - 500,000 simulations 2 e⁻
    - 100,000 simulations 10 e⁻
    - 50,000 simulations 100 e⁻
  - --> around 2 e⁻

Postprocessing and visualization by HY
Results

- Investigate more carefully around 2 e⁻
- 500,000 simulations each

1 e⁻  
2 e⁻  
3 e⁻

leading e⁻  
Energy gain  

trailing e⁻  
Energy loss  

Postprocessing and visualization by HY
Outlook

‣ Instead of an outlook...

‣ If you have problems that interesting for simulation, contact me
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