A study of linear dispersive models for nanoplasmonics.

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Workshop Numerical Waves, Nice
Outline

1 Motivations

2 Modelling equations: one approach

3 Theoretical study

4 Numerical framework
   • Academic context
   • Is the model with $\beta \neq 0$ physically relevant?

5 Validity of the model
Motivations

Control of the interaction of light with nano-scaled structures.

Nanophotonics
Motivations

Control of the interaction of light with metallic nano-scaled structures.

Nanoplasmonics
Control of the interaction of light with metallic nano-scaled structures.\hfill $\Rightarrow$ Nanoplasmonics

Light + subwavelength metallic structures
\downarrow
Collective oscillations of the electrons of the metal.
\downarrow
Plasmons
Motivations

Surface plasmons

Bulk plasmons

Gap plasmons
Optical frequencies: $\approx [300\text{nm}, 700\text{nm}]$.

Size of the nano-structuration $< \lambda$.

Subwavelength phenomena.
Motivations

Interaction of electromagnetic waves with complex heterogeneous media.

Metals at the nanoscales at optical frequencies:

\[ \rightarrow \text{Computational nanophotonics.} \]
Motivations

Interaction of electromagnetic waves with complex heterogeneous media.

Metals at the nanoscales at optical frequencies:

⇝ Computational nanophotonics.

Challenges in this context

- Geometrical characteristics of the physical domain.
- Physical characteristics of the propagation medium.
- Need of numerical accuracy
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Modelling equations

Free electrons of the metal $\leadsto$ electron gas.

Hydrodynamic description ( $v$: speed, $n$: density. )

$$m \left( \frac{\partial}{\partial t} + v \cdot \nabla \right) v = -e(E + v \times B) - m\gamma v - \frac{m\beta^2}{n} \nabla n$$

$$\frac{\partial}{\partial t} n + \text{div} (n v) = 0$$

$$J = -en v$$

+ Time Domain Maxwell’s equations ($J$, $E$ and $B$).
Modelling equations

Free electrons of the metal $\rightsquigarrow$ electron gas.

Hydrodynamic description ( $v$: speed, $n$: density. )

\[
m \left( \frac{\partial}{\partial t} + v \cdot \nabla \right) v = -e (E + v \times B) - m\gamma v - \frac{m\beta^2}{n} \nabla n
\]

\[
\frac{\partial}{\partial t} n + \text{div} (n v) = 0
\]

\[
J = -en v
\]

+ Time Domain Maxwell’s equations ($J$, $E$ and $B$).

\[
\frac{m\beta^2}{n} \nabla n \rightsquigarrow \text{quantum pressure term.}
\]
Linearized Hydrodynamic model

Formal linearization around an equilibrium state \((n_0, v_0, E_0, B_0, J_0) = (n_0, 0, E_0, 0)\),

\[
\begin{aligned}
\varepsilon_0 \varepsilon_r \frac{\partial \mathbf{E}}{\partial t} &= \text{curl} \mathbf{H} - \mathbf{J} \\
-\mu_0 \frac{\partial \mathbf{H}}{\partial t} &= \text{curl} \mathbf{E} \\
\frac{\partial \mathbf{J}}{\partial t} &= \beta^2 \nabla Q - \gamma \mathbf{J} + \varepsilon_0 \omega_p^2 \mathbf{E} \\
\frac{\partial Q}{\partial t} &= \nabla \cdot \mathbf{J},
\end{aligned}
\]

Maxwell’s equations

\[
\begin{aligned}
\beta \neq 0 & \implies \text{PDE for polarization current} \\
\beta = 0 & \implies \text{ODE for polarization current}
\end{aligned}
\]

Charge preservation

\[
\text{div}(\varepsilon_0 \varepsilon_r \mathbf{E}) = -Q
\]
Modeling equations: one approach

\[
\begin{align*}
\varepsilon_0 \varepsilon_r \frac{\partial E}{\partial t} &= \text{curl } H - J \\
\frac{\partial H}{\partial t} &= \text{curl } E \\
-\mu_0 \frac{\partial J}{\partial t} &= \beta^2 \nabla Q - \gamma J + \varepsilon_0 \omega_p^2 E \\
\frac{\partial Q}{\partial t} &= \nabla \cdot J,
\end{align*}
\]

~~~

**Key remark.**

The energy

\[
\mathcal{E}(t) = \frac{1}{2} \varepsilon_0 \varepsilon_r \| E \|^2 + \mu \| H \|^2 + \frac{1}{\varepsilon_0 \omega_p^2} \| J \|^2 + \frac{\beta^2}{\varepsilon_0 \omega_p^2} \| Q \|^2
\]

is decreasing (if adequate BCs)

\[
\frac{d}{dt} \mathcal{E}(t) = -\frac{\gamma}{\varepsilon_0 \omega_p^2} \| J \|^2 \leq 0.
\]

**Well-posedness** with semi-group theory is at reach!
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Theoretical study

Synthetic formulation

\((\mathcal{L}, D(\mathcal{L})) \) unbounded operator, \( U = (E, H, J, Q)^T \).

\[
\begin{aligned}
\begin{cases}
\partial_t U = \mathcal{L} U, \\
U(0) = U_0,
\end{cases}
\end{aligned}
\]

\( \mathcal{L} = \mathcal{A} + \mathcal{K} + \mathcal{F} \)

- \( \mathcal{A} \) unbounded operator,
- \( \mathcal{K} \) bounded operator,
- \( \mathcal{F} \) bounded operator.

\[
\mathcal{A} U = \begin{pmatrix}
\varepsilon_0^{-1} \varepsilon_L^{-1} \text{curl} H \\
-\mu_0^{-1} \text{curl} E \\
\beta^2 \nabla Q \\
\nabla \cdot J
\end{pmatrix}
\]

\[
\mathcal{K} U = \begin{pmatrix}
-\varepsilon_0^{-1} \varepsilon_L^{-1} J \\
0 \\
\varepsilon_0 \omega_p^2 E \\
0
\end{pmatrix}
\]

\[
\mathcal{F} U = \begin{pmatrix}
0 \\
0 \\
-\gamma J \\
0
\end{pmatrix}
\]
Theoretical study

First study for perfect medium (joint work with S. Nicaise).

- \( \Omega \) Lipschitz open bounded simply connected domain \( \subset \mathbb{R}^3 \).
- Boundary conditions \( B_{\text{perfect}}(U) = 0 \): \( E \times n = 0, \; Q = 0 \) on \( \partial \Omega \)

\[
\mathcal{H} = \{ U \in H(\text{div}, \Omega) \times H_0(\text{div} = 0, \Omega) \times L^2(\Omega)^3 \times L^2(\Omega), \text{div}(\varepsilon \varepsilon_1) = -\mathcal{U}_4 \text{ on } \Omega \},
\]

\[
\langle U, U' \rangle_{\mathcal{H}} := \varepsilon_0 \varepsilon L \langle U_1, U'_1 \rangle + \mu \langle U_2, U'_2 \rangle + \frac{1}{\varepsilon_0 \omega_p^2} \langle U_3, U'_3 \rangle + \frac{\beta^2}{\varepsilon_0 \omega_p^2} \langle U_4, U'_4 \rangle
\]

- Hilbert space: \( (\mathcal{H}, \langle \cdot, \cdot \rangle_{\mathcal{H}}) \)
- Energy scalar product: \( \langle \cdot, \cdot \rangle_{\mathcal{H}} \)
- Domain \( D(\mathcal{L}) \) dictated by \( A \) and \( B_{\text{perfect}} \):

\[
\text{"} \{ U \in \mathcal{H}, \; AU \in L^2, \; B_{\text{perfect}}(U) = 0 \} \text{"}.
\]

Properties of the operators.

- \( A \) is skew-adjoint, \( \Re \langle AU, U \rangle_{\mathcal{H}} = 0 \),
- \( \Re \langle KU, U \rangle_{\mathcal{H}} = 0 \),
- \( \langle FU, U \rangle_{\mathcal{H}} = -\frac{\gamma}{\varepsilon_0 \omega_p^2} \| J \|^2 \leq 0 \).
Well-posedness (joint work with S. Nicaise\textsuperscript{1})

\[ \mathcal{E}(t) = \frac{1}{2} \langle U, U \rangle_\mathcal{H} \]

Energy principle

\[ \frac{d\mathcal{E}}{dt} = -\frac{\gamma}{\varepsilon_0 \omega_p^2} \| J \|^2 \]

Dissipative operator

\[ \mathcal{R}(\langle \mathcal{L} U, U \rangle_\mathcal{H}) = \langle \mathcal{F} U, U \rangle_\mathcal{H} \leq 0 \]

\[ + \mathcal{L} \text{ is maximal.} \]

Theorem

The operator \( \mathcal{L} \) with domain \( D(\mathcal{L}) \) generates a \( C_0 \)-semigroup of contractions on \( \mathcal{H} \).

\textsuperscript{1}S. Nicaise, C. Scheid, CAMWA, 2020
What about the decay of the energy? (joint work with S. Nicaise)

Polynomial decay of the energy

Theorem

There exists a positive constant $C$ such that for all $U_0 \in D(A)$, $\forall t > 0$,

$$\mathcal{E}(t) \leq C t^{-1} \|U_0\|^2.$$

Sketch of the proof

- Imaginary axis in the resolvent set,

$$i\mathbb{R} \subset \rho(\mathcal{L})$$

- "Control at high frequencies"

$$\limsup_{|\xi| \to \infty} \frac{1}{\xi^2} \|(i\xi - \mathcal{L})^{-1}\| < \infty.$$
What about the decay of the energy (joint work with S. Nicaise)

Optimality of the decay

- Expansion of some eigenvalues at high frequencies.

Theorem

There exists $k_0$ large enough such that $\mathcal{L}$ has eigenvalues $\lambda_k^\pm$, for all $k \geq k_0$ satisfying

$$
\lambda_k^\pm = \pm i \left( (\varepsilon_0 \varepsilon_r \mu)^{-1/2} \lambda_{M,k} + \sqrt{\frac{\mu \varepsilon_0}{\varepsilon_r} \frac{\omega_p^2}{2 \lambda_{M,k}}} \right) - \frac{\gamma \varepsilon_0 \omega_p^2 \mu}{2 \lambda_{M,k}^2} + o \left( \frac{1}{\lambda_{M,k}^2} \right), \forall k \geq k_0.
$$

where $(\lambda_{M,k}^2)_k$ eigenvalues of curl(curl(.)) operator with PEC BC.

- For all $\varepsilon > 0$, construct an initial data that decay more slowly than $\frac{1}{t^{1+\varepsilon}}$.

Corollary

The decay rate is optimal.
Generalization to other BCs (joint work with S. Nicaise)?

\( \Omega \) exterior of \( O \) a bounded domain of \( \mathbb{R}^3 \) (\( \partial O = \Gamma_S \)), truncated by an artificial boundary \( \Gamma_A \).

Boundary conditions

- On \( \Gamma_S \), \( \mathcal{B}_{\text{perfect}}(U) = 0 \).
- On \( \Gamma_A \), \( \mathcal{B}_{\text{abs}}(U) = 0 \):

\[
E \times n - z(H \times n) \times n = 0 \text{ on } \Gamma_A, \quad \text{and}
\]

\[
\beta_1 J \cdot n + \beta_2 Q = 0 \text{ on } \Gamma_A,
\]

with \( (\beta_1, \beta_2) \in \mathbb{R}^+ \times \mathbb{R}^+ \) such that \( \beta_1 + \beta_2 > 0 \) and \( z = \sqrt{\frac{\mu}{\epsilon}} \).
Properties of the operators.

- $\mathcal{A}$ is not skew-adjoint, $\Re \langle AU, U \rangle_{\mathcal{H}} \leq 0$ because of $B_{abs}$,
- $\Re \langle KU, U \rangle_{\mathcal{H}} = 0$,
- $\langle FU, U \rangle_{\mathcal{H}} = -\frac{\gamma}{\varepsilon_0 \omega_p^2} \| J \|^2 \leq 0$.

Strategy: Prove that $\mathcal{A}$ is densely defined and closed and that $\mathcal{A}$ and $\mathcal{A}^*$ are dissipative.

Characterization of the adjoint

One has $D(\mathcal{A}^*) = \{ U \in \mathcal{H} \mid OU \in D(\mathcal{A}) \}$, and

$$\mathcal{A}^* = \mathcal{O} \mathcal{A} \mathcal{O},$$

with $\mathcal{O}(F, G, R, S)^\top = (F, -G, -R, S)^\top$.

$\Rightarrow$ The problem is well posed!
Decay rate of the energy? (joint work with S. Nicaise, 2021)

Polynomial decay of the energy

Theorem

There exists a positive constant $C$ such that for all $U_0 \in D(A)$, $\forall t > 0$,

- If $\beta_2 > 0$,
  \[ \mathcal{E}(t) \leq C t^{-1} ||U_0||^2. \]

- If $\beta_2 = 0$
  \[ \mathcal{E}(t) \leq C t^{-\frac{1}{3}} ||U_0||^2. \]
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3. Theoretical study
4. Numerical framework
   - Academic context
   - Is the model with $\beta \neq 0$ physically relevant?
5. Validity of the model
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Numerical framework: semi-discretization

Discontinuous Galerkin discretization framework in space.

- Nodal DG, piecewise polynomials on each cells of the mesh: $\mathcal{V}_h \subset L^2$ $V_h \not\subset D(\mathcal{L})$.

Semi-discrete formulation

*Find $U_h \in C^1(0,T,\mathcal{V}_h)$ such that for all $U'_h \in \mathcal{V}_h$,*

$$\langle \frac{\partial U_h}{\partial t}, U'_h \rangle = \langle \mathcal{L}_h(U_h), U'_h \rangle_h,$$

$$\langle \mathcal{L}_h(U_h), U'_h \rangle_h = (U_h, A_h^* U'_h)_h + \langle \tilde{B}_h(U^*_h), U'_h \rangle_\partial + \langle (\mathcal{K} + \mathcal{F})(U_h), U'_h \rangle$$

- Use of centered fluxes or upwind fluxes. Hidden in $\tilde{B}_h$
Semi-discrete formulation

Find $U_h \in C^1(0,T,\mathcal{V}_h)$ such that for all $U'_h \in \mathcal{V}_h$,

$$
\langle \frac{\partial U_h}{\partial t}, U'_h \rangle = \langle \mathcal{L}_h(U_h), U'_h \rangle_h,
$$

$$
\langle \mathcal{L}_h(U_h), U'_h \rangle_h = (U_h, A^*_h U'_h)_h + \langle \tilde{B}_h(U^*_h), U'_h \rangle_\partial + \langle (\mathcal{K} + \mathcal{F})(U_h), U'_h \rangle
$$

Properties of the semi-discrete energy: $\mathcal{E}_h = \frac{1}{2} \langle U_h, U_h \rangle_H$

\begin{itemize}
  \item $\frac{d\mathcal{E}_h}{dt} = \langle \mathcal{F} U_h, U_h \rangle_H$ for centered fluxes $\rightsquigarrow$ Preserved energy principle.
  \item $\frac{d\mathcal{E}_h}{dt} = \langle \mathcal{F} U_h, U_h \rangle_H - \frac{\gamma}{\varepsilon_0 \varepsilon_r} \| U_h \|_{\text{faces}}^2 - \frac{\gamma}{\varepsilon_0 \varepsilon_r} \| U_h \|^2$ for upwind fluxes $\rightsquigarrow$ Numerical dissipation.
\end{itemize}

Stability and a priori convergence analysis are at reach!
$\rightsquigarrow$ standard numerical analysis.
Fully discrete schemes

Time integration with explicit schemes:

- Leap frog scheme of order 2 (LF2),
- Runge Kutta (RK2/RK4).

～～～

(LF2 & centered fluxes) or (RK2/RK4 & upwind fluxes)

～～～

Standard numerical analysis

- Theoretical proofs of stability (under CFL), convergence estimates via discrete energy principles and numerical validation in 2D and 3D
- CFL impacted by physical coefficients: especially $\omega_p$.

Discrete preservation of properties

<table>
<thead>
<tr>
<th></th>
<th>LF2 &amp; centered fluxes</th>
<th>RK2/RK4 &amp; upwind fluxes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Constraint (weakly)</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Energy principle</td>
<td>yes</td>
<td>no (num. dissipation)</td>
</tr>
<tr>
<td>Numerical decay</td>
<td>yes</td>
<td>no</td>
</tr>
</tbody>
</table>

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5 Validity of the model
Is the model with $\beta \neq 0$ physically relevant?

$\beta = 0$ is usually sufficient!

Mandatory improvements.
- Full 3D parallel
- PML’s
- TF/SF
- Curvilinear elements
- p-local approximations
- Hybrid meshes
- Quantities of interest
- Oblique incidence
- ...

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Validity of the model (PhD of N. Schmitt).

In lots of situations $\beta = 0$ is usually sufficient!

Is the model with $\beta \neq 0$ physically relevant?

Goal: Find structures for which the model impacts the position of measured resonances.

joint work with N. Schmitt (Inria), A. Moreau, A Pitelet, E. Centeno (Clermont-Ferrand), D. Loukrezis, H. De Gersem (T.U. Darmstadt), C. Ciraci (ITT, Italia).

4Pitelet et al, JOSA B, 2019
Validity of the model

Grating study

Key observation

Increase of the permittivity of the dielectric

\[ \downarrow \]

Increase of sensitivity of surface plasmons to quantum and internal pressure, i.e. to $\beta$

$\beta$

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A study of linear dispersive models for nanoplasmatics

07/10/2021 29/32
Overall goal

- Track surface plasmons resonances positions: dip in the reflectance spectra,
- Investigate whether they are captured by the model with $\beta \neq 0$ or $\beta = 0$.

Procedure

- Calibration step: find ”good” dimensions for the structure
- Investigate resonances positions with the two models.
- Generate a noisy reflectance spectra
- Estimate whether differences are significative with respect to variation of geometrical parameters.
Validity of the model

Grating

Taking into account for geometric uncertainties \(^5\)
Impact on the reflectance spectrum.

Blue \(\rightarrow \beta = 0\), Orange \(\rightarrow \beta \neq 0\).

Perspectives

Towards users ⟷ More physical test cases

Some methodological improvements
Design of new Finite Elements methods, include strategies of optimization...

Improvement of models