A study of linear dispersive models for nanoplasmonics.

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Work in collaboration with ATLANTIS INRIA PROJECT TEAM

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S. Nicaise.

Workshop Numerical Waves, Nice

Outline

Motivations

Control of the interaction of light with nano-scaled structures.
$\rightsquigarrow$ Nanophotonics

Motivations

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

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

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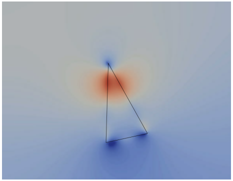
Light + subwavelength metallic structures

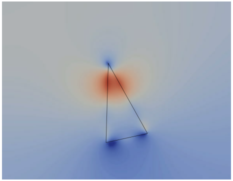
$\downarrow$

Collective oscillations of the electrons of the metal.

$\downarrow$

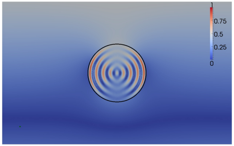
Plasmons



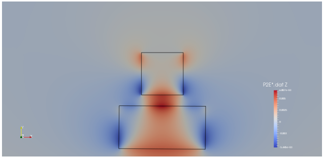


Surface plasmons

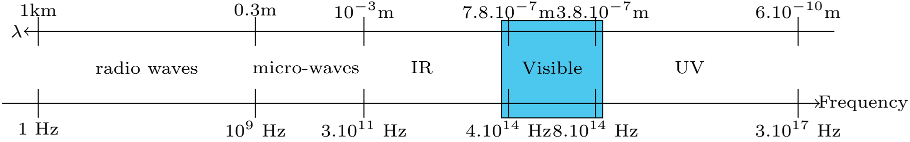
Bulk plasmons

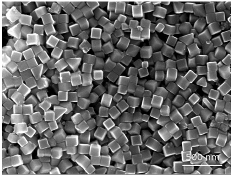


Gap plasmons



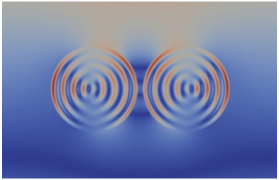
Optical frequencies: $\approx [300nm, 700nm]$.



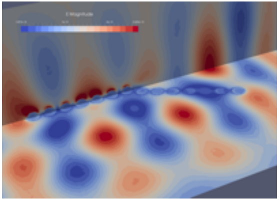


$\rightsquigarrow$ Size of the nano-structuration $< \lambda$.

$\sim\sim\sim$



Subwavelength phenomena.



Motivations

Interaction of electromagnetic waves with complex heterogeneous media.

Metals at the nanoscales at optical frequencies:
$\rightsquigarrow$ Computational nanophotonics.

Motivations

Interaction of electromagnetic waves with complex heterogeneous media.

Metals at the nanoscales at optical frequencies:
$\rightsquigarrow$ Computational nanophotonics.

Challenges in this context

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

Outline

Modelling equations

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

Hydrodynamic description ($\mathbf{v}$: speed, n: density.)

$$
\begin{aligned}
m\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla\right)\mathbf{v} &= -e\left(\mathbf{E} + \mathbf{v}\times\mathbf{B}\right) - m\gamma\mathbf{v} - \frac{m\beta^2}{n}\nabla n \\
\frac{\partial}{\partial t}n + \operatorname{div}(n\mathbf{v}) &= 0 \\
\mathbf{J} &= -en\mathbf{v}
\end{aligned}
$$

+ Time Domain Maxwell's equations ($\mathbf{J}$, $\mathbf{E}$ and $\mathbf{B}$).

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\end{aligned}
$$

$+$ Time Domain Maxwell's equations ($\mathbf{J}$, $\mathbf{E}$ and $\mathbf{B}$).

$\frac{m\beta^2}{n}\nabla n \rightsquigarrow$ quantum pressure term.

Linearized Hydrodynamic model

Formal linearization around an equilibrium state $(n_0, v_0, \mathbf{E}_0, \mathbf{B}_0, J_0) = (n_0, 0, \mathbf{E}_0, 0)$,

$$\begin{cases} \varepsilon_0 \varepsilon_r \dfrac{\partial \mathbf{E}}{\partial t} = \text{curl}\, \mathbf{H} - \mathbf{J} \\ -\mu_0 \dfrac{\partial \mathbf{H}}{\partial t} = \text{curl}\, \mathbf{E} \\ \dfrac{\partial \mathbf{J}}{\partial t} = \beta^2 \nabla Q - \gamma \mathbf{J} + \varepsilon_0 \omega_p^2 \mathbf{E} \\ \dfrac{\partial Q}{\partial t} = \nabla \cdot \mathbf{J}, \end{cases}$$

Maxwell's equations

$+$

$\beta \neq 0 \rightsquigarrow$ PDE for polarization current
$\beta = 0 \rightsquigarrow$ ODE for polarization current

Charge preservation

$$\text{div}(\varepsilon_0 \varepsilon_r \mathbf{E}) = -Q$$

$$\begin{cases} \varepsilon_0 \varepsilon_r \dfrac{\partial \mathbf{E}}{\partial t} = \text{curl}\, \mathbf{H} - \mathbf{J} \\ -\mu_0 \dfrac{\partial \mathbf{H}}{\partial t} = \text{curl}\, \mathbf{E} \\ \dfrac{\partial \mathbf{J}}{\partial t} = \beta^2 \nabla Q - \gamma \mathbf{J} + \varepsilon_0 \omega_p^2 \mathbf{E} \\ \dfrac{\partial Q}{\partial t} = \nabla \cdot \mathbf{J}, \end{cases}$$

$\sim\sim\sim\sim$

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!

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

Synthetic formulation

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

$$\begin{cases} \partial_t U = \mathfrak{L} U, \\ U(0) = U_0, \end{cases}$$

$$\mathfrak{L} = \mathcal{A} + \mathcal{K} + \mathcal{F}$$

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

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

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

$$\mathcal{F}U = \begin{pmatrix} 0 \\ 0 \\ -\gamma J \\ 0 \end{pmatrix}$$

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

- Ω Lipschitz open bounded simply connected domain $\subset \mathbf{R}^3$.
- Boundary conditions $\mathcal{B}_{perfect}(U) = 0$: $E \times \boldsymbol{n} = 0$, $Q = 0$ on $\partial\Omega$

$$\mathcal{H} = \{\mathcal{U} \in H(\mathrm{div}, \Omega) \times H_0(\mathrm{div} = 0, \Omega) \times L^2(\Omega)^3 \times L^2(\Omega), \mathrm{div}(\varepsilon \mathcal{U}_1) = -\mathcal{U}_4 \text{ on } \Omega\},$$

$$\langle \mathcal{U}, \mathcal{U}' \rangle_{\mathcal{H}} := \varepsilon_0 \varepsilon_L \langle \mathcal{U}_1, \mathcal{U}_1' \rangle + \mu \langle \mathcal{U}_2, \mathcal{U}_2' \rangle + \frac{1}{\varepsilon_0 \omega_p^2} \langle \mathcal{U}_3, \mathcal{U}_3' \rangle + \frac{\beta^2}{\varepsilon_0 \omega_p^2} \langle \mathcal{U}_4, \mathcal{U}_4' \rangle$$

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

$$\text{"} \left\{ U \in \mathcal{H},\, \mathcal{A}U \in L^2,\, \mathcal{B}_{perfect}(U) = 0 \right\} \text{"}.$$

Properties of the operators.

- $\mathcal{A}$ is skew-adjoint, $\Re \langle \mathcal{A}U, U \rangle_{\mathcal{H}} = 0$,
- $\Re \langle \mathcal{K}U, U \rangle_{\mathcal{H}} = 0$,
- $\langle \mathcal{F}U, U \rangle_{\mathcal{H}} = -\frac{\gamma}{\varepsilon_0 \omega_p^2} \|J\|^2 \leq 0$.

Well-posedness (joint work with S. Nicaise[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

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

$+ \mathfrak{L}$ is maximal.

Theorem

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

[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 $U_0 \in \mathcal{D}(\mathcal{A})$, $\forall t > 0$,

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

Sketch of the proof

- Imaginary axis in the resolvent set,

$$i\mathbf{R} \subset \rho(\mathfrak{L})$$

- "Control at high frequencies"

$$\limsup_{|\xi| \to \infty} \frac{1}{\xi^2} \, \|(i\xi - \mathfrak{L})^{-1}\| < \infty.$$

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 $\mathfrak{L}$ 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[2])?

Ω exterior of O a bounded domain of $\mathbf{R}^3$ ($\partial O = \Gamma_S$), truncated by an artificial boundary Γ_A.
Boundary conditions

- On Γ_S, $\mathcal{B}_{perfect}(U) = 0$.
- On Γ_A, $\mathcal{B}_{abs}(U) = 0$:

$$E \times \mathbf{n} - z(H \times \mathbf{n}) \times \mathbf{n} = 0 \text{ on } \Gamma_A, \text{ and} \quad (1)$$
$$\beta_1 J \cdot \mathbf{n} + \beta_2 Q = 0 \text{ on } \Gamma_A, \quad (2)$$

with $(\beta_1, \beta_2) \in \mathbf{R}^+ \times \mathbf{R}^+$ such that $\beta_1 + \beta_2 > 0$ and $z = \sqrt{\frac{\mu}{\epsilon}}$

[2] S. Nicaise, C. Scheid, preprint, 2021

Properties of the operators.

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

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 $D(\mathcal{A}^*) = \{U \in \mathcal{H} \,|\, \mathcal{O}U \in D(\mathcal{A})\}$, and

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

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

$\rightsquigarrow$ **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 $U_0 \in \mathcal{D}(\mathcal{A})$, $\forall t > 0$,

- *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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Numerical framework: semi-discretization

Discontinuous Galerkin discretization framework in space.

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

Semi-discrete formulation

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

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

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

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

Semi-discrete formulation

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

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

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

- $\frac{d\mathcal{E}_h}{dt} = \langle \mathcal{F}U_h, U_h \rangle_{\mathcal{H}}$ for centered fluxes $\rightsquigarrow$ Preserved energy principle.
- $\frac{d\mathcal{E}_h}{dt} = \underbrace{\langle \mathcal{F}U_h, U_h \rangle_{\mathcal{H}}}_{-\frac{\gamma}{\varepsilon_0 \varepsilon_r}\|U_h\|^2} - \|[\![U_h]\!]\|^2_{faces}$ for upwind fluxes $\rightsquigarrow$ Numerical dissipation.

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).

$\sim\sim\sim$

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

$\sim\sim\sim$

Academic studies. [3]

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 ω_p.

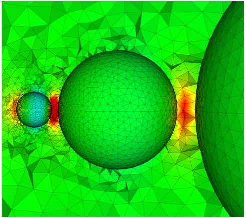
Discrete preservation of properties

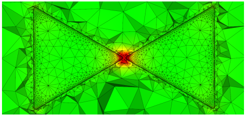
	LF2 & centered fluxes	RK2/RK4 & upwind fluxes
Constraint (weakly)	yes	yes
Energy principle	yes	no (num. dissipation)
Numerical decay	yes	no

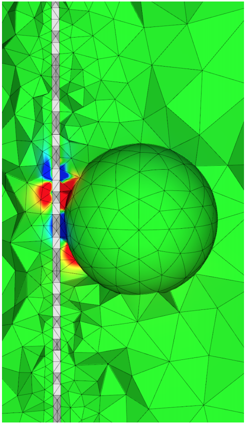
[3] Schmitt, C. S., Lanteri, Viquerat, Moreau, JCP(2016), Lanteri, C. SS., Viquerat, SISC (2017), Schmitt, S., Viquerat, Lanteri, JCP (2018), S. Nicaise, C. S., CAMWA(2020)

Outline

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



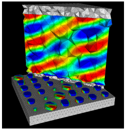




DIOGENES
A DG-based software suite for nano-optics

Mandatory improvements.

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



$\beta = 0$ **is usually sufficient !**

Outline

Validity of the model (PhD of N. Schmitt [4]).

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).

[4] Pitelet et al, JOSA B, 2019

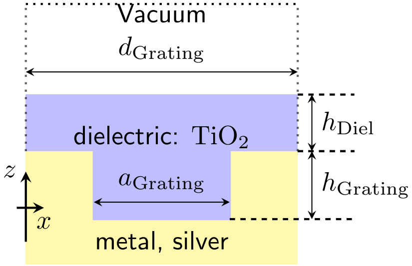
Grating study

Key observation

Increase of the permittivity of the dielectric

$\Downarrow$

Increase of sensitivity of surface plasmons to quantum and internal pressure i.e. to β



Metallic grating

"not so small" structure (> 20 nm)

Overall goal

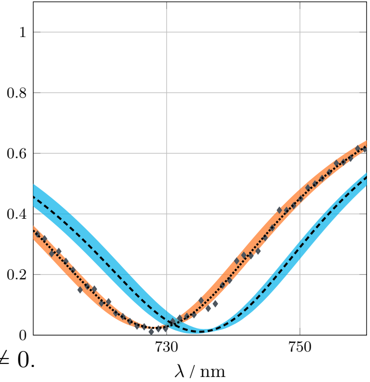
- Track surface plasmons resonances positions: dip in the reflectance spectra,
- Investigate whether they are captured by the model with $\beta \neq 0$ or $\beta = 0$.

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.

Grating

Taking into account for geometric uncertainties [5]
Impact on the reflectance spectrum.



Blue⇝$\beta = 0$, Orange⇝$\beta \neq 0$.

[5] Influence of spatial dispersion on surface plasmons and grating couplers, A. Pitelet et. al., JOSA B, 2019.

Perspectives

Towards users $\rightsquigarrow$ More physical test cases

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

Improvement of models