Harmonic Generation at Nanoscale in Strategic Materials for Nanophotonics

[1]L. Rodríguez-Suné, [1]C. Cojocaru, [2]M. Scalora, and [1]J. Trull
[1]Department of Physics, Universitat Politècnica de Catalunya, Rambla Sant Nebridi 22, 08222 Terrassa, Spain
[2]Aviation and Missile Center, US Army CCDC, Redstone Arsenal, Huntsville, AL, United States

ABSTRACT

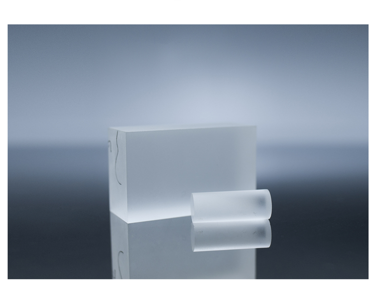
Nowadays structures with **nanometric size** are produced and integrated in different photonic devices and applications. At this scale light-matter interaction displays completely new phenomena. Understanding how light interacts at the nanoscale with metals, semiconductors, or ordinary dielectrics is pivotal if one is to properly engineer and implement nano-antennas, filters and, more generally, devices that aim to harness the effects of new physical phenomena that manifest themselves at the nanoscale. For this reason, we study the processes of **second and third harmonic generation** in nanolayers of different strategic materials for nanophotonics as semiconductors, metals and conductive oxides. We compare our measurements with numerical results based on our theoretical model which allows us to understand and explain the different contributions to the nonlinear light-matter interaction at the nanoscale.

BULK NONLINEAR OPTICS

Leading NL polarization term: **bulk contribution** described through $\chi^{(2)}$ and $\chi^{(3)}$.

$\vec{P}_{ED}^{NL} = \chi^{(2)}\vec{E}\vec{E} + \chi^{(3)}\vec{E}\vec{E}\vec{E} + \cdots$

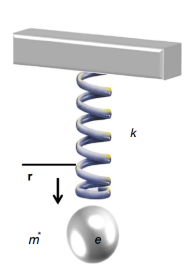
High conversion efficiencies (>10^{-2}): thick NL materials (cm, mm, μm) with high nonlinearities, phase matching conditions, and low material absorption (BBO, LBO, KTP, $LiNbO_3$…).


BBO crystal


$LiNbO_3$ crystal

NONLINEAR POLARIZATION: THEORETICAL MODEL

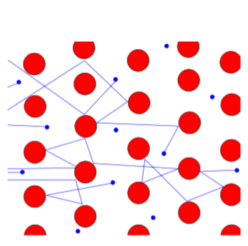
- Bound electrons



$$\ddot{\boldsymbol{P}}_{bj} + \tilde{\gamma}_{bj}\dot{\boldsymbol{P}}_{bj} + \tilde{\omega}_{0,bj}^2\boldsymbol{P}_{bj} + \boldsymbol{P}_{bj}^{Bulk} = \frac{e^2\lambda_0^2 n_{0,b}}{m_{bj}^* c^2}\boldsymbol{E} + \frac{e\lambda_0}{m_{bj}^* c^2}(\boldsymbol{P}_{bj}\cdot\nabla)\boldsymbol{E} + \frac{e\lambda_0}{m_{bj}^* c^2}\dot{\boldsymbol{P}}_{bj}\times\boldsymbol{H}$$

Bulk; Surface; Magnetic

- Free electrons



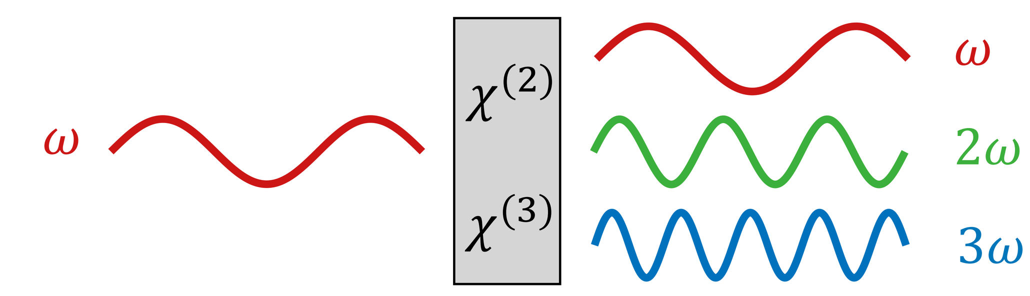
$$\ddot{\boldsymbol{P}}_f + \tilde{\gamma}_f\dot{\boldsymbol{P}}_f = \frac{e^2\lambda_0^2 n_{0,f}}{m_0^* c^2}\boldsymbol{E} \pm \tilde{\Lambda}(\boldsymbol{E}\cdot\boldsymbol{E})\boldsymbol{E} - \frac{e\lambda_0}{m_0^* c^2}(\nabla\cdot\boldsymbol{P}_f)\boldsymbol{E} + \frac{e\lambda_0}{m_0^* c^2}\dot{\boldsymbol{P}}_f\times\boldsymbol{H} + \frac{3E_F}{5m_0^* c^2}\left[\nabla(\nabla\cdot\boldsymbol{P}_f) + \frac{1}{2}\nabla^2\boldsymbol{P}_f\right] - \frac{1}{n_{0,f}e\lambda_0}\left[(\nabla\cdot\dot{\boldsymbol{P}}_f)\dot{\boldsymbol{P}}_f + (\dot{\boldsymbol{P}}_f\cdot\nabla)\dot{\boldsymbol{P}}_f\right]$$

Hot electrons; Nonlocal; Convection

$$\boldsymbol{P}_{TOT}^{NL} = \boldsymbol{P}_b^{NL} + \boldsymbol{P}_f^{NL} \longrightarrow \quad \nabla\times\boldsymbol{E} = -\frac{1}{c}\frac{\partial\boldsymbol{H}}{\partial t} \qquad \nabla\times\boldsymbol{H} = \frac{1}{c}\frac{\partial\varepsilon\boldsymbol{E}}{\partial t} + \frac{4\pi}{c}\frac{\partial\boldsymbol{P}_{TOT}^{NL}}{\partial t}$$

SECOND AND THIRD HARMONIC GENERATION

SHG and THG are two **nonlinear (NL) processes**.
The input wave interacts with the NL material. As a result, **new frequencies are generated**.
These NL processes are described through the **NL polarization** $\vec{P}_{NL}$.



NONLINEAR OPTICS AT NANOSCALE

The reduced thickness of the material drastically diminish the bulk contribution. Typical efficiencies: 10^{-7} – 10^{-10}.

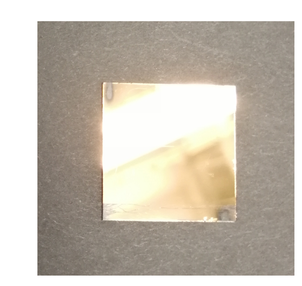
New light matter interaction phenomena: $\vec{P}^{NL} = \vec{P}_{ED}^{NL} + \vec{P}_{MD}^{NL} + \vec{P}_{EQ}^{NL}$.

Most models rely on assigning **effective surface and volume** $\chi^{(2)}$ for SHG and **effective volume** $\chi^{(3)}$ for THG.

Lack of a detailed **microscopic description** of light propagation and light-matter interactions.

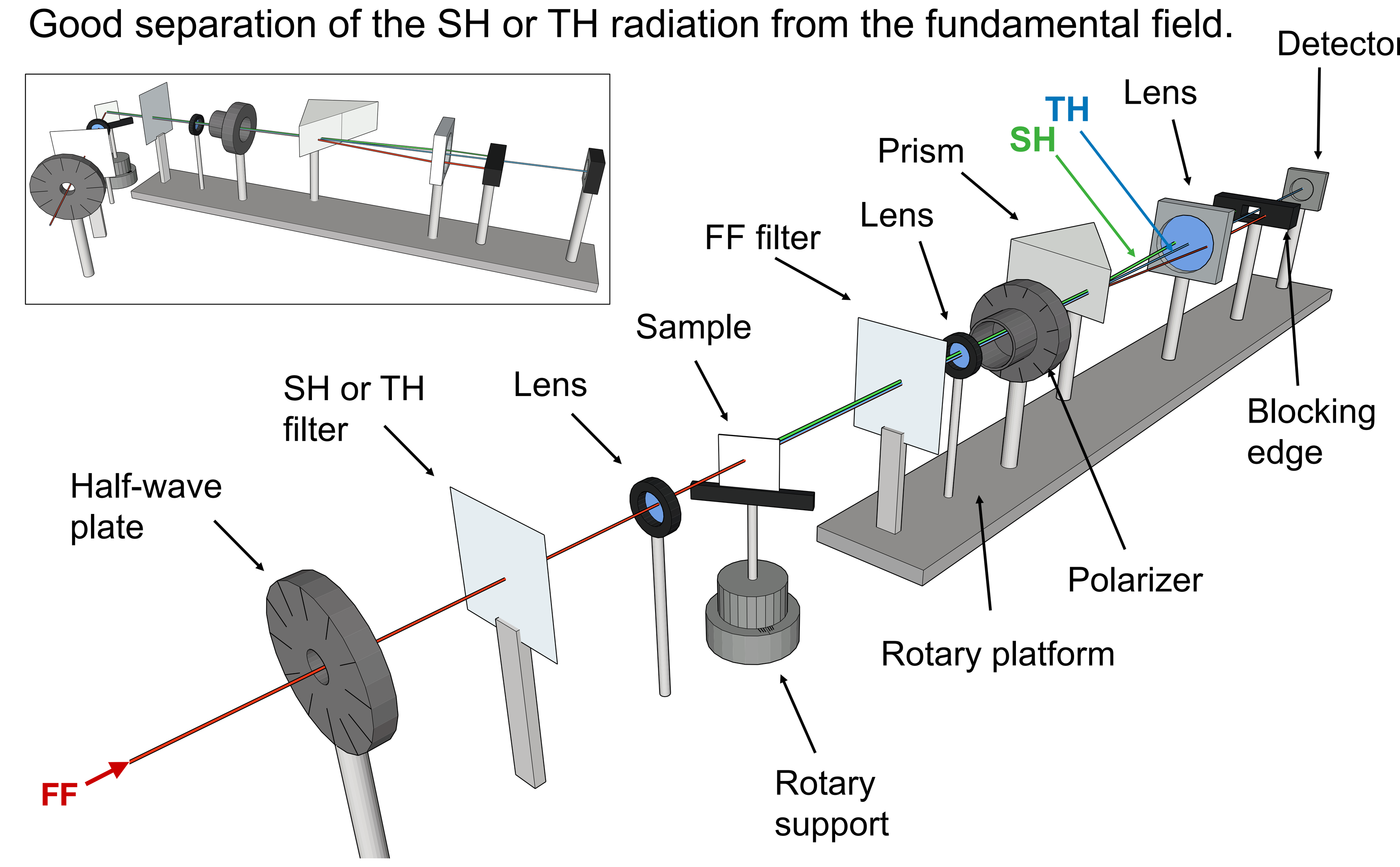

ITO nanolayers


Si/GaAs wafers

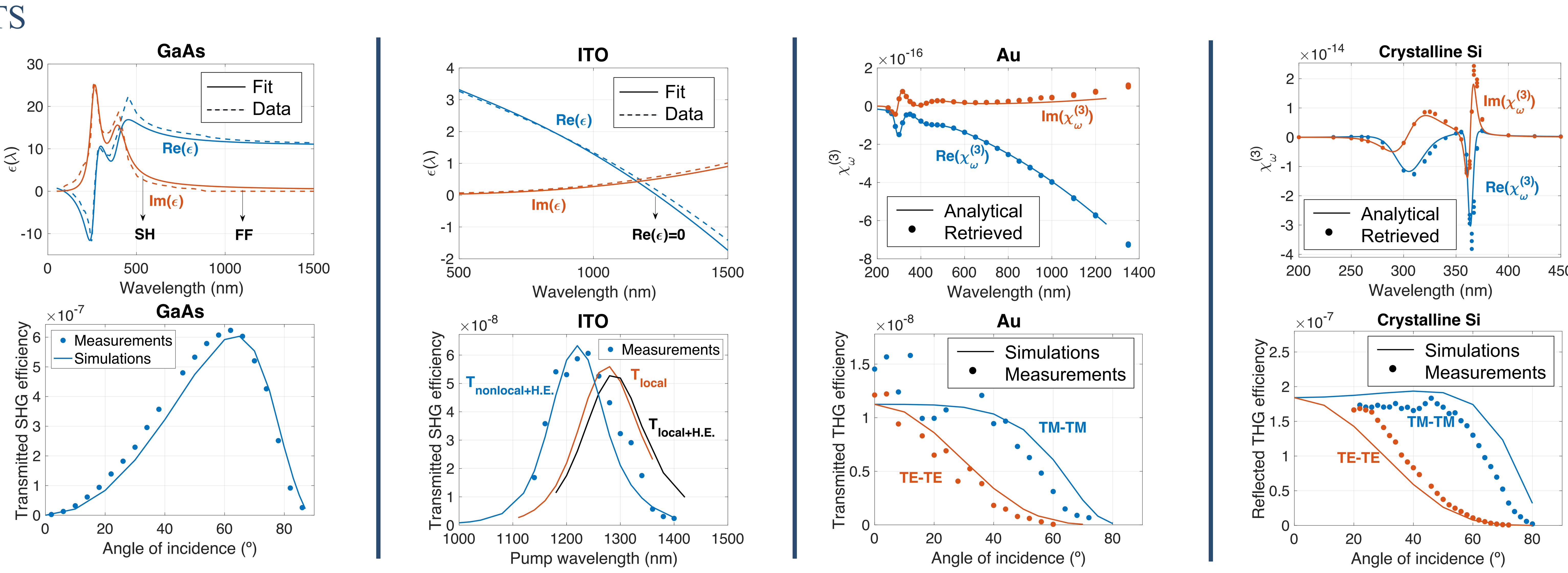

Au nanolayer

EXPERIMENTAL SET-UP

SHG and THG measurements in transmission and reflection.
Control of the polarization and angle of incidence.
Good separation of the SH or TH radiation from the fundamental field.



RESULTS



The combination of experimental measurements and numerical simulations gives important information of the particular contributions of each NL polarization source on the generation of the SH and TH fields.

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