NONLINEAR OPTICS AT NANOSCALE

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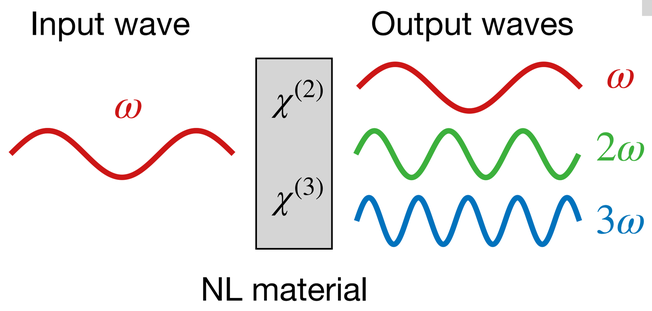
ABSTRACT

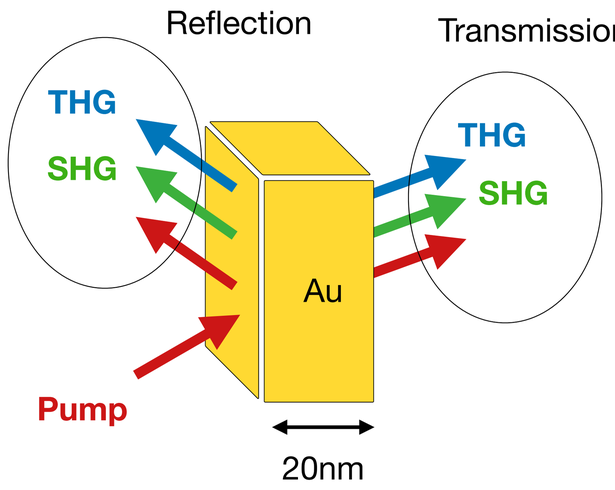
Nowadays structures with nanometric size are produced and integrated in different photonic devices and applications. At this scale light-matter interaction displays completely new phenomena. Understanding how light interacts at the nanoscale with metals, semiconductors, or ordinary dielectrics is pivotal 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 experimentally the processes of second and third harmonic generation in different nanolayers and compare our measurements with numerical results based on our theoretical model which allows us to understand and explain the light-matter interaction at the nanoscale.

SECOND AND THIRD HARMONIC GENERATION



➡SHG and THG are two **NL processes**.

➡The input wave interacts with the NL material. As a result, the frequency of the output waves is doubled (**SHG**) or triplicated (**THG**).

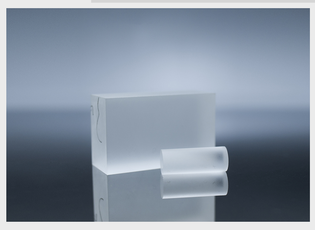


➡As NL material we use structures with **nanometric size.**

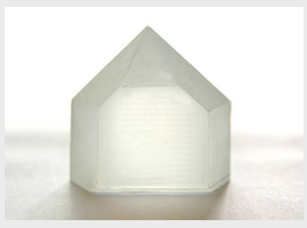
➡SH and TH signals are generated in transmission and in reflection. Our experimental set-up allows us to measure the signals in both configurations.

➡We compare the experimental results with numerical simulations that allow us to understand the observations.

BULK NONLINEAR OPTICS



BBO crystal



$LiIO_3$ crystal

➡Higly NL material

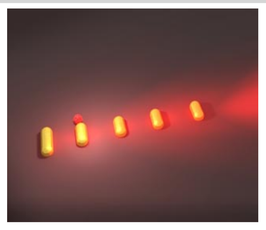
➡High SHG efficiencies

➡$\mathbf{P}_{ED} = \epsilon_0[\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{E}^2 + \chi^{(3)}\mathbf{E}^3 + \dots]$

SURFACE NONLINEAR OPTICS

GaAs wafer

ITO nanolayer



Gold nanoantenas

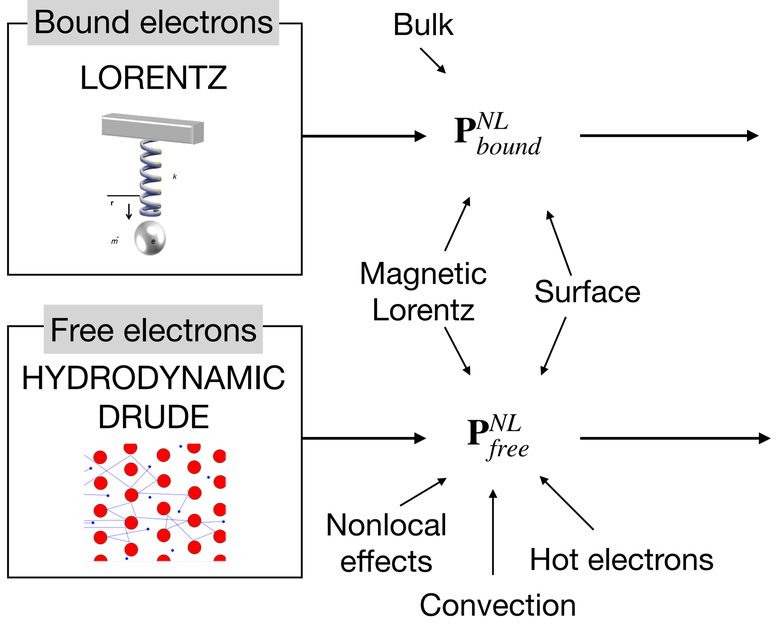
Gold nanolayer

➡Bulk contribution to the SHG drastically decrease and other terms like MD and EQ become important

➡New light-matter interaction phenomena: $\mathbf{P}^{NL} = \mathbf{P}_{ED} + \mathbf{P}_{MD} + \mathbf{P}_{EQ} + \dots$

➡Conventional approximations no longer valid

NONLINEAR POLARIZATION



$$\ddot{\mathbf{P}}_b + \tilde{\gamma}_b\dot{\mathbf{P}}_b + \tilde{\omega}^2_{0,b}\mathbf{P}_b + \mathbf{P}^{bulk}_b = \frac{n_{0,b}e^2\lambda_0^2}{m^*_b c^2}\mathbf{E} + \frac{e\lambda_0}{m^*_b c^2}\mathbf{E}(\nabla\cdot\mathbf{P}_b) + \frac{e\lambda_0}{m^*_b c^2}\dot{\mathbf{P}}_b\times\mathbf{H}$$

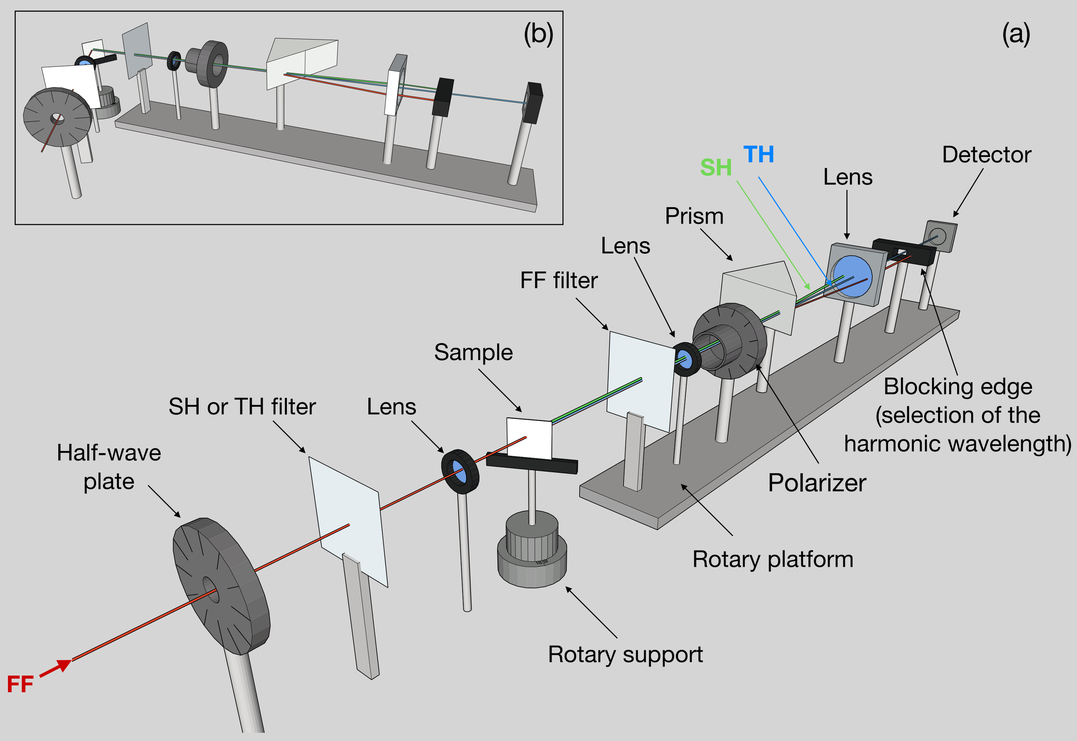
$$\ddot{\mathbf{P}}_f + \tilde{\gamma}\dot{\mathbf{P}}_f = \frac{n_{0,f}e^2\lambda_0^2}{m^*_{0,f}c^2}\mathbf{E} - \frac{e\lambda_0}{m^*_{0,f}c^2}\mathbf{E}(\nabla\cdot\mathbf{P}_f) \pm \tilde{\Lambda}(\mathbf{E}\cdot\mathbf{E})\mathbf{E} + \frac{e\lambda_0}{m^*_{0,f}c^2}\dot{\mathbf{P}}_f\times\mathbf{H}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

$$\mathbf{P}^{NL}_{total} = \mathbf{P}^{NL}_{free} + \mathbf{P}^{NL}_{bound}$$

$$\nabla\times\mathbf{E} = -\frac{1}{c}\frac{\partial\mathbf{H}}{\partial t}$$

$$\nabla\times\mathbf{H} = \frac{1}{c}\frac{\partial\mathbf{E}}{\partial t} + \frac{4\pi}{c}\frac{\mathbf{P}^{NL}_{total}}{\partial t}$$

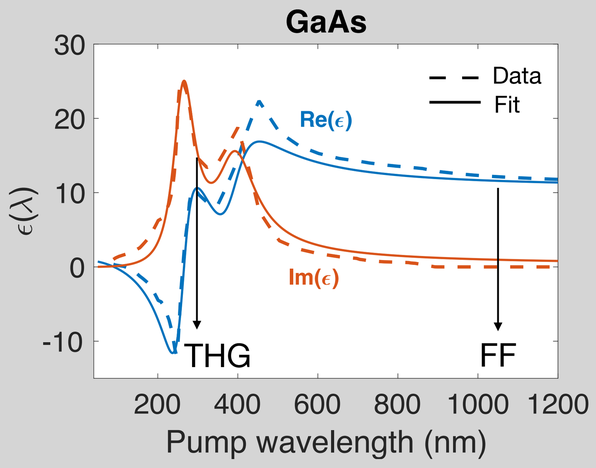
EXPERIMENTAL SET-UP

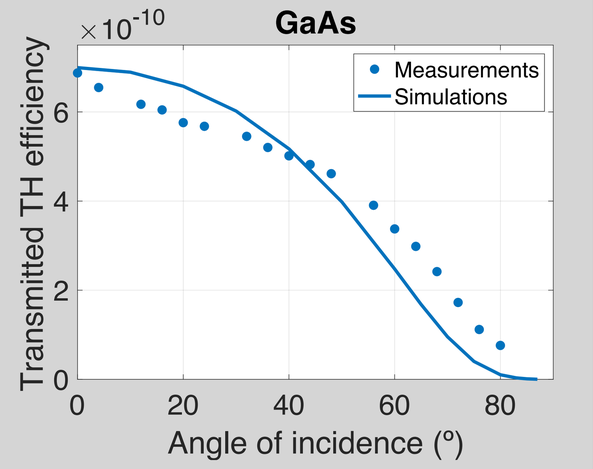


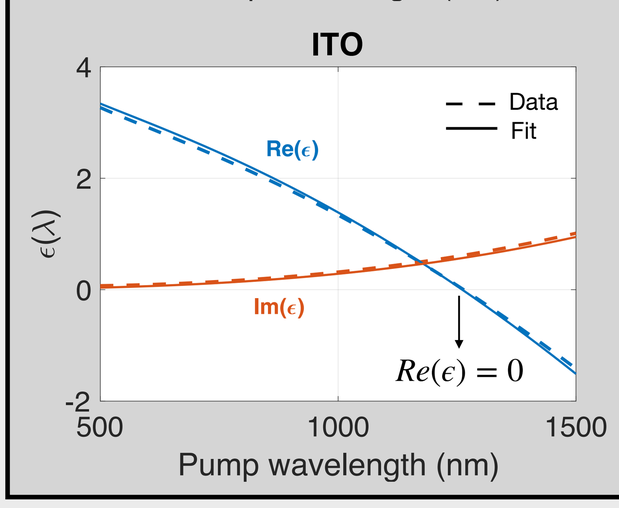
Experimental set-up implemented for the measurement of SH and TH signals. The detection platform is mounted on a rotating arm to be used for (a) **transmission** and (b) r**eflection** measurements.

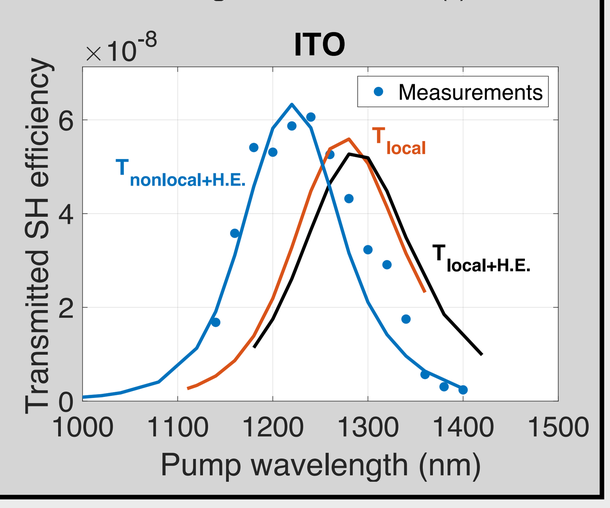
HARMONIC GENERATION FROM GaAs AND ITO

1. **Measurements:** we take measurements of the **harmonic signal** as a function of the angle of incidence and input wavelength.
2. **Simulations:** reproducing the experimental situation we calculate the nonlinear polarization and with Maxwell's equations we extract the fields.
3. A very good agreement is found.









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- L. Rodríguez-Suné *et. al.*, "*Study of second and third harmonic generation from an indium tin oxide nanolayer: Influence of nonlocal effects and hot electrons*," APL Photonics 5, 010801 (2020)