Nonlinear Optics in Plasmonic Nanostructures

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Abstract

The unique optical properties of plasmon resonances in noble metal nanoparticles have been extensively investigated owing to their ability to enhance the electric field amplitude but also to tailor its spectral and spatial distribution. Among all application domains, nonlinear optics play a singular role since the efficiency of the underlying physical processes are driven by specific symmetry properties and local density of states, being drastically affected by the metal response. In particular, these resonances enable the observation of non-linear optical responses such as second harmonic generation (SHG) and two photon luminescence (TPL) [1, 2]. The latter has been widely used to infer the field localization at the nanoscale, becoming a key tool for investigating the plasmon resonances even in the context of gap induced hot spots formation.

The nonlinear currents responsible for SHG in metal nanostructures arise from the breakdown of the centrosymmetry at the metal surface (local surface response) or from intensity gradients inside the bulk (nonlocal bulk response). Including these contributions in numerical simulations has been performed in pioneering work using COMSOL Multiphysics® software [3]. Since then, coupling numerical and analytical tools (LiveLink for MATLAB®) have opened new opportunities in order to account for the field focusing by high numerical aperture objectives [4], the presence of the substrates and of oil immersion and finally the collection of the SHG fields by the same objective (see Figure 1). As a final step, in order to reproduce as close as possible the experimental configuration, CAD facilities have been incorporated for designing the investigated nanostructures (LiveLink for SOLIDWORKS®). The extensive resource cost in order to obtain a 2D map of the SHG signal (by scanning the sample) has motivated deploying the COMSOL simulation on a new cluster at the Neel Institute. Depending on the actual simulations, this allowed drastically reducing the computation time bringing the prohibitive month time-scale of the first proof-of-concept calculation below the day threshold required for reasonable parametric investigation (wavelength, sizes…).

Typical results are shown in Figure 2 for the non-local bulk current (a) and the local surface current (b) in a crossed polarization configuration (the SHG being polarized horizontally). Clearly, the SHG intensity collected in the far-field by scanning the sample under the focused excitation beam are dissimilar for both contributions. Owing to the detailed description of the real setup, the 2D map obtained theoretically (2a and 2b) can be quantitatively compared to the

experimental measurements (2c). This constitutes an invaluable tool for getting new insight in the involved physical processes. This allowed us to show that, for these very thin gold prisms (typically 25 nm), the SHG is dominated by the bulk currents owing to the large field gradients, emphasizing the importance of this contribution often disregarded.

Reference

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3. G. Bachelier et al., “Multipolar second harmonic generation in noble metal nanoparticles”, J. Opt. Soc. Am. B, 25, 955 (2008).

Figures used in the abstract

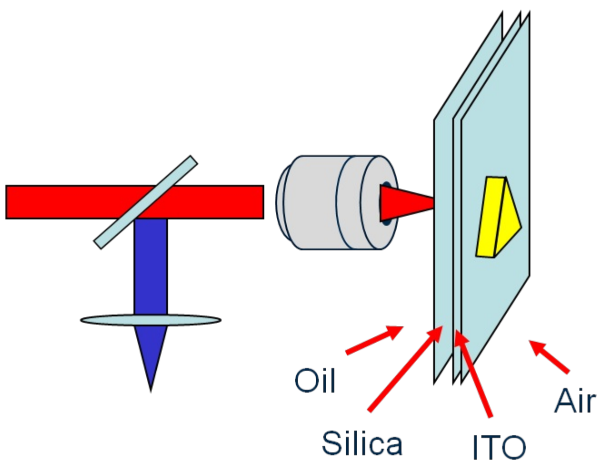

Figure 1: Experimental/simulation configuration.

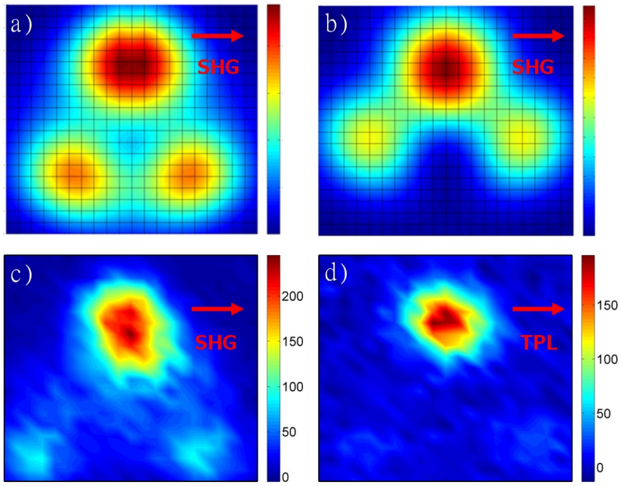

Figure 3: Intensity maps of the simulated SHG signal with nonlocal bulk (a) and local surface (b) contributions. Panels c) and d) correspond to the experimentally measured SHG and TPL intensities respectively.