Nonlinear Optics Driven by Out-of-Equilibrium Electron Dynamics in Epsilon-Near-Zero Media

A Marini[1]

[1] *Department of Physical and Chemical Sciences, University of L'Aquila, Via Vetoio, 67100, L'Aquila, Italy.*
Contact Phone: +393477757899
Contact Email: andrea.marini.tlp@gmail.com

Nonlinear (NL) radiation-matter interactions give rise to many physical phenomena particularly relevant to quantum mechanical processes and offer a promising platform to devise compact ultraviolet (UV) radiation sources. Harmonic generation (HG) is one of the main nonlinear effects amplified by photonic materials with low dielectric permittivity, also known as epsilon near zero (ENZ) media. These materials enhance coherent NL radiation-matter interactions thanks to their natural ability to bypass phase-matching requirements, thus playing a crucial role in several NL phenomena, e.g., optical switching. Currently, ENZ media are adopted to enhance NL optical effects, particularly in the near-infrared part of the spectrum, where doped semiconductors, oxides, and nitrides display this behavior. Poor materials show marked ENZ features in the UV that are promising for guidance [1] and for resonant third-harmonic generation (THG) in thin films [2], and for manipulation and guidance of extreme-UV (XUV) radiation.

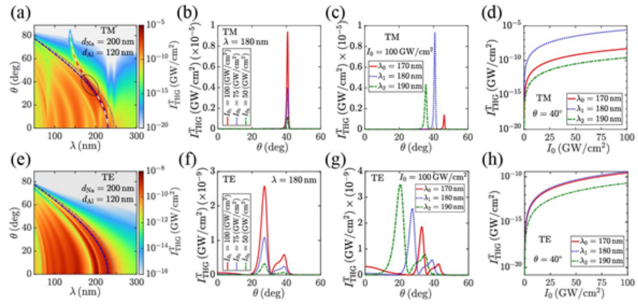

Figure 1: (a,e) Dependence of the FW intensitiy of the THG signal over the pump wavelength λ and incidence angle θ for fixed pump intensity $I_0 = 100$ GW/cm^2. The dashed white curve in (a) indicates the SPP dispersion curve, while the dot-dashed grey and blue curves indicate the Al and Na NZI dispersion curves, respectively for $\lambda_{\rm THG}$ and λ. (b,c,f,g) Dependence of over the pump incidence angle for (b,f) fixed pump wavelength $\lambda = 180$ nm and several intensities $I_0 = 50, 75, 100$ GW/cm^2, and (c,g) fixed pump intensities $I_0 = 100$ GW/cm^2 and several wavelengths $\lambda = 170, 180, 190$ nm [highlighted by the black circled area in (a)]. (d,h) Dependence of over the pump intensity I_0 for fixed incidence angle and several wavelengths $\lambda = 170, 180, 190$ nm. All plots are obtained for $d_{\rm Na} = 200$ nm and $d_{\rm Al} = 120$ nm and refer to either (a-d) TM or (e-h) TE polarisation of the impinging pump field

Here, we investigate the potential of three different setups: the first one involves a Na – based thin film [2], the second one a Na/Al-based nanosphere [3], and the third one a Na-Al bilayer [4], all with nanometer-size thicknesses. We focus on the latter to model the third-harmonic generation (THG) process produced by collision-driven NL electron dynamics. We attain this task by considering a hydrodynamical model, analytically derived in the Fokker-Planck-Landau theoretical framework in the weak coupling limit [5]. By using this set of hydrodynamical equations (HDEs) and by solving them perturbatively, we obtain an analytical expression for the collision-driven THG NL-susceptibility. Moreover, under the undepleted pump approximation, we derive an analytical expression for the forward (FW) THG intensity by both transverse electric (TE) and magnetic (TM) pump polarisation. In Fig. 1a, where we depict the dependence of the FW intensity of the THG signal over the pump wavelength λ and incidence angle θ for the latter configuration, one can see that the THG process exhibits a double resonance for TM pump excitation $\lambda_{\rm THG} = \lambda/3$ in the ENZ regime due to the coupling of the pump field enhancement and the radiation of the microscopic dipoles, producing a THG enhancement factor of 2/3 order of magnitude greater than a Na-based thin film [2]. Moreover, in the ENZ regime of Na it's possible to excite surface plasmon polaritons (SPPs) at the Na-Al interface, because in this regime this setup is analogous to an Otto configuration. Indeed, in this setup air acts as a prism, providing sufficient impulse to excite surface plasmon polaritons (SPPs) at Na-Al interface thanks to attenuated total internal reflection, see Fig. 1a, where the dotted white line represents the SPP dispersion curve. Finally, Al/Na-based nanoparticles sustain localized surface plasmon excitations enabling enhanced functionalities in nanoscale environments [4]. These results highlight the potential of NZI heterogeneous nanostructure devices in the development of integrated XUV sources.

References

[1] L Assogna, C Ferrante, Ao Ciattoni and A Marini, J. Phys.: Photonics **5**, 045001 (2023)

[2] M Silvestri, A Sahoo, L Assogna, P Benassi, C Ferrante, Al Ciattoni and A Marini, Nanophotonics **13**, 2003 (2024)

[3] M Silvestri, M Venturi, M Di Muzio, R Adhikary, C Ferrante, P Benassi and Andrea Marini, J. Chem. Phys. **161**, 054111 (2024)

[4] M Silvestri, A Sahoo, C Ferrante, P Benassi, A Ciattoni and A Marini, Phys. Rev. A **110**, 023524 (2024)

[5] A Marini, A Ciattonib and C Conti, Faraday Discuss. **214**, 235 (2019)