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Nonlinear plasmonics: from classical to quantum effects

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Modern photonic devices rely on nonlinear optical effects to carry out their functionalities. Yet, the realization of efficient nanoscale nonlinear optical components remains a chimera. In this talk, we explore three strategies based on the exploitation of plasmonic systems that might allow to overcome the main challenges and pave the way for all-optical integrated circuits.

Achieving multi-resonance mode matching: Plasmonic enhancement of nonlinear optical processes confronts severe limitations arising from the strong dispersion of metal susceptibilities and small interaction volumes that hamper desirable phase-matching-like conditions. Maximizing nonlinear interactions in nanoscale systems require simultaneous excitation of resonant modes that spatially and constructively overlap at all wavelengths involved in the process. Here, we present a hybrid rectangular patch antenna design [1] for optimal second harmonic generation (SHG) that is characterized by a non-centrosymmetric dielectric/ferroelectric material at the plasmonic hot spot. The optimization of the rectangular patch allows for the independent tuning of various modes of resonances that can be used to enhance the SHG process. Furthermore, we propose a novel configuration with a periodically-poled ferroelectric layer for orders-of-magnitude enhanced SHG at normal incidence.

Exploiting quantum effects: Second-order nonlinear optical processes may occur in the angstroms-thick layer at surfaces. At such length-scales, quantum mechanical effects come into play which could be crucial for an accurate description of plasmonic systems. Using a nonlinear quantum hydrodynamic description, we study free-electron nonlinear dynamics in plasmonic systems [2]. Our model predicts strong resonances induced by the spill-out of electron density at the metal surface. These resonances can boost SHG efficiency up to four orders of magnitude and can be arbitrarily tuned by controlling the electron spill-out at the metal surface with the aid of thin dielectric layers.

Shifting to mid-infrared frequencies: Heavily doped semiconductors have emerged as tunable low-loss plasmonic materials at mid-infrared wavelengths (2-20 μm). Because of small carrier densities in semiconductors compared to noble metals, hydrodynamic effects result strongly amplified. More precisely, a measure, l, of hydrodynamic effects can be linked to the ratio of Fermi velocity, $v_{\rm F} = \left(3\pi^2 n_0\right)^{\frac{1}{3}} \hbar/m$, over the plasma frequency, $\omega_{\rm p} = \sqrt{e^2 n_0/(m\varepsilon_0)}$, with n_0 and m being the charge concentration and the effective charge mass respectively (e, $\hbar$, and ε_0 represent the usual physical constants). Leaving out all the constants, we ultimately obtain that $l \sim v_{\rm F}/\omega_{\rm p} \propto 1/(n_0^{\frac{1}{6}} m^{\frac{1}{2}})$.

Because, both n_0 and m are much smaller in semiconductors than in noble metals, nonlocal and hence nonlinear effects are expected to be much larger. We show that contrarily to noble metals, in fact, free-electron nonlinearities in doped semiconductors can be several orders of magnitude larger than crystalline lattice nonlinearities [3].

References

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