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Electron spill-out in plasmonic systems enhances second-harmonic generation

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Second-harmonic generation in centrosymmetric metals originates from convective and Lorentz-force interactions in the free-electron fluid, as well as from the angstrom-scale thin layer near the surface where the symmetry is broken. At such length-scales classical electrodynamics fails to account for the microscopic details and consideration of nonlocal and quantum mechanical effects become crucial for accurately characterizing plasmonic optical behavior. Recently, quantum hydrodynamic theory has been used to effectively take into account the quantum effects and a very good agreement with the first-principle calculations has been demonstrated in the linear approximation [1, 2, 3]. Here we develop a new theoretical model based on quantum hydrodynamic description to study second-order nonlinearities of a plasmonic system of an arbitrary shape and size. The presented approach can efficiently handle realistic profiles associated to the ground-state electron density (see Fig. 1a), without having to rely on a simplistic idealization of the metal surface [4] Using this method we perform second-harmonic generation calculations for Na and Ag slabs. We show that the results predicted by our theoretical model for Ag slab are in very good agreement with the angle-dependent experimental data available in the literature. In the spectral analysis of the metal films, we observe strong resonances induced by the electron spill-out of electron density at the metal surface (see Fig. 1b). These resonances can boost the SHG efficiency by several orders of magnitude (see Fig. 1c). We also report that these spill-out induced resonances can be tuned at the optical frequencies by controlling the electron spill-out from the metal surface.

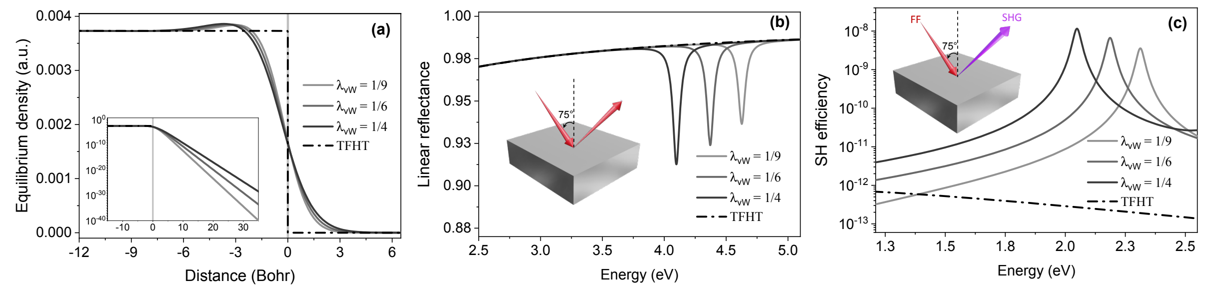

Figure 1. (a) Equilibrium electron density calculated self-consistently for different values of spill-out parameter λ_{vW}. The inset shows the decay of the electron density for each λ_{vW} and the solid grey line indicates the position of the metal interface. (b) Linear reflectance and (c) SH efficiency as a function of exciting (fundamental) energy of the incident field at $\theta = 75^\circ$ for different types of charge density profiles.

References

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