Nonlinear plasmonics in atomically-thin materials

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Nanoscale nonlinear optics has received a recent stimulus with the isolation of graphene and other atomically-thin crystals, which combine a large electro-optical response with strong intrinsic optical nonlinearities. In particular, the conical electronic dispersion of graphene boosts its nonlinear response through both intra- and interband transitions [1,2], which are predicted to be further increased by coupling to plasmons—the collective oscillations of electrons in conducting media—sustained by highly doped graphene nanostructures [3,4]. Also, transition-metal dichalcogenides such as MoS$_2$ are observed to produce efficient harmonic generation [5]. Here we study the plasmon-assisted nonlinear optical response of graphene and other atomically-thin materials. Atomic simulations provide an accurate description of such phenomena in graphene nanostructures, both in perturbative (weak field) and non-perturbative (strong field) regimes, although their computational cost is prohibitive for large systems [3]. In the perturbative regime, this limitation can be overcome by exploiting an eigenmode decomposition of the optical field in the framework of classical electrodynamics [4], yielding an analytical prescription that can be used to quantify the nonlinear optical response in 2D nanostructures. For strong external fields, the light-intensity threshold for extreme nonlinear phenomena such as saturable absorption and higher-order harmonic generation is dramatically reduced by graphene plasmons [2,3]. In this non-perturbative regime, incoherent plasmon-assisted electron heating compliments the intrinsically-large nonlinear absorption. We anticipate that these findings will elucidate the role of coherent and incoherent nonlinearities for future studies and applications of nonlinear plasmonics in atomically-thin materials.


FIG. 1. High-harmonic generation (HHG) assisted by graphene plasmons. (a) Schematic illustration of a doped graphene nanoribbon illuminated by an intense optical pulse that is resonant with the ribbon transverse dipole plasmon. The latter produces strong in-plane electric field intensity enhancement (see color scale) that boosts the generation of high-harmonics. (b) Absorption cross-section of a 20-nm wide armchair edge-terminated graphene nanoribbon doped to a Fermi energy $E_F=0.4$ eV, as predicted by atomistic tight-binding/random phase approximation (TB-RPA, red curves) and classical electrodynamics (CEM, local-RPA conductivity at 300 K, blue curves) simulations for transverse light polarization (see upper graphic). The prominent resonance feature corresponds to the transverse dipolar plasmon within the $2E_F$ optical gap. The dashed curve shows the classical simulation for the undoped ribbon. (c) Spectral decomposition of the light emission energy under illumination by a normally-incident pulse (100 fs FWHM duration, centered at the frequency $\omega_p$ of the ribbon plasmon), as calculated in the time-domain for three different pulse peak intensities (see legend) within the atomistic (TB-RPA, filled curves, $\hbar\omega_p=0.336$ eV) and massless Dirac fermion/classical electromagnetic (MDF-CEM, unfilled curves, $\hbar\omega_p=0.330$ eV) descriptions. Each curve is normalized to its own maximum value around the fundamental frequency.