Nonlinear Graphene Plasmonics

Joel D. Cox[1] and F. Javier García de Abajo[1,2]

[1]*ICFO-Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, Castelldefels, 08860 Barcelona, Spain*
[2]*ICREA-Institució Catalana de Recerca i Estudis Avançats, Passeig Lluís Companys 23, 08010 Barcelona, Spain*
joel.cox@icfo.es

Abstract
The Combination of graphene's intrinsically-high nonlinear optical response and its proven ability to support long-lived, electrically-tunable plasmons that couple strongly with light has garnered significant interest in the application of the atomically-thin material to new nonlinear nanophotonic devices. This interest is mainly reinforced by classical analyses performed using the response derived from extended graphene, neglecting finite and nonlocal effects that become important when the carbon layer is structured on the nanometer scale in actual device designs. Based on a quantum-mechanical description of graphene using tight-binding electronic states combined with the random-phase approximation, we show that finite-size effects can produce large contributions that increase the nonlinear optical response associated with plasmons in nanostructured graphene to significantly higher levels than those predicted by previous classical theories [1,2]. Motivated by this finding, we discuss and compare saturable absorption in extended and nanostructured graphene, and in particular, we study the role of near-field plasmonic enhancement to simultaneously enhance absorption and decrease threshold saturation intensities. We further explore high-harmonic generation in doped graphene nanoribbons and nanoislands, where illumination by an infrared pulse of moderate fluence, tuned to a plasmon resonance, is predicted to generate light at high-harmonics of the fundamental frequency, extending over the visible and ultraviolet regimes. Our atomistic description of graphene's nonlinear optical response reveals its complex nature both in extended and nanostructured systems, while further supporting the exceptional potential of this material for nonlinear nanophotonic devices.

References
[1] J. D. Cox and F. J. García de Abajo, Nat. Commun. **5** (2014) 5725.
[2] J. D. Cox, I. Silveiro, and F. J. García de Abajo, ACS Nano **10** (2016) 1995.

Figures

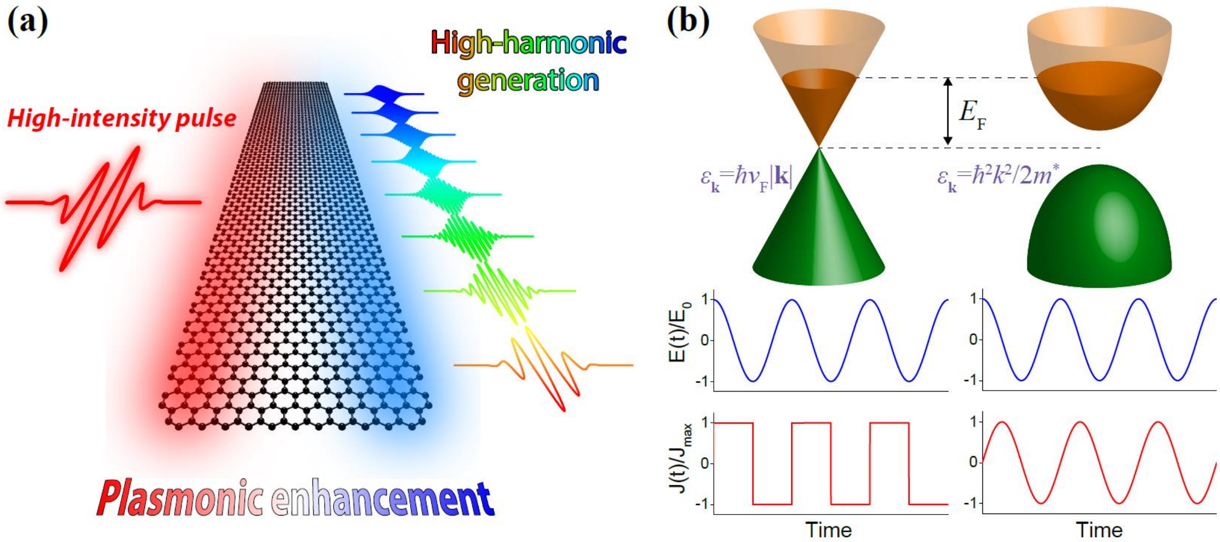

Fig. 1: High harmonic generation from graphene plasmons. (a) Illustration of a nanoribbon illuminated by an intense optical pulse that resonates with a plasmon mode in the structure, depicted by the regions of induced charge on either side of the ribbon (red and blue colors symbolize opposing induced charge densities), providing strong local fields that enhance high-harmonic generation. (b) The low-energy band structures of graphene (upper left) and a conventional 2-D semiconducting crystal (upper right) response differently to a sinusoidally-varying electric field $\mathbf{E}(t)=\mathbf{E}_0\cos(\omega t)$: In graphene, the induced current $\mathbf{J}(t)$ (lower left) has a square-wave form, containing all odd-order harmonics in its Fourier decomposition, while the semiconductor response harmonically at the driving frequency ω.