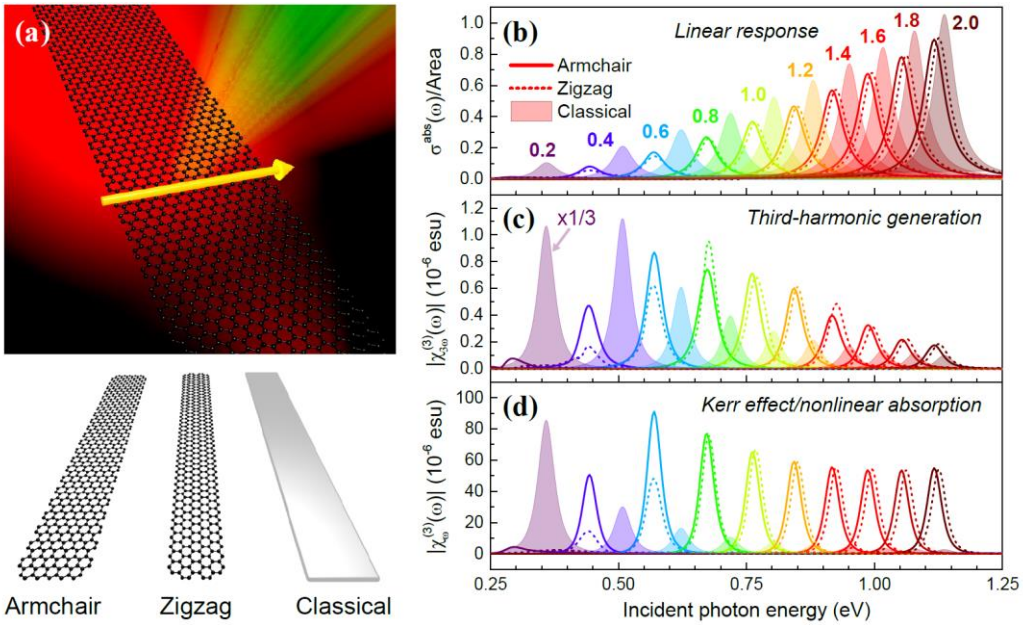
Quantum effects in the nonlinear response of graphene plasmons

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

Graphene's ability to support long-lived, electrically tunable plasmons that interact strongly with light [1], combined with its highly nonlinear optical response [2], has generated great expectations for application of the atomically-thin material to nanophotonic devices [3, 4]. Many of these expectations are reinforced by classical analyses performed using the response derived from extended graphene, neglecting finite-size and nonlocal effects that become important when the carbon layer is structured on the nanometer scale in actual device designs. Here we reveal strong finite-size and atomistic effects in the nonlinear response associated with plasmon resonances in graphene nanoribbons and nanoislands, predicted to take place from a realistic, quantum-mechanical (QM) description of these structures, beyond a semi-classical electromagnetic description, which produces dramatically different results for structure sizes up to several tens of nanometers. The discrepancy between classical and QM descriptions is particularly large for the complex third-order susceptibility associated with the Kerr effect and nonlinear absorption, where the classical theory underestimates the strength of the third-order response by several orders of magnitude even for ~40 nm structures. This is at odds with the conclusions previously drawn by examining the linear response regime, where classical and QM descriptions were found to agree well when either the plasmon energy is below the Fermi energy or when the sizes of the structure exceeds a few tens of nanometers. The QM effects reported here for such large sizes represent a rather unusual scenario in plasmonics, while they support the use of doped nanographene structures as plasmon-driven nonlinear enhancers that perform much better than previously estimated from the study of extended graphene.

[1] Z. Fang et al., ACS Nano, **7** (2013) 2388.
[2] E. Hendry et al., Phys. Rev. Lett., **105** (2010) 097401.
[3] A. N. Grigorenko, M. Polini, and K. S. Novoselov, Nat. Photon., **6** (2012) 749.
[4] F. J. García de Abajo, ACS Photon., **1** (2014) 135.



Linear and nonlinear spectral response of graphene nanoribbons. (a) We consider cw incident light linearly polarized across the ribbons (top) and present results derived from a quantum-mechanical (QM) model (tight-binding+random-phase approximation) for structures with either armchair or zigzag edges, compared with classical electromagnetic simulations (local conductivity) for a homogeneous planar sheet. **(b-d)** Linear absorption cross-section **(b)**, and third-order susceptibilities for THG **(c)** and the Kerr nonlinearity **(d)**, as obtained from the QM model for armchair (solid curves) and zigzag (dashed curves) nanoribbons, compared with classical electrodynamic simulations (filled curves). Different Fermi energies (color-coded numerical values in **(b)**, eV) are considered, taking the ribbon width as ~10 nm and the damping $\hbar\gamma^{-1}$ =50 meV in all cases.