Nonlinear Graphene Plasmonics

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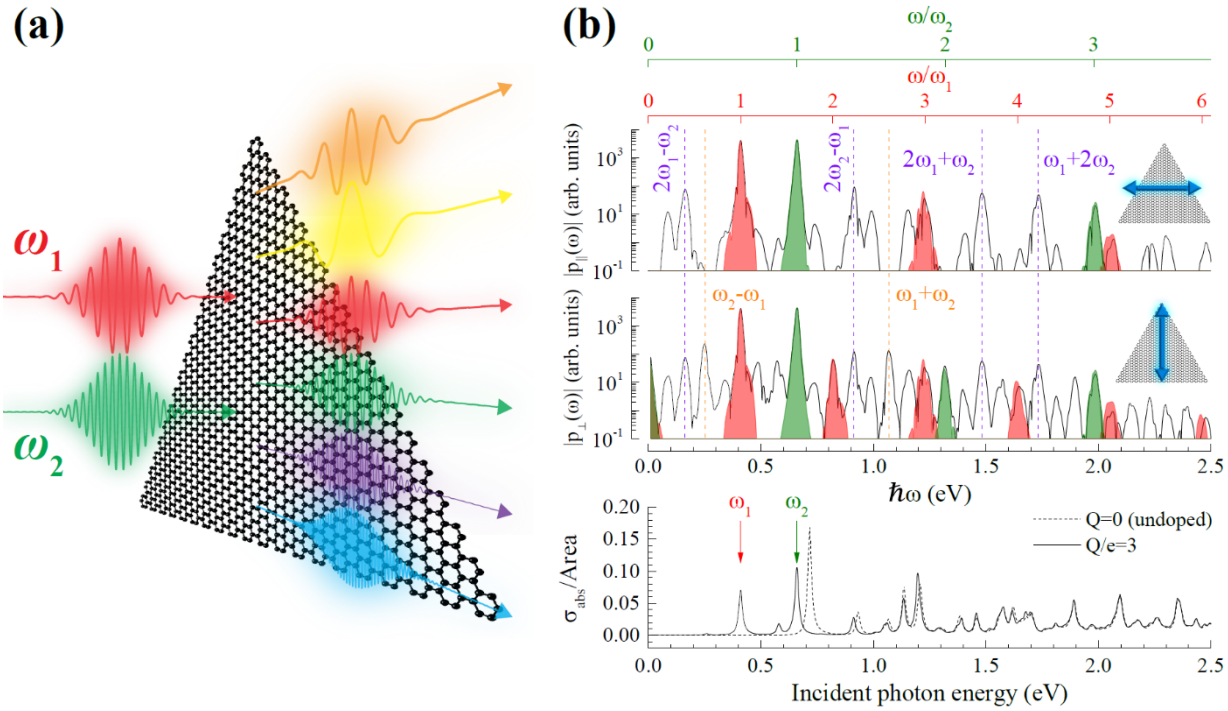
Nonlinear optical processes host a wide range of applications in nanophotonics, including all-optical signal processing, ultrafast switching, and sensing, all of which are inhibited by the inherently weak nonlinear optical susceptibilities found in conventional materials. Noble metal nanoparticles can help overcome this limitation on the nanoscale, as they enhance optical nonlinearities through the strong electromagnetic field concentration enabled by localized plasmons. Due to plasmonic enhancement, noble metal nanoparticles have demonstrated nonlinear frequency conversion with the highest recorded efficiencies per volume [1], and are generally regarded as the best available nonlinear materials.

Compared to noble metals, doped graphene nanostructures support plasmon excitations that couple more strongly with light, possess longer lifetimes, and are electrically tunable [2]. Here we show that nonlinear optical processes, including second- and third-harmonic generation, sum and difference frequency generation, and four-wave mixing, can be realized in small, doped graphene nanoislands with extraordinarily high efficiencies, surpassing those of noble metal nanoparticles with much greater size by several orders of magnitude. We model the optical response of these nanoislands using rigorous quantum mechanical simulations, taking into account significant contributions from nonlocal and finite-size effects. Due to quantum effects, a graphene nanoisland is capable of supporting multiple plasmon resonances, facilitating strong nonlinear frequency conversion for illumination with light of one or more incident frequencies that target these plasmons.

[1] M. Kauranen and A. V. Zayats, Nat. Photon. 6 (2012) 737.
[2] F. J. García de Abajo, ACS Photon. 1 (2014) 135.



Wave mixing in graphene nanoislands: (a) Illustration of a triangular graphene nanoisland illuminated by multifrequency collinear light pulses. **(b)** Spectral density of the induced dipole moment under excitation by light polarized along a direction parallel (upper panel) or perpendicular (middle panel) to one of the nanotriangle sides. The filled curves show the spectra produced by individual pulses of central frequency ω_1 (red) or ω_2 (green), while the black curves show the response when these pulses are applied simultaneously. The linear absorption cross-section of the nanoisland, which reveals the presence of multiple plasmonic resonances with electrical doping, is presented in the lower panel.