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Graphene plasmonics: THz nonlinearities beyond thermal effects

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Graphene is a very versatile material for optoelectronics or nonlinear optics in a large spectral range, in particular for THz radiation. One drawback is the low interaction volume between THz radiation and the single atomic layer, which limits the light matter interaction at elevated photon frequencies. During the recent years it has been shown that patterning graphene into plasmonic structures, e.g. ribbons or disks, can shift the rather strong optical response of free charge carriers at low frequencies to a more confined plasmonic resonance at higher frequencies. The size of the structure, in combination with the carrier density, determines the plasmon frequency, that can be tailored in a wide range. Beyond the linear absorption, the nonlinear optical properties are enhanced by about two orders of magnitude under resonance compared to unpatterned graphene. Here we present a set of studies that quantify the thermal effect when the structures are excited with strong laser pulses: the charge carriers are heated efficiently, as their specific heat is rather low, which leads to a decreased chemical potential and therewith a reduced plasmon frequency. Compared to thermal nonlinearities in conventional materials, thermal nonlinearities in graphene are very fast as the hot charge carriers cool down within several tens of picoseconds. Polarization-resolved pump-probe measurements on graphene disks revealed nonlinear absorption beyond thermal effects, i.e. plasmonic nonlinearities: thermal and nonthermal effect can be distinguished by using cross- and co-polarized pump-probe measurements. Numerical simulations considering thermal as well as plasmonic nonlinearities, match the observed signals well, giving a complete picture of the nonlinear processes in graphene plasmons.

Primary author(s) : Prof. MITTENDORFF, Martin (University of Duisburg-Essen); Dr HAN, Jeong Woo (University of Duisburg-Essen); Dr WINNERL, Stephan (Helmholtz-Zentrum Dresden-Rossendorf (HZDR)); Mr CHIN, Matthew L. (University of Maryland); Prof. MURPHY, Thomas E. (University of Maryland)

Presenter(s) : Prof. MITTENDORFF, Martin (University of Duisburg-Essen)

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