

# Terahertz Bandwidth Optical Nonlinearity of Graphene Metamaterial

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**Abstract:** We provide the first observation that hybridizing graphene with plasmonic metamaterial results in order of magnitude resonant enhancement of its nonlinear optical properties and enables THz-bandwidth optical switching at near-infrared wavelengths.

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We present the first experimental demonstration of enhancement of the graphene's nonlinear optical properties by hybridization with a plasmonic metamaterial. We show that nonlinear changes of transmission of a hybrid graphene-metamaterial structure can be resonantly increased by at least a factor of 20 while retaining an ultrafast ( $\sim 100$  fs) response time. Our approach allows to spectrally engineer and enhance graphene's nonlinearity within a broad wavelength range and is expected to result in applications in optical signal processing, where broadband ultrafast switching is a much desirable property, but also in mode-locking, noise suppression and pulse shaping, where the metamaterial design provides extensive control over the frequency dispersion of graphene nonlinearities.

Graphene, a two dimensional atomic layer of carbon atoms, exhibits unique optical properties that have attracted enormous research interest in recent years: the nonlinear optical response of graphene is non-resonant in a broad spectral range from the visible to the infrared with ultrafast sub-ps response times. Such properties establish graphene as a very attractive candidate for broadband all-optical modulation and laser mode-locking applications. However, the nonlinear response of graphene is weak requiring very high levels of excitation of the order of  $\text{GW}/\text{cm}^2$  to engage it. Hybridization of nonlinear agents with plasmonic metamaterials [1], on the other hand, is known to boost the nonlinear response due to the field enhancement provided by the metamaterial resonances [2]. Following this approach we demonstrate an order of magnitude enhancement of graphene's nonlinear properties.

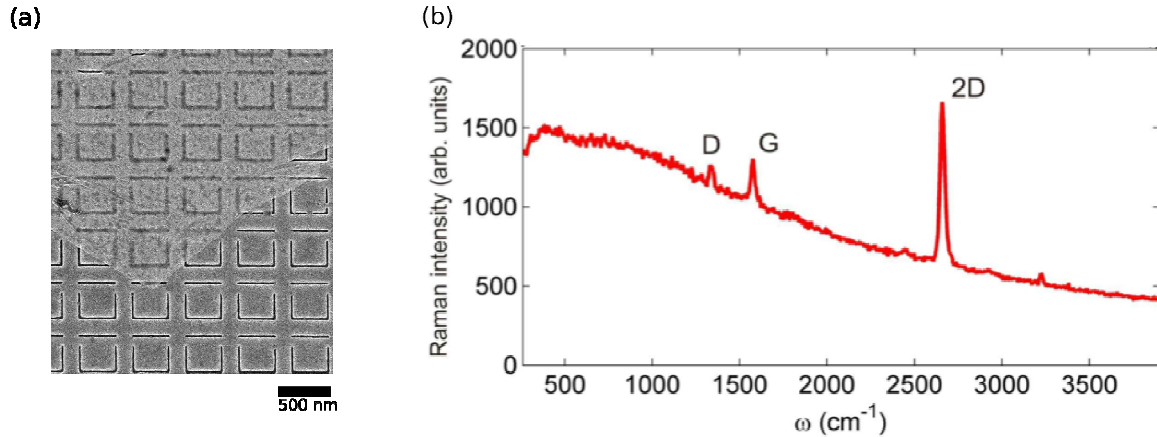


Figure 1: (a) He-ion scanning microscope image of a metamaterial array partially covered by graphene. (b) Raman spectrum of the single-layer graphene.

The metamaterial consisted of complementary metallic asymmetrically-split rings (see Fig. 1a) that exhibit strong resonances in the near infrared. The metamaterial structures were fabricated by focused ion beam milling

through a 65 nm thick gold film evaporated on a 100 nm thick silicon nitride membrane. Single layer graphene was grown on polycrystalline copper films by low-pressure chemical vapor deposition and was then transferred on the metamaterial substrate. The single-layer nature and high quality of the graphene was confirmed with Raman spectroscopy, where the 2D peak is dominant (see Fig. 1b). The nonlinear response of the hybrid structure was investigated by single-color non-collinear pump-probe spectroscopy, while the intensity of the probe pulse was 20 times weaker than the intensity of the pump pulse. Pump and probe beams had orthogonal linear polarizations.

The graphene-metamaterial structure exhibits a very strong nonlinear response in the vicinity of the metamaterial resonance (1600 nm), which decays rapidly at wavelengths away from the resonance (see Fig. 2). At the metamaterial resonance relative changes in transmission reach values as high as 0.8% at moderate pump fluence levels of  $30 \mu\text{J}/\text{cm}^2$ . In contrast, graphene on a silicon nitride membrane exhibits a much weaker, non-resonant response which remains constant at levels below 0.04% over the measurement wavelength range. This 20-fold enhancement of the nonlinear response of the bare graphene layer results from the strong field enhancement associated with the metamaterial resonance.

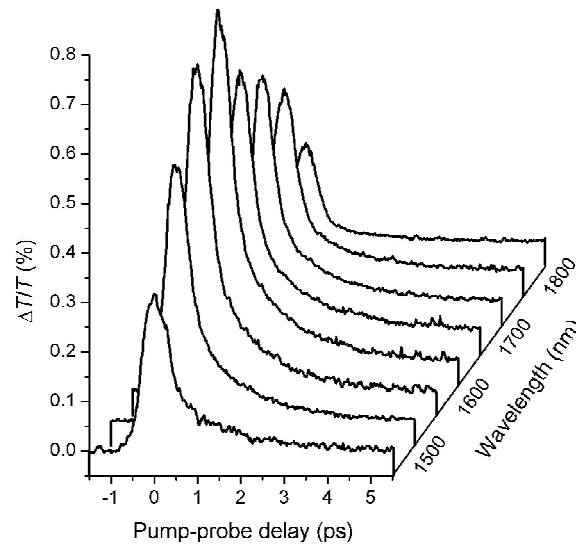


Figure 2: Non-linear response of the graphene metamaterial in the vicinity of the metamaterial resonance.

Importantly, the nonlinear response of graphene, even after hybridization with the plasmonic metamaterial, retains its ultrafast character. The transient nonlinear changes of transmission of graphene follow a bi-exponential dependence with the fast component  $\tau_1 \sim 100$  fs (not resolved in our measurements) attributed mainly to carrier-carrier intraband scattering and the slow component  $\tau_2 \sim 1$  ps attributed to the carrier-phonon intraband scattering and electron-hole interband recombination processes. The dynamics of nonlinear changes of transmission of the hybrid graphene-metamaterial structure nearly replicate the ultrafast dynamics of graphene proving that the transient nonlinear optical response of graphene metamaterial is governed by relaxation processes in graphene and is not affected by the coupling to the plasmonic resonances.

In summary, we show that metamaterials can enhance by an order of magnitude the nonlinear response of bare graphene without affecting its ultrafast relaxation times. Hybridization with metamaterials also allows to spectrally tailor the nonlinear properties of the composite material through the resonant features of the metamaterial enabling applications in optical switching, mode-locking and pulse shaping.

## References

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