Strongly Enhanced Second- and Third-harmonic Generation in Graphene Metasurfaces*

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***Abstract*— Using a new homogenization technique, we demonstrate that the effective second- and third-order nonlinear susceptibility of a graphene sheet can be enhanced by more than 100× by patterning it into a graphene metasurface. This giant enhancement of the nonlinear optical response of graphene metasurfaces is attributable to excitation of surface plasmons on the graphene components of the metasurface. This work may open new avenues to explore novel physical properties of metasurfaces based on graphene.**

I. Introduction

Since it has been first isolated from graphite [1], the unique and striking properties of graphene have generated intense research efforts to develop and synthesize new two-dimensional (2D) materials. Thus, in addition to graphene, these research endeavors have led to the discovery of several families of 2D materials, including hexagonal boron nitride, transition metal dichalcogenides, and black phosphorus [2,3]. These and other 2D materials have already had great impact both as facilitators of key advancements in fundamental research and as enablers of new devices operating in a broad spectral domain [2-4]. Equally important, the nonlinear optical properties of 2D materials have facilitated the development of new active photonic devices and the study in new settings of more fundamental phenomena, including systems with tunable Dirac points [5] and Anderson light localization at the nanoscale [6].

In this paper, using a novel homogenization method for graphene-based metasurfaces, we demonstrate that the second- and third-harmonic generation (SHG, THS) can be enhanced by more than two order of magnitude as compared to that of a homogeneous graphene sheet if the metasurface is operated at wavelengths at which surface plasmons are excited on the graphene components of the metasurface.

II. Physical Configuration and Material Parameters of Graphene Metasurfaces

The schematics of the graphene metasurface used in this work to investigate the enhancement of the THG is presented in Fig. 1, with the unit cell being depicted in Fig. 1(a). The metasurface lies in the x–y plane, and consists of a rectangular array of cruciform graphene patches. In the case of SHG, we considered a similar metasurface, the main differences being that the cruciform patches were replaced with rectangular ones and the metasurface was place on a silica substrate (n_{SiO2}=1.4), so that the inversion symmetry is broken. The symmetry axes of the array and graphene patches are along the x- and y-axes, the corresponding periods being P_x and P_y. The length and width of the arms of the cruciform are (L_x, L_y) and (W_x, W_y), respectively. The values of these geometrical parameters are $P_x=P_y=200$ nm, $L_x=L_y=180$ nm, and $W_x=W_y=75$ nm. The periods of the rectangular patches metasurface are $P_x=P_y=100$ nm and the length of the graphene patches along the y-axis is $w_y=30$ nm. The length w_x of the graphene patches along the x-axis is a free parameter that will be optimized so as to achieve a double-resonance effect, namely plasmons exist both at the fundamental frequency (FF) and SH. In both cases the graphene metasurfaces are illuminated by a normally incident, x-polarized plane wave. Moreover, the electric permittivity of graphene, ε_g, is described by the Kubo's formula [7], and is a function of frequency, ω, chemical potential, μ_c, temperature, T, and relaxation time, τ. In the case of the cruciform (rectangle) metasurfaces, μ_c=0.2 eV (μ_c=0.6 eV), τ=0.1 ps (τ=0.25 ps), and T=300 K.

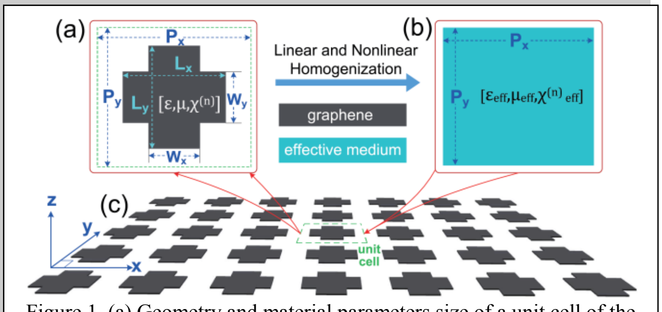

Figure 1. (a) Geometry and material parameters size of a unit cell of the metasurface. (b) The homogenized metasurface, characterized by effective linear and nonlinear optical constants. (c) Schematics of a graphene-based metasurfaces consisting of a rectangular array of graphene cruciform patches.

III. Results and Discussion

In this section we present the main results regarding the enhancement of the THG and SHG in the two graphene metasurfaces investigated in this work.

A. Enhancement of third-harmonic generation

The symmetry group of graphene lattice is D_{6h}, so that its third-order susceptibility has two non-zero components: $\chi^{(3)}_{g,xyxy}=\chi^{(3)}_{g,xxyy}=\chi^{(3)}_{g,xyyx}=\chi^{(3)}_{g,yxxy}=\chi^{(3)}_{g,yxyx}=\chi^{(3)}_{g,yyxx}$ and the dominant component, $\chi^{(3)}_{g,xxxx}=\chi^{(3)}_{g,yyyy}=\chi^{(3)}_{gra}$. It can be readily shown that the nonlinear susceptibility of graphene and that of the homogenized metasurface have the same non-

*Research supported by the European Research Council (ERC) (ERC-2014-CoG-648328).

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zero components. In order to quantify the enhancement of the nonlinear optical response of the graphene metasurface, we also computed the ratio between the dominant components of the nonlinear susceptibilities of graphene and that of the homogenized metasurface.

The results of these calculations [8], presented in Fig. 2, reveal several important conclusions. First, the dominant component of the third-order susceptibility of the homogenized metasurface is $\chi^{(3)}_{eff,xxxx}=\chi^{(3)}_{eff,1}$, but unlike the monotonous frequency dependence of graphene third-order susceptibility, the frequency dependence of $\chi^{(3)}_{eff,xxxx}$ suggests a resonant nonlinear optical response. These resonance wavelengths of the optical nonlinearity of the homogenized metasurface coincide with the wavelengths of surface plasmons excited on the graphene patches. Second, due to the plasmon resonances, the effective third-order susceptibility of the graphene metasurface is strongly enhanced as compared to that of graphene; e.g., at the wavelength λ=5.4 μm of the main resonance the enhancement is about 100×.

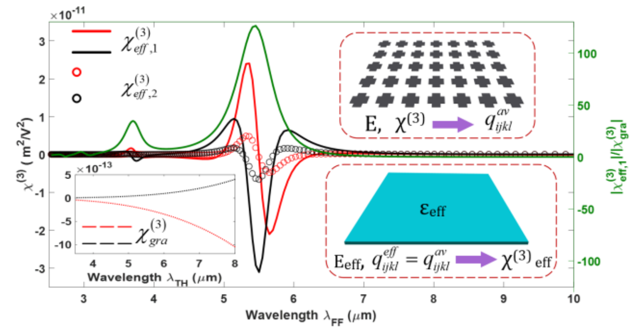

Figure 2. Wavelength dependence of the third-order susceptibility of graphene, of the two components of the homogenized graphene metasurface, and of the susceptibility enhancement factor.

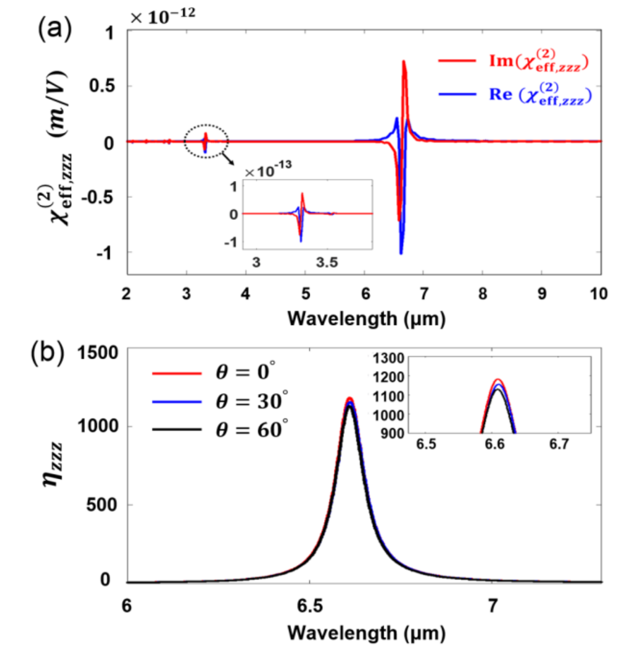

Figure 3. Top: Effective second-order susceptibility of graphene metasurface *vs*. wavelength. Bottom: Spectrum of enhancement of the second-order susceptibility, determined for several incidence angles.

B. Enhancement of the second-harmonic generation

We have applied this formalism to the SHG [9], too, the results regarding the effective second-order susceptibility of the homogenized metasurface and the enhancement of the nonlinear optical response of the metasurface being presented in Fig. 3 in the top and bottom panels, respectively. An important conclusion that can be inferred from this figure is that the second-order susceptibility tensor shows a resonant behavior around the plasmon resonance wavelength, which means that the enhanced nonlinearity of the graphene metasurfaces is plasmon driven. Moreover, the spectrum of this component of the second-order susceptibility tensor is similar to that of a nonlinear optical medium containing resonators of Lorentzian nature, which suggests that the graphene nanostructures that constitute the building blocks of metasurfaces can be viewed as meta-atoms responsible for the effective nonlinear optical response of these optical nanostructures. Since the size of these meta-atoms is much smaller than the resonance wavelength at the SH, the nonlinear graphene nanostructures investigated in this study operate in the metasurface regime. Indeed, as can be seen in Fig. 3, the nonlinear optical response of the metasurface does not depend on the angle of incidence.

IV. Conclusions

In summary, in this study we investigated the third- and second-harmonic generation in graphene metasurfaces and their homogenized counterparts. In particular, we retrieved the effective third- and second-order susceptibilities of the homogenized metasurfaces and compared them with those of a homogeneous graphene sheet. Our analysis revealed that the nonlinear optical response of graphene metasurfaces with nano-sized graphene constituents can be enhanced by more than two orders of magnitude.

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