Efficient Third Harmonic Generation with THz Graphene Metasurfaces

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Abstract. We theoretically study graphene-based metasurfaces for efficient third-harmonic generation in the THz regime. The graphene sheet is judiciously patterned into patch and cross geometries, in order to support sharp metasurface resonances at the fundamental and third harmonic frequencies. The reported conversion efficiencies reach -19dB (1.2%).

1. Introduction

Graphene is a two-dimensional (2D) material that possesses attractive properties for linear and nonlinear photonic applications [1], since it is characterized by a strong third-order nonlinearity and its linear and nonlinear conductivities can be tuned electrically. In addition, graphene can support tightly-confined surface plasmons in the technologically important THz regime. In particular, the short wavelength of propagating graphene plasmons enables the design of metasurfaces (MSs) supporting higher-order resonances, while the lattice constant remains subwavelength even for higher harmonic frequencies, avoiding diffraction effects (i.e., higher diffraction orders becoming propagating).

In this work, we design graphene-based MSs for efficient third harmonic generation (THG) in the THz regime. The design strategy is based on exploiting multiple resonances of the metasurface [2] and placing sharp resonances in the spectral vicinity of the fundamental (FF) and third harmonic (TH) frequencies, in order to harness the associated spatiotemporal energy confinement and boost the conversion efficiency (CE). This strategy has been termed double-resonant enhancement in Ref. [3]. Having as a reference the CE of -26dB (at input intensity of 0.1 MW/cm²) reported for graphene ribbons on a metal-backed dielectric substrate in Ref. [4], we investigate and propose structures with 2D patterning, in order to further confine the plasmons and produce stronger local fields, as well as to have the possibility for polarization insensitive response in the x and y axes. By careful alignment of the fundamental and third-harmonic frequencies with MS resonances, we achieve efficiencies as high as -19dB (1.2%).

2. Graphene metasurfaces for third harmonic generation

We first study rectangular patches with size \( w_x \times w_y \) in a square lattice [figure 1(a)]. The lattice constant is \( a = 3.88 \mu m \). The patches reside on a dielectric substrate of thickness \( h = 6.3 \mu m \), backed by a gold backreflector. A parametric study is conducted with respect to the length and width of the...
resonating patches, \( w_x = r_x \times a \) and \( w_y = r_y \times a \), with \( r_x \) and \( r_y \) varying from 0.1 up to 0.95. The conversion efficiency is calculated under CW conditions with the frequency-domain finite element method (FEM) by using two linear simulations at the FF and TH frequencies [4, 5] (see Methods). Control over the underlying resonant frequencies is exerted by varying \( w_y \) and consequently engineering the dispersion of the propagating plasmon; modifying \( w_x \) changes the length of the finite waveguide segment and specifies the frequencies satisfying the resonant condition. The optimum point in the \( w_x - w_y \) map results in a CE of -19dB (1.2%).

However, this performance is obtained only for the \( E_x \) polarization, since after optimization we find \( r_x \neq r_y \) meaning that the resulting patch is rectangular. To achieve isotropic response, we investigate different geometries such as the cross geometry in figure 1(b), which, following a similar parametric optimization procedure, leads to a THG conversion efficiency of -20dB (1%). To unveil the physical mechanism underlying the optimum point we plot the linear reflection and absorption spectrum (power coefficients for plane-wave incidence), as shown in figure 2. The operating frequency \( \omega_{FF} \) is positioned at the first absorption peak (2.2 THz marked in figure 2 with a solid arrow), associated with the fundamental resonance of the structure. The TH frequency \( \omega_{TH} = 3 \times \omega_{FF} \) (6.6 THz marked in figure 2 with a solid arrow) is also well aligned with an absorption peak of the MS. These absorption peaks are associated with underlying resonances of the metasurface, as verified through eigenvalue simulations [6]; the respective eigenfrequencies are marked in figure 2 with dashed (red) arrows.

**Figure 1.** Graphene-based metasurfaces for efficient third harmonic generation. (a) Rectangular graphene patches reside on a metal-backed dielectric substrate. A conversion efficiency of 1.2% is attained for \( E_x \) incident polarization. (b) Cross geometry of patches for isotropic response. The attained conversion efficiency is 1% for either \( E_x \) or \( E_y \) incident polarization.

**Figure 2.** Linear plane-wave scattering spectrum (power coefficients) for cross geometry. Plotted data correspond to the optimum structure of the cross geometry with \( r_x = 0.9 \) and \( r_y = 0.6 \). Two resonances of the metasurface are in close proximity to the fundamental and third harmonic frequencies. The resonances are identified by the absorption peaks and verified through eigenvalue simulations (dashed arrows).
3. Nonlinear modelling framework for continuous wave operation

3.1. Simulation details
For assessing the nonlinear response of the metasurfaces under study we perform full wave 3D electromagnetic simulations using COMSOL Multiphysics. A single unit cell is simulated with periodic boundary conditions at the $x$ and $y$ boundaries and input/output ports at the $z$ boundaries. In order to simulate the THG nonlinear response under continuous wave (CW) illumination, we can decouple the nonlinear problem into two linear frequency domain simulations at the fundamental (FF) and third harmonic (TH) frequency [4,5]. In the first step, the metasurface is illuminated at the fundamental frequency, inducing currents on the graphene patches. Via frequency mixing mediated by the third order conductivity, a nonlinear current is produced oscillating at the third harmonic. This acts as a source for the second simulation radiating a plane wave at $3\omega$. The THG conversion efficiency (CE) is given by $P_{TH}/P_{FF}$, where $P_{TH}$ is the power outflow at the third harmonic frequency and $P_{FF}$ is the injected power at fundamental frequency.

3.2. Graphene properties
Graphene is naturally modelled as an infinitesimally thin material via a surface current boundary condition. It is a highly dispersive material in the THz regime; its linear conductivity can be described by a Drude dispersion model:

\[ \sigma^{(1)} = -\frac{iD}{(\omega - \text{Re}F)}, \]  
\[ D = \frac{q^2\mu_c}{\hbar^2}, \]

where $\tau = 500$ fs is the electron relaxation time, $q$ is the elementary charge, $\mu_c = 0.3$ eV is the chemical potential and $\hbar$ is the reduced Planck’s constant.

The nonlinear current density that develops on graphene and oscillates at the third harmonic frequency is given by:

\[ J_{3\omega} = \sigma^{(3)} \cdot E_{||} \cdot E_{||} \cdot e^{j3\omega t}. \]  

Graphene’s third order conductivity formula is taken from Cheng et al. [7]

\[ \sigma^{(3)} = \frac{i\sigma_0(h\omega_F)^2}{48\pi(h\omega_F)^4} T\left(\frac{h\omega_F}{2\mu_c}\right), \]

where

\[ \sigma_0 = q^2(4\hbar)^{-1}, \]
\[ T(x) = 17G(x) - 64G(x) + 45G(3x), \]
\[ G(x) = \ln\left|\frac{1+x}{1-x}\right| + (i\pi\theta|x| - 1), \]

in which $u_F$ is the Fermi velocity and $\theta(x)$ the Heaviside step function.
4. Conclusion
We have proposed nonlinear THz metasurfaces based on 2D-patterned graphene patches atop a metal-backed dielectric substrate for efficient third harmonic generation. The two-dimensional patterning tightly confines the supported graphene plasmons and results in strong local fields, boosting the nonlinear conversion process. Both a rectangular and a cross graphene patch geometry have been studied, and are found to exhibit high conversion efficiencies of approximately -20dB (1%). The cross geometry leads, moreover, to polarization-independent (isotropic) response. Toward future work, additional metasurfaces combining graphene with metallic structures have been investigated and preliminary results indicate even higher CEas, reaching values as high as -9dB (12%).

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