Numerical Simulation of Nonlinear Effects in Multilayer Graphene Metasurfaces in Terahertz and Infrared Ranges

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Abstract— Using the developed numerical-analytical method the nonlinear effects in multilayer metasurfaces of graphene ribbons with finite length was investigated numerically at the resonant frequencies of the surface plasmon–polariton (SPP) modes in the terahertz (THz) and infrared (IR) frequency ranges. It is shown that the efficiency of nonlinear effects (the frequency multiplication) can be enhanced by increasing the number of array layers, its packing density, dielectric mirrors in substrates and tuned by changing the chemical potential of graphene (applying an external electric field).

Keywords— frequency multiplication, surface plasmon–polariton, multilayer metasurface, graphene ribbons, resonant frequency

I. Introduction

HIGHLY doped graphene has recently emerged as an appealing platform for plasmonics due to its unique optoelectronic properties, which give rise to relatively long-lived, highly confined, and actively tunable plasmon resonances that mainly appear in the THz and IR frequency regimes [1]. Intriguing and unusual physical properties of graphene offer remarkable potential for advanced, photonics-related technological applications, particularly in the area of nonlinear optics at the deep-subwavelength scale [2]. The combination of large field confinement and enhancement produced by graphene plasmons, the strong nonlinear response in this atomically thin material, its extraordinary sensitivety to external dc fields, and its low electronic heat capacity enable applications in important frontiers of nanophotonics and, in particular, in the achievement of unity-order nonlinear effects at the nanoscale [1, 3, 4]. Furthermore, electrostatic biasing in multilayer graphene is enhanced with respect to single-layered one due to the redistribution of carriers over different layers, thus extending the spectral tuning range of the plasmonic structures [5].

The proposed research is directed toward the numerical investigation into the nonlinear interactions of electromagnetic waves with multilayer metasurfaces, based on graphene nanostructures, using mathematical modeling by solving the nonlinear Maxwell`s boundary problems combine with a model of the graphene electronic conductivity. The goal is to investigate nonlinear physical phenomena and effects, diffraction phenomena, geometry and size effects, related to the design of prospective reconfigurable nonlinear, parametric devices based on the multifunctional graphene metasurfaces in the THz and IR frequency ranges.

II. MATHEMATICAL MODEL NUMERICAL APPROACH

The mathematical model of nonlinear interactions of electromagnetic waves with multilayer graphene metasurfaces are based on the solution of nonlinear diffraction boundary problems simultaneously with a model of nonlinear graphene surface conductivity.

The perturbation method is used to solve the nonlinear diffraction problem. First the linear problem of the diffraction of electromagnetic waves on the multilayer graphene ribbon metasurfaces was solved using two methods [6]. The

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numerical approaches are based on different methods: the method of the approximate boundary conditions [7] and the method for solving a volume integrodifferential equation [8, 9].

The model of surface conductivity σ of graphene as a nonlinear function of the intensity of electric field $\boldsymbol{E}$ is

$$\sigma = \sigma^{(1)} + \sigma^{(3)} \left|\vec{E}(x, y, t)\right|^2, \quad (1)$$

where $\sigma^{(l)}$ is the linear part of the conductivity, determined by the Kubo formula [10], $\sigma^{(3)}$ is a nonlinear conductivity of the third order [11]

$$\sigma^{(3)} = -i\frac{3}{32}\frac{e^4 v_F^2}{\omega^3 \hbar^2 \mu_c}, \quad (2)$$

here e is the electron charge, μ_c is the chemical potential (Fermi energy), $v_F = 10^6$ m/s is the Fermi velocity, ω is the incident wave frequency, $\hbar$ is the reduced Planck's constant.

The computational algorithm was created at several stages:

- at first electromagnetic fields in the multilayer dielectric are determined using the method described in [8];
- the boundary conditions on the graphene ribbons with the linear surface conductivity $\sigma^{(l)}$ for $\sigma^{(3)} = 0$ in (1) are satisfied;
- as a result a system of paired integral equations (SPI Eqs.) about the unknown surface current densities on the ribbons is obtained. The SPIEqs. is close to the PIEqs. [7] for the case of one graphene ribbon in the unit cell of the single-layer metasurface.
- SPIEqs. is solved by using the Galerkin's method [7];
- electromagnetic fields (on the ribbons and in the far radiation zone) are determinated by solving SPIEqs;
- the obtained solution is used to find the nonlinear surface currents on the ribbons and simulate the nonlinear effects in the multilayer graphene metasurfaces.

III. RESULTS OF NUMERICAL MODELLING

The multilayer metasurfaces (inset in Fig. 1) consist of several two-dimensional periodic arrays ($d_x = d_y$ - periodicity along the x, y axes) of graphene ribbons of finite length (width w, length l) located on dielectric layers (SiO_2) with a refractive index n = 1.45, thickness $d_1 = d_2$ and d_3 on a semi-infinite substrate n = 1.77. Graphene parameters: T = 300 K, τ = 1 ps, μ_c = 0.3 eV.

The results of numerical simulation of the nonlinear effect of THz frequency multiplication: the normalized power T_3 of the triple frequency wave radiated in the forward direction and the absorption coefficient P depending on the frequency of the incident TEM-wave are shown in Fig. 1 for s-, p- polarization (Fig. 1a) and for different number of array layers (Fig. 1b). The T_3 values are calculated by normalizing to the incident power density P_0.

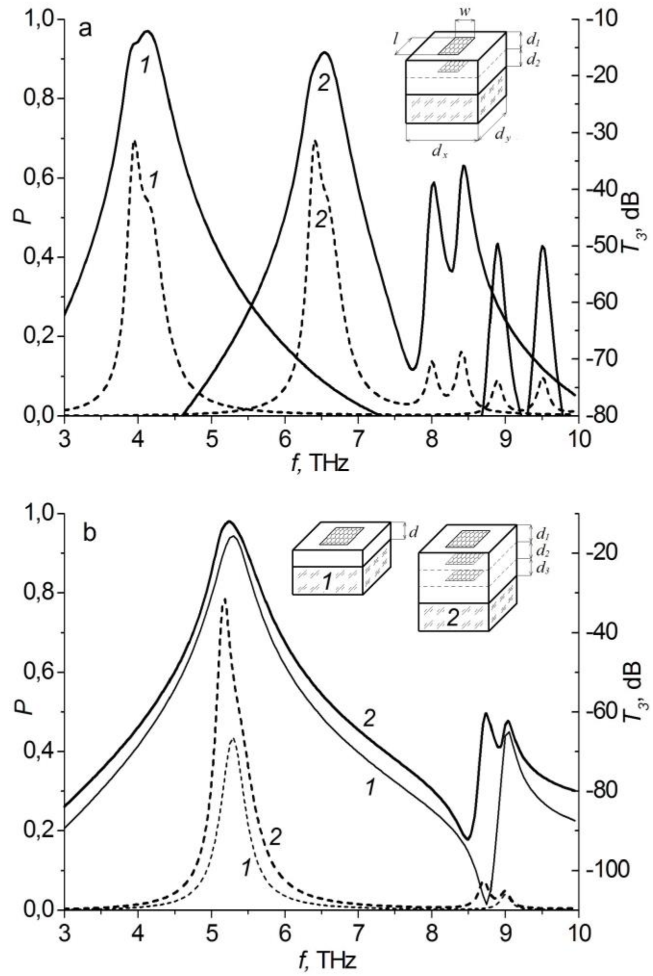

Fig. 1. Efficiency of THz frequency multiplication (solid lines) by multilayer graphene ribbon metasurfaces and the linear absorption spectrum P (dashed lines): a) for the 2-layered array of rectangular graphene ribbons (inset): curves *1* – *s*-polarization, d_1 = 9 μm, d_2 = 4 μm; curves *2* – *p*-polarization, d_1 = 5 μm, d_2 = 3 μm; w_1 = 1.4 μm, l_1 = 2.5 μm, $w_{2,3}$ = 1.1 μm, $l_{2,3}$ = 2.1 μm; b) for the different number of layers of square graphene ribbon array: curves *1*- for single-layered – $w_1 = l_1$ = 1.9 μm, d =11 μm; curves *2* – 3-layered - $w_1 = l_1$ = 1.9 μm, $w_{2,3} = l_{2,3}$ = 1.5 μm, $d_1 = d_2$ = 4 μm, d_3 = 2 μm; $d_x = d_y$ = 3.5 μm. μ_c = 0.3 eV, $P_0 = 10^{-3}$ mW / mm^2.

The efficiency of frequency multiplication increases by several orders of magnitude when the frequency of the incident wave is close to the resonance frequencies of the strongly-confined SPP modes (Fig. 1a) allowing for significant field enhancement which, combined with the strong nonlinearity of graphene.

The maxima of the absorption coefficient P in Fig. 1a correspond to resonances determined by SPP modes in graphene ribbons of finite length and depend on the polarization of the incident wave. For s-polarization, the first resonant frequency is determined by the fundamental SPP mode (resonance of the electric current along the wide side of the ribbons). For p-polarization, this is the resonance of the current along the narrow side of the ribbons, and the resonance

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frequency is higher than for s-polarization (Fig. 1a). Several (two) resonance frequencies of the nearest higher SPP modes are observed (Fig.1 a).

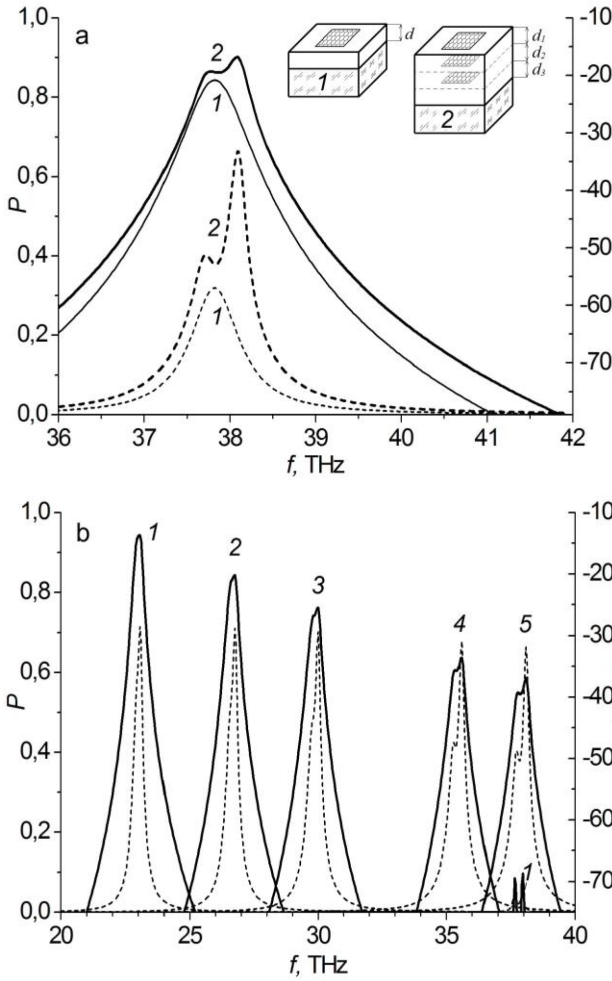

Fig. 2. a) Efficiency of frequency multiplication (solid lines) by multilayer metasurfaces of square graphene ribbons in the mid-IR regime and the linear absorption spectrum *P* (dashed lines) for the different number of array layers (inset): curves *1*- single-layered– $w_1 = l_1$=100 nm, d =1.4 µm; *2* – 3-layered - $w_1 = l_1$=100 nm, $w_{2,3} = l_{2,3}$=75 nm, $d_1 = d_2 = d_3$ = 1.4 µm; μ_c = 0.8 eV, $P_0 = 10^{-7}$ mW/mm^2; b) frequency multiplication tunability (3-layered array) with the various values of chemical potential: curves *1* – μ_c = 0.3 eV, $d_1=d_2=d_3$= 2.2 µm; *2* – μ_c = 0.4 eV, $d_1=d_2=d_3$ = 1.9 µm; *3* – μ_c = 0.5 eV, $d_1=d_2=d_3$= 1.7 µm; curve *4* – μ_c = 0.7 eV, $d_1=d_2=d_3$ = 1.5 µm; *5* – μ_c = 0.8 eV, $d_1 = d_2 = d_3$ = 1.4 µm; $d_x = d_y$ = 0.2 µm. $P_0 = 10^{-8}$ mW / mm^2.

In Fig. 1b shows the results of numerical simulation of the THz frequency multiplication in the multilayer metasurfaces with a different number of layers of graphene ribbon array. With an increase in the number of array layers, the efficiency of nonlinear effect can be significantly enhanced as compared to single-layer one (Fig. 1b).

As the sizes of graphene ribbons and the periodicity of array decreases, the SPP resonance frequency and, therefore, the maximum of normalized power T_3 of the triple frequency wave shift towards higher THz frequencies and IR frequency range (Fig. 2, 3).

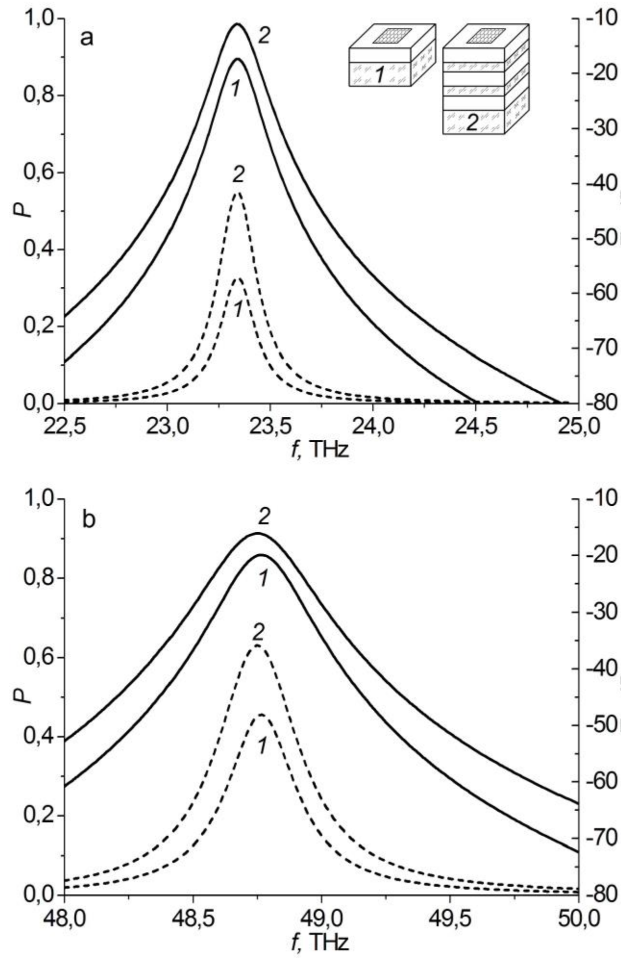

Fig. 3. Enhancement of frequency multiplication by multilayer graphene ribbon metasurfaces using the dielectric mirrors in substrates: a) in mid-IR frequency range *(w = l* = 100 nm, *d* (*n*=1.45) = 2.2 µm, *d* (*n*=1.77) = 1.8 µm, $d_x = d_y$ = 400 nm, μ_c = 0.3 eV, $P_0 = 10^{-7}$ mW/mm^2); b) in near-IR frequency range (*w* = *l* = 20 nm, *d* (*n*=1.45) = 1 µm, *d* (*n*=1.77) = 0.85 µm, $d_x = d_y$ = 400 nm, μ_c = 0.3 eV, $P_0 = 10^{-11}$ mW/mm^2. Solid lines – normalized power T_3 of the triple frequency IR wave; dashed lines - the linear absorption spectrum *P*. Curves *1*- metasurface with the dielectric mirror, *2* - without the dielectric mirror.

Fig. 2a shows an enhancement of frequency multiplication effect in the multilayer metasurfaces using several layers of graphene ribbon arrays in the mid-IR frequency range.

Fig. 2b shows the triple-frequency regime of IR wave through the multilayer graphene ribbon metasurfaces at different chemical potential values. Upon decreasing the value of chemical potential the SPP resonance frequency decrease but the efficiency of frequency multiplication significantly increases in the mid-IR frequency range. Thus, tunability of the resonant operating frequencies of reconfigurable frequency multipliers by changing chemical potential (applying an external electric field) without changing its geometry and sizes in the IR frequency range is shown.

Another possibility of enhancement of nonlinear effects in the graphene metasurfaces is realized using multilayer substrates (dielectric mirrors) and demonstrated by the correct choice of the layering, the thickness of the dielectric layers and their dielectric constants in mid- and near-IR frequency ranges (Fig. 3a, b). The normalized power T_3 of the triple frequency wave increases with increasing number of dielectric mirror's layers in mid- and near-IR frequency ranges (Fig. 3a, b).

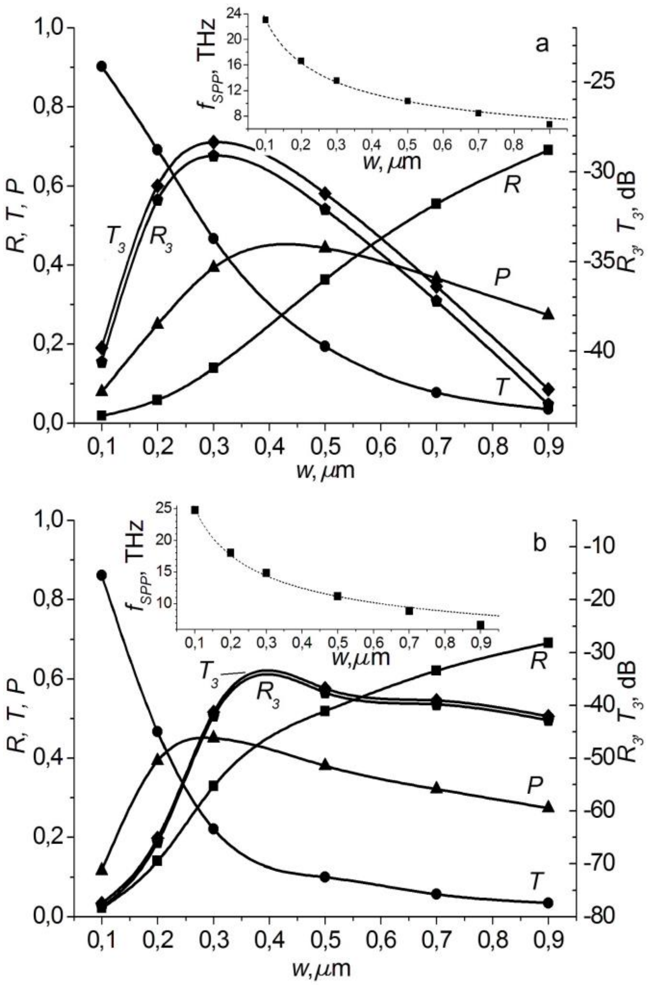

Fig. 4. Dependencies of the normalized power of the triple frequency wave radiated in the forward T_3 (transmission) and an opposite direction R_3 (reflection) (the nonlinear operation regime) and the transmission T, reflection R, absorption P coefficients (the linear regime) at the resonance THz,-IR frequencies on graphene ribbons sizes of metasurface (№*1* insert in Fig. 3a): a) for square shape ($w = l$); b) for rectangular shape ($w \neq l$, l = 0.9 μm, *p*-polarization). $d_x = d_y = 1$ μm, $\mu_c = 0.3$ eV, $P_0 = 10^{-6}$ mW/mm^2.

The results of calculations of the normalized power of the triple frequency wave radiated in the forward T_3 (transmission) and backward R_3 (reflection) directions (the nonlinear operation regime) and the transmission T, reflection R, absorption P coefficients (the linear regime) at the resonance frequencies in the THz and mid-IR regimes depending on sizes of square and rectangular graphene ribbons of multilayer metasurfaces are shown in Fig. 4.

Each ribbon size in Fig.4 corresponds to its own resonant frequency of the SPP fundamental mode f_{SPP} and the quarter-wavelength thickness d of the dielectric layer. As the ribbon size increases, the f_{SPP} frequency decreases (symbols on the insets). This decreasing is approximated by the function $f_{SPP} = k/\sqrt{w}$ (dashed lines symbols on the insets), where the coefficient k is determined for a particular type of metasurface by substituting the width w and the corresponding frequency f_{SPP} [12].

Fig. 4 shows the closely spaced regions of the maxima of normalized power T_3 of the triple frequency and the absorption coefficient P. For metasurfaces with square ribbons (Fig. 4a) the efficiency of frequency multiplication T_3 is maximal at $w \sim$ 0.2…0.5 μm, and the absorption P is maximal at $w \sim$ 0.25…0.7 μm. For rectangular ribbons (Fig. 4b) the region of maxima T_3 at $w \sim$ 0.3…0.5 μm, P at $w \sim$ 0.2…0.5 μm. A high level of normalized power T_3 of the triple frequency can be obtained at sufficiently low values of the width w with respect to the periodicity d_x due to an increase in the ribbons' current density. It was also verified that when the periodicity of the metasurface increases with the same ratio $w/d_x(l/d_y)$, the maxima of the absorption and the efficiency of frequency multiplication are in the same regions as in Fig. 4. Hence, for the SPP fundamental mode the maximum level of nonlinear effects is observed for the geometric dimension ratio of around $w/d_x(l/d_y) \sim 0.3$.

Thus, by increasing the number of layers (Fig. 1a, 2a) and the packing density and sizes of graphene ribbon arrays (Fig. 4), it is possible to control the change in the resonant operating frequencies of the frequency multipliers and the efficiency of frequency multiplication by multilayer graphene metasurfaces in the THz and IR frequency ranges.

IV. CONCLUSION

Using the developed numerical-analytical method we numerically demonstrate the enhanced frequency multiplication nonlinear effect in multilayer graphene ribbon metasurfaces in THz and mid-IR ranges.

Here we report on strong optical nonlinearities in multilayer graphene ribbon metasurface, which demonstrate an enhancement by three orders of magnitude in the third-harmonic signal compared with that of single-layered one.

It is shown that multilayer graphene metasurfaces graphene present opportunities for enhancing nonlinear optical processes at the nanoscale by using the correct choice of parameters, thus of particular array layering, the thickness of dielectric layers and its dielectric permeability.

By varying geometrical sizes of ribbons, the periodicity and the packing density of graphene ribbon arrays, enhanced the frequency multiplication and wave mixing can be realized over a wide spectral range in the in THz and mid-IR or. near IR ranges.

The improved tunability of multilayer ribbon metasurfaces applying an external electric field should enable a plethora of future IR plasmonic devices with high optical performance and wide tunability [4].

The nonlinear effects can be used in variety of applications including THz, IR multipliers and generators, nonlinear IR

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modulators, IR multiplexors, IR logic, and sensing devices [1].

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