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Graphene-Tuned Plasmonic Metasurface Based on Split-Ring Resonators for Enhanced Second Harmonic Generation in the Mid-Infrared

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Abstract— The second harmonic generation (SHG) plays a crucial role in a wide range of optical and photonic applications. In this study, we propose and optimize a novel plasmonic nanoantenna based on a split-ring resonator (SRR) configuration engineered to enhance SHG efficiency. The designed metasurface consists of a quadruple SRR arrangement, in which both near-field and far-field responses confirm the presence of strong second-harmonic radiation. The orthogonal orientation of the SRR elements enables polarization-orthogonal field coupling, resulting in SHG resonances at 60 THz and 130 THz . Due to the symmetric layout of the nanoparticle array, the structure exhibits polarization-independent behavior. Furthermore, a graphene layer is integrated into the metasurface to provide dynamic tunability of the resonance frequencies through adjustment of its chemical potential. To validate the effectiveness of the SRR configuration, a comparison is made with an array of rectangular nanoparticles lacking split gaps. The results indicate that the introduction and placement of gaps are essential for controlling the near-field localization and the corresponding far-field radiation pattern. These engineered gaps facilitate the excitation and manipulation of dipolar and quadrupolar plasmonic modes, thereby enabling and enhancing SHG. Overall, the proposed metasurface demonstrates the ability to tailor nonlinear optical responses through geometric design and material integration.

Keywords— Graphene, Plasmonic, Nonlinear Optics, Second Harmonic Generation.

I. INTRODUCTION

Second harmonic generation (SHG) is a well-known nonlinear optical phenomenon that has attracted significant interest in plasmonic nano antennas [1] as well as in other engineered photonic platforms, including photonic crystal waveguides [2] and optical absorbers [3],[4]. SRR is a type of metamaterial characterized by configuration that enables extraordinary electromagnetic properties which are utilized in sensors,nano antennas and etc[5],[6],[7]. The primary motivation for employing SHG in nano scale structures lies in its ability to dramatically enhance the local electromagnetic field—both in the near-field and the radiated far-field beyond what is achievable in conventional plasmonic systems [8]. In addition, SHG has been investigated as an effective mechanism for phase control in

advanced nano photonic devices [9]. As a fundamental nonlinear optical process, SHG plays a critical role in laser physics and has been incorporated into a wide range of applications, such as optical diodes [10] and optical modulation systems [11]. SHG can be realized either by utilizing nonlinear material composites [12] or by engineering complex resonant structures, such as nanoparticle arrays with tailored field enhancement characteristics [13]. In general, SHG is associated with strong near-field amplification at the fundamental resonance and increased radiation at the second harmonic frequency [14]. This property has been leveraged to design polarization-independent multi-Fano resonant systems [15]. Notably, structures exhibiting SHG typically demonstrate distinct near- and far-field profiles compared to conventional nano antennas due to the nonlinear mode conversion involved [16]. Plasmonic nano antennas derive their unique electromagnetic response from localized surface plasmon resonances [17]. By introducing split gaps [18] or employing hybridized plasmonic coupling schemes [19], it is possible to engineer multi-band resonances with controllable field confinement. The strategic placement of such gaps is particularly effective for enhancing the near-field intensity and enabling nonlinear frequency conversion [20]. Furthermore, metasurface-based nano antenna architectures, incorporating metamaterial-inspired geometries, have recently emerged as promising candidates for achieving efficient SHG within the optical and infrared regimes [21]. In recent developments, SHG-active metasurfaces have demonstrated high nonlinear response [22], and plasmonic nano antennas supporting SHG have been explored for sensitive biosensing and enhanced energy harvesting applications [23]. In this work, a symmetric nano antenna configuration is investigated to enhance SHG and improve radiation performance in the mid-infrared region. Initially, a basic structure composed of four rectangular loops is examined. However, analysis of its extinction cross-section, near-field distribution, and far-field pattern demonstrates that this geometry is not suitable for supporting SHG due to insufficient field confinement and distorted mode formation. To enable SHG, engineered gaps are introduced into the structure, forming a refined SRR design. The modified geometry effectively controls the local field distribution and resonance behavior, resulting in two distinct resonances at 60 THz and 130 THz corresponding to dipolar and quadrupolar radiation patterns, respectively, confirming SHG activity. The symmetric configuration also ensures polarization-independent response under varying incident field orientations. Furthermore, a graphene layer is integrated beneath the nano antenna to enable dynamic tuning of resonance frequencies through adjustment of its chemical potential. Comparative evaluation confirms that, unlike the original rectangular-loop array, the proposed SRR structure provides strong near-field enhancement, well-defined mode profiles, and stable far-field SH radiation suitable for mid-infrared photonic applications.

II. NANO ANTENNA DESIGN WITH SHG ATTRIBUTES

SHG in plasmonic nanoparticles occurs when the localized surface plasmon resonance is engineered such that a higher-order mode oscillates at twice the frequency of the fundamental mode (2ħω) [24]. The SH response originates from the second-order nonlinear polarization:

$$\nabla^2 E(r,\omega) + \frac{\omega^2}{c_0^2} E(r,\omega) = \mu_0 \omega^2 P(r,\omega) \quad (1)$$

$$P(r,\omega) = \varepsilon_0 \chi^{(1)}(\omega) E(r,\omega) \quad (2)$$

$$P(t) = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} EE + \varepsilon_0 \chi^{(3)} EEE + ... \quad (3)$$

Where ε_0 is the vacuum permittivity, $\chi^{(1)}$is the nonlinear susceptibility, and E is the incident electric field [25]. Fig. 1 illustrates the initial nano antenna structure, incorporates four split-ring resonators arranged orthogonally. The introduced gaps function as capacitive regions, while the metallic loops provide inductive behavior, enabling strong near-field confinement. These gaps serve two key purposes: (1) shaping the near-field distribution and corresponding far-field radiation pattern, and (2) controlling the interaction between dipolar and quadrupolar plasmonic modes. The metasurface is simulated on a 50 nm SiN substrate (n = 1.98). A plane wave with 1 V/m amplitude polarized along the x-direction excites the structure. Numerical simulations are performed using CST Microwave Studio with time-domain finite-integration techniques and PML boundary conditions. The SRRs are made of 40 nm gold modeled using Palik dispersive parameters. The optimized geometry (L_1 = 1400 nm, L_2 = 65 nm, L_3 = 220 nm, L_4 = 640 nm, L_5 = 200 nm) demonstrates strong SHG due to enhanced field localization and controlled mode coupling.

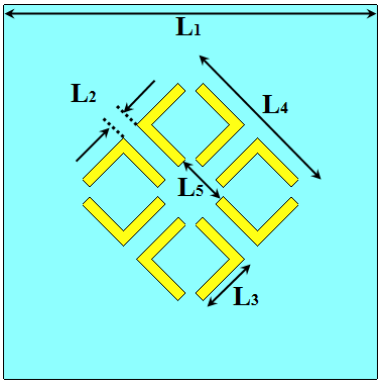


Fig. 1. The nano antenna with the SHG performance for mid infrared application.

III. THE SIMULATION RESULTS AND DISCUSSIONS

The simulation results confirm the presence of second harmonic generation through the extinction cross-section analysis, which combines both absorption and scattering contributions. As shown in Fig. 2, the metasurface exhibits two pronounced resonances at 60 THz and 130 THz, with corresponding extinction cross-section values of approximately 1.4E+6 and 1.8E+6, respectively. The lower-frequency resonance is characterized by a narrow spectral linewidth associated with a dipolar plasmonic mode, while the higher-frequency resonance displays a broader profile, indicative of a quadrupolar mode. The coupling between these modes enables efficient SHG and confirms the nonlinear optical activity of the proposed SRR-based metasurface.

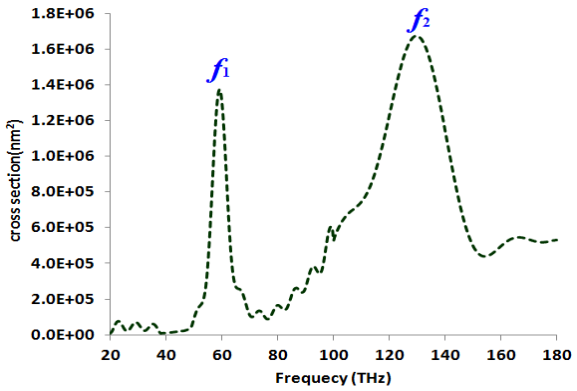


Fig. 2. The extinction cross section of the SHG nano antenna.

Over the past decade, graphene a two-dimensional carbon material arranged in a honeycomb lattice has attracted extensive attention due to its exceptional mechanical, electrical, magnetic, and thermal properties [22]. In nanophotonics, graphene has been widely utilized for tunable and reconfigurable optical devices such as plasmonic nanoantennas and absorbers. Its tunability arises from the ability to modify its chemical potential, which directly influences the extinction spectrum and enables the generation or suppression of Fano resonances [23]. The plasmonic behavior of graphene is governed by its surface conductivity, which can be described using dyadic Green's functions for two-dimensional electron systems [24]. According to the Kubo formalism, the complex optical conductivity of graphene consists of both intraband and interband contributions, as expressed in Eq. (4). The intraband and interband conductivity terms are typically evaluated for single-layer graphene, and their analytical forms are given in Eqs. (5) and (6), respectively [24]. These conductivity components determine the tunable optical response of graphene when electrically or chemically biased, making it an effective platform for dynamic control of plasmonic resonances in metasurfaces.

$$\sigma\ (\omega) = \sigma^{\text{intra}}(\omega) + \sigma^{\text{inter}}(\omega) \tag{4}$$

$$\sigma^{\text{intra}}(\omega) = \frac{e^2 K_B T}{\pi\hbar^2(\omega - i2\Gamma)}\left[\frac{\mu_c}{K_B T} + 2\ln(e^{-\frac{\mu_c}{K_B T}} + 1)\right] \tag{5}$$

$$\sigma^{\text{inter}}(\omega) = \frac{e^2}{4\pi\hbar}\ln\left(\frac{2|\mu_c| - (\omega - i2\Gamma)\hbar}{2|\mu_c| + (\omega - i2\Gamma)\hbar}\right) \tag{6}$$

The k_B is Boltzmann constant, ħ is reduced Plank, T is the temperature in Kelvin, e is electron's charge, τ is relaxation time, ω is the angular frequency, Γ = 1/(2τ) is scattering rate and also μ_c is the chemical potential of graphene. For mid-infrared applications $\sigma^{\text{intra}}(\omega)$ and $\sigma^{\text{inter}}(\omega)$ are considered but in this paper only $\sigma^{\text{intra}}(\omega)$ is used. In this work, graphene is employed as a tunable layer to control the resonance behavior of the metasurface and enhance SHG performance. Specifically, a graphene film is positioned between the gold nanoantenna and the Si_3N_4 substrate. This configuration not only enables dynamic modulation of the resonance frequencies through adjustment of the graphene chemical potential but also helps compensate for fabrication-related imperfections in the gold layer. The optical response of graphene is modeled in CST using the Kubo formalism, which accounts for both intraband and interband conductivity contributions. In this study, a multilayer graphene sheet with an effective thickness of 1 nm is considered at 300 K with a relaxation time of 0.5 ns. To evaluate the tunability of the metasurface, the extinction cross-section is calculated for various chemical potentials in the range of 0.2–0.8 eV. As shown in Fig. 3, increasing the graphene chemical potential results in a noticeable spectral shift of the resonance peaks, confirming the effectiveness of graphene biasing for dynamic tuning of SHG-active plasmonic modes.

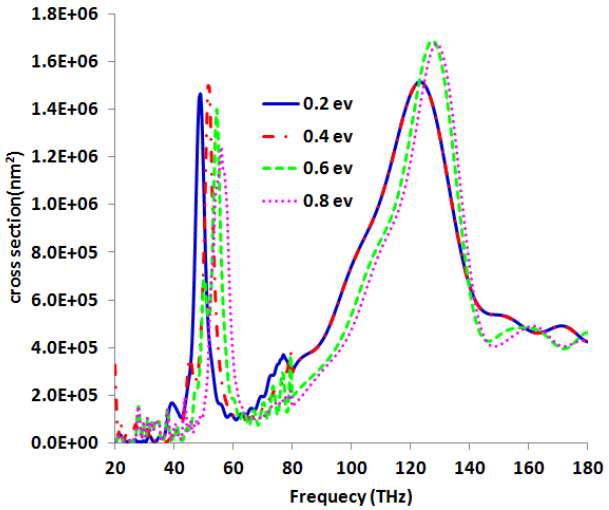
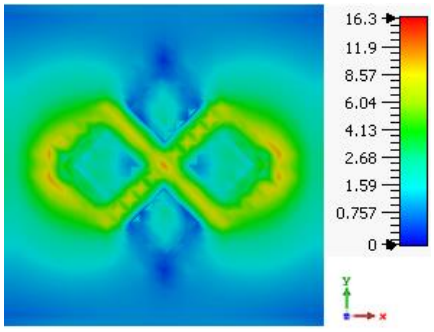
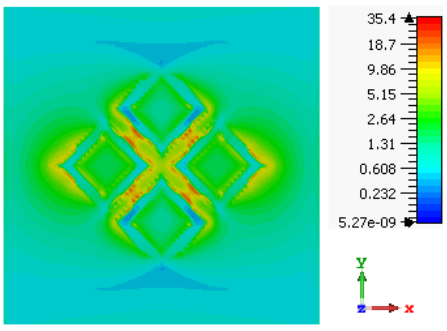


Fig. 3. The extinction cross section of the proposed nano antenna with various Graphene biases.

The electric field distribution of both nano antenna configurations was evaluated under plane-wave excitation with an incident field amplitude of 1 V/m. As shown in Fig. 4, the rectangular nano antenna exhibits a dipolar field distribution at its fundamental resonance, with a maximum electric field intensity of approximately 16.3 V/m (Fig. 4(a)). At the second resonance (130 THz), the field profile transitions to a quadrupolar mode, yielding a peak field intensity of about 35.4 V/m (Fig. 4(b)). These results indicate limited field confinement and weak nonlinear interaction efficiency in the rectangular design, underscoring its inadequacy for supporting strong SHG.



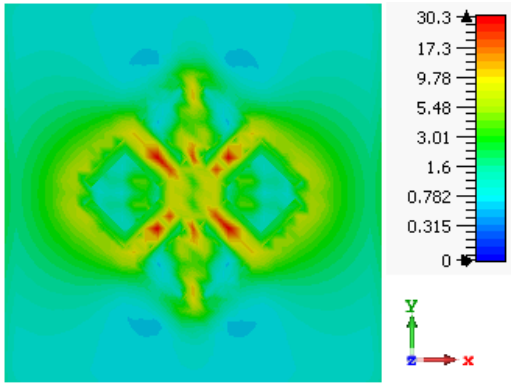
(a)



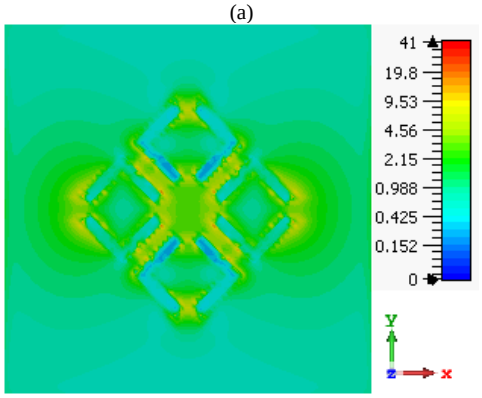
(b)

Fig. 4. The electric field (a) for the rectangular nano antenna at first resonance (b) for the rectangular nano antenna at second resonance.

For the SRR-based nano antenna, the first resonance exhibits a dipolar field distribution with a peak electric field intensity of approximately 32 V/m, as shown in Fig. 5(a). Compared to the rectangular structure, this represents an enhancement of more than 250%, indicating significantly stronger near-field confinement. At the second resonance (130 THz), the SRR structure supports a quadrupolar mode with a maximum field magnitude of about 42 V/m, as illustrated in Fig. 5(b). This corresponds to an energy enhancement of roughly 36% relative to the rectangular configuration. These results confirm that the introduction of split gaps in the SRR geometry effectively strengthens both fundamental and harmonic field localization, thereby facilitating efficient SHG.



(a)



(b)

Fig. 5. The electric field (a) For the SRR nano antenna at first resonance (b) For the SRR nano antenna at second resonance.

IV. Experimental Feasibility

The proposed quad-SRR graphene-integrated metasurface can be fabricated using standard nanofabrication techniques compatible with mid-infrared plasmonic devices. A feasible fabrication workflow is outlined below: 1. Substrate Preparation: A Si_3N_4 film (~50 nm thick) is deposited on a silicon wafer using low-pressure chemical vapor deposition (LPCVD).

2.Graphene Transfer: Single- or few-layer graphene grown by chemical vapor deposition (CVD) is transferred onto the Si_3N_4 substrate using a PMMA-assisted wet transfer technique, followed by annealing to remove polymer residues. 3.Electron-Beam Lithography (EBL):A PMMA resist layer is spin-coated on the graphene and patterned using EBL to define the quad-SRR geometry. 4.Metal Deposition: A 40 nm gold layer is deposited using electron-beam evaporation. A thin chromium or titanium layer (1–2 nm) may be used as an adhesion layer. 5.Lift-Off Process: The sample is immersed in acetone to remove the resist, leaving the gold SRR structures on top of the graphene–Si_3N_4 substrate. The performance of the proposed quad-SRR metasurface can be influenced by several practical factors, including thermal effects, intrinsic material losses, and fabrication tolerances. These considerations are important in assessing the robustness and real-world feasibility of the design.

1. Thermal Effects

Mid-infrared plasmonic nanoantennas generally experience lower absorption-induced heating compared to visible-frequency structures. Under the simulation excitation level (1 V/m), the absorbed optical power is insufficient to cause significant local temperature rises. Thermal-induced resonance shifts are therefore expected to remain below 1%, which is smaller than the tunability achievable through the graphene chemical potential. In practical operation, moderate thermal fluctuations can be further compensated through electrical tuning of the graphene layer.

2. Material Losses

Gold losses, modeled using Palik's dispersive parameters, primarily affect the linewidth of the quadrupolar resonance. Although material absorption reduces the Q-factor slightly, it does not suppress the field localization required for SHG. Graphene introduces additional intraband absorption, which is accounted for using the full Kubo conductivity model in our simulations. The combined loss mechanisms mainly broaden the resonance peaks, but the nonlinear frequency-doubling mechanism remains intact due to strong dipole–quadrupole coupling in the SRR geometry.

3. Fabrication Imperfections

Typical nanofabrication variations—such as deviations in gap width (±10 nm), surface roughness, and slight asymmetries—may shift the resonance frequencies by approximately 2–3 THz. These variations do not fundamentally disrupt SHG, as the nonlinear response is primarily governed by field localization within the engineered gaps. Importantly, the incorporation of graphene provides a practical method to compensate for fabrication-induced spectral shifts, enabling post-fabrication tuning via control of the chemical potential. This feature increases the robustness and reproducibility of the proposed metasurface.

CONCLUSION

In this study, a plasmonic nano antenna based on a quad SRR configuration was designed and analyzed to achieve efficient SHG in the mid-infrared regime. The introduction of engineered split gaps was shown to play a crucial role in controlling the local electric field distribution, thereby influencing both the near-field enhancement and the corresponding far-field radiation patterns. The metasurface supports two resonant modes at 60 THz and 130 THz; however, the presence of dual resonances alone is not sufficient to ensure SHG. Instead, the SHG capability is directly linked to the enhanced field localization and the coupling between dipolar and quadrupolar modes. The symmetric arrangement of the SRR elements enables polarization-independent performance, while the integration of a graphene layer beneath the nanoantenna provides dynamic tunability of the resonance frequencies through adjustment of its chemical potential. Furthermore, the far-field response of the optimized structure exhibits a quadrupolar radiation pattern at the second harmonic frequency, which is absent in the non-SHG rectangular configuration. These results demonstrate that the proposed SRR metasurface offers an effective platform for tunable nonlinear photonics, mid-infrared sensing, and reconfigurable optoelectronic applications.

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