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STUDY ON THE INTEGRATION OF TWO-DIMENSIONAL PLASMONIC MATERIALS, GRAPHENE AND MoS_2, INTO METAMATERIAL ABSORBER STRUCTURES OPERATING IN THE GHz AND THz FREQUENCY RANGES

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INTRODUCTION

1. Research Urgency

To date, research on metamaterials (MM) with artificial structures having physical dimensions many times smaller than their operating wavelength has been a prioritized field of development in both theory and practice. During the development of MM, there have been many difficulties and challenges hindering their practical applications, such as expanding/actively controlling the frequency band, realizing three-dimensional structures, integrating with current electronic devices, or miniaturizing structures at micro-nanometer scales. We have witnessed many major improvements, notably the concept of combining MM with their advantages in artificial structures and potential plasmonic materials through the interaction between light and free electrons in noble metals. The interesting properties of surface plasmon polaritons (SPPs, propagating at the interface between dielectrics and metals) have been utilized in many MM structures to create miraculous effects not observed in natural materials, including: negative refractive index, reverse Cherenkov/Doppler effects. In general, MM have discrete/periodic structures and their operating mechanism is based on strong light-matter interactions with internal electromagnetic waves and artificial sub-wavelength scale unit cells. Therefore, when combined with plasmonic materials (especially two-dimensional materials - 2DP), they will bring many advanced technological solutions (MM-2DP) helping to solve the mentioned challenges.

Currently, some advanced properties of 2DP materials (transmittance, good electrical conductivity, and conductivity values that can be easily controlled through chemical potential, electric/magnetic fields) have mostly been studied based on theoretical calculations or simulations at THz frequencies for applications in adjusting amplitude and absorption frequency bands. The fabrication and application of these structures still face many challenges due to high costs, as they typically use expensive modern manufacturing technologies. On the other hand, telecommunications technology and communications in smart devices operating in the GHz range (4G/5G) for military, civilian, and healthcare purposes are currently priority development areas in our country and many countries worldwide. Therefore, the early realization of electromagnetic wave absorption material models based on metamaterials integrated with two-dimensional plasmonic materials (2DP-MPA) in the GHz range with simple structures, large sizes, and reasonable costs is being actively pursued by research groups abroad.

Some research directions that need to be implemented to bring 2DP-MPA models into practical operation in the GHz range include developing diverse manufacturing technologies for MA materials while simultaneously developing technologies for integrating 2DP materials into MPA structures to: control changes in amplitude and frequency of absorption spectra through the thickness of used 2DP materials; eliminate the need for masks to save costs and manufacturing time; control absorption properties when completely replacing the metal layer with periodic 2DP; realize two-dimensional absorption based on isotropic structures; control absorption amplitude and frequency based on forced effects (mechanical-thermal, electrical, optical, and doping); control polarization conversion effects in asymmetric models. Therefore, we have chosen the thesis topic: "***Study on the Integration of Two-Dimensional Plasmonic Materials, Graphene and MoS_2, into Metamaterial Absorber Structures Operating in the GHz and THz Frequency Ranges***" to optimize single/multi-band and broadband absorption characteristics.

2. Research Objectives

- Clarify the operating mechanism of some metamaterial models integrated with two-dimensional plasmonic materials (MPA-2DP), based on electromagnetic resonance characteristics and impedance matching, dielectric resonance.

- Fabricate and experimentally verify some MPA-2DP models in the GHz frequency range to control multi-peak absorption amplitude and frequency.

3. New Contributions

The dissertation has achieved several main results related to the integration of two-dimensional plasmonic materials (graphene and Molybdenum-disulfide), including:

+ The dissertation clarifies the operating mechanism of the MPA-2DP model integrating low-conductivity ink on a 2D graphene substrate (resistance varying from 7.2 to 30.0 Ω/sq in 5-10 GHz range), obtaining multi-peak absorption spectrum (maximum absorption over 90% at 6.85 GHz), absorption can be controlled up to 60% at 8.4 GHz. Due to the symmetric structure, this model works well with incident angles less than

60° and at different polarization angles (0-90°).

+ The dissertation has controlled changes in amplitude and absorption frequency of some MPA-2DP over a wide frequency band based on MoS_2 integration (absorption over 90% from 10.1-17.8 GHz), relative bandwidth (FBW) reaching over 55.2%.

CHAPTER 1: OVERVIEW OF ELECTROMAGNETIC-WAVE ABSORPTION CHARACTERISTICS OF METAMATERIALS INTEGRATED WITH TWO-DIMENSIONAL PLASMONIC MATERIALS

1.1. Absorption Characteristics in Metamaterial (MM) Environment

1.1.1. General theory of perfect electromagnetic wave energy absorption in MM

The electromagnetic interaction between materials and electromagnetic waves can be described through Maxwell's equations with appropriate boundary conditions. Reflection and transmission coefficients can be determined by the following equations:

$$\tilde{r} = \frac{E_r}{E_i} \equiv |r|e^{i\theta_r}$$

$$\tilde{t} = \frac{E_t}{E_i} \equiv |t|e^{i\theta_t}$$

where θ_r and θ_t are reflection and transmission phases respectively. Reflectance (R) and transmittance (T) are determined by:

$$R = |\tilde{r}|^2$$

$$T = |\tilde{t}|^2$$

Absorption (A) is calculated according to energy conservation law:

$$A = 1 - R - T$$

1.1.2. Perfect impedance matching in MPA-2DP

The impedance matching is a necessary condition for perfect absorption, where $\varepsilon_r = 1$ and $\mu_r = 1$ or $Z_r = 1$. In the general case:

$$R = |\tilde{r}|^2 = 0$$

$$T = |\tilde{t}|^2 = e^{-2n_2k_0d}$$

$$A = 1 - e^{-2n_2k_0d}$$

The above equation indicates that if the medium has a large loss element (n_2) and optimized thickness *d*, therefore, perfect absorption (100%) can be achieved inside the sub-wavelength scale.

1.1.3. Effective theoretical model for single-layer anisotropic MPA structure

Consider a traditional MPA structure as a homogeneous material consisting of 3 layers: periodic metallic structure/dielectric/continuous metal layer [59]. We can examine the selective interaction of electromagnetic waves when transmitted to the front structure, typically caused by magnetic and electric resonance. At the absorption wavelength $\lambda = 5.94$ µm, $\varepsilon_{eff} = 2.432 + 24.175i$ and $\mu_{eff} = 1.003 + 25.517i$ lead to $z_{eff} = 1.02 + 0.03i$, and $n_{eff} = 1.736 + 24.849i$. The large imaginary part of the refractive index n" = 24.849 is the original mechanism of absorption-energy dissipation at this wavelength. The electromagnetic wave intensity when penetrating through the MPA sample surface can be represented by: $I = I_0e^{-2\alpha z}$, where $\alpha = n''k = 2\pi n/\lambda = 26,28$ µm^{-1}. When electromagnetic waves are reflected back due to the continuous metal back layer, the intensity decreases according to $I/I_0 = e^{-4\alpha ts} = 7,8x10^{-5}$. Therefore, 99,98% of the incident electromagnetic wave energy is absorbed inside the MPA.

1.1.4. Anisotropic MPA structure model based on electromagnetically induced transparency effect

To create multiple resonances, we can exploit the interaction between resonant structures in metamaterials based on electromagnetically induced transparency (EIT) effect. A typical model of this method is designed including two layers: a resonant structure layer consisting of 3 metal bars on top and a dielectric layer below. The vertical metal bars act as bright modes directly excited by the incident field while the two horizontal metal bars cannot be directly excited and are considered dark modes. The dark mode can be excited by the bright mode through the displacement of the vertical metal bar position (indirect excitation). The direct excitation path $|0\rangle \rightarrow |1\rangle \rightarrow |0$ and indirect excitation path $|0\rangle \rightarrow |1\rangle \rightarrow |2\rangle \rightarrow |0\rangle$ will destructively interfere to create a transmission window. This phenomenon is commonly known as the EIT effect. In the case of symmetric structure, the spectrum only shows one transmission dip corresponding to the resonant excitation of the bright mode. When the structure is asymmetric, a transmission region appears at the position of the initial transmission dip and lies between two newly created resonances. The splitting of 2 transmission dips due to near-field (NF) interaction leads to the formation of two new resonances. At these two new resonance frequencies, impedance matching of the structure with the operating environment can occur, when combined with strong magnetic resonance, the incident electromagnetic wave energy will be completely absorbed inside the structure.

1.1.5. Absorption characteristics of isotropic MPA structure

MM materials designed with a continuous metal layer at the back are generally called anisotropic structures due to their one-dimensional absorption. Unlike anisotropic MPA structures, isotropic structures need to control two resonances: electric resonance and magnetic resonance close together. Although isotropic MPAs have advantages in independent practical operation, they face difficulties in fabrication techniques: requiring absolute coaxial alignment between two metal structure layers of the dielectric plate.

1.2. Electromagnetic properties of two-dimensional materials with periodic structure

1.2.1. Kubo theoretical model for graphene's electrical conductivity

The graphene layer is characterized by surface conductivity value, determined through surface current appearing when the electric field acts tangentially to the graphene plane. Using the Kubo conductivity model, graphene's conductivity can be determined. In the non-magnetic state, conductivity is considered isotropic with two conductivity components: intraband (σ_{intra}) and interband (σ_{inter}) contributing mainly. At room temperature (300K), the contribution of inter-band conductivity is determined to be very small compared to the intra-band conductivity component. Therefore, we can describe the surface conductivity of the graphene layer according to Drude theory, where graphene's surface conductivity depends on frequency and chemical potential (Fermi energy).

1.2.2. Theoretical model for electrical conductivity characteristics of molybdenum disulfide (MoS_2)

MoS_2 in nanometer-scale film form (MoS_2-NS) is a multilayer material similar to graphene. At high frequency region, the real part value of dielectric permittivity in both bulk and nanofilm cases decreases according to Debye theory:

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2}$$

$$\varepsilon'' = \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2}\omega t + \frac{\sigma}{\omega\varepsilon_0}$$

1.3. Some MPA models integrated with two-dimensional plasmonic materials (MPA-2DP)

1.3.1. MPA structure partially integrated with Graphene

The unit cell design of the MPA-graphene structure includes four layers: metal (gold) resonators with thickness of $t_m = 0,1$ μm [25,26] and a metal plane separated by a dielectric layer with relative permittivity $\varepsilon_d = 1,6$ and integrated with a graphene layer of thickness $t_g = 0,34$ nm directly below the periodic metal structure on the front. Research results show that absorption amplitude and frequency are strongly affected by the resonance ring radius. This research group constructed super unit cells by arranging resonance rings of different radii and arranging coaxial resonance rings. Results show that when the unit cell structure is optimized, the absorption spectrum bandwidth has been significantly expanded.

1.3.2. MPA structure based on front resonance entirely made of 2DP materials

MPA with resonant structures on the front made entirely of graphene have been studied. Recent results show that absorption and absorption spectrum bandwidth significantly depend on graphene's conductivity. When graphene's conductivity is low, the absorption spectrum is narrow-band; when conductivity increases, narrow absorption peaks broaden and form broadband MPA.

1.4. Application potential of MM-2DP

*** THz radiation impact reduction applications**

Currently, there aren't many comprehensive studies on THz waves' effects on human health. Therefore, research on electromagnetic interference, information security, and military stealth of materials in the GHz-THz range is still being intensified. 2DP materials integrated into MMs structures have advantages in flexible control of absorption amplitude and frequency in multilayer structures, hybrid structures, or ultra-thin-light structures. Some MMs models integrating 2DP materials for electromagnetic wave shielding in the THz region can achieve 90% absorption in the operating frequency range from 0.1 THz to 2.5 THz.

*** THz modulation applications**

Graphene's electron density and Fermi energy level can be controlled through external applied voltage. Therefore, graphene-integrated MPA have good modulation capability, showing potential for expanding MPA-2DP applications with multi-peak/broadband applications through multilayer structure design, selective and electromagnetic wave energy storage in GHz region.

* **THz sensing applications**

In 2019, Qiu *et al.* proposed an MPA structure model integrating MoS_2 material in the visible light region. For sensing applications, this model can achieve sensitivities of 500 nm/RIU and 200 nm/RIU corresponding to square and circular structures. The operating mechanism is based on dielectric resonance and surface plasmon resonance of 2DP material structures integrated periodically inside MPA.

CONCLUSION

In this chapter, the dissertation has presented an overview of research on electromagnetic wave absorption characteristics of some metamaterial models integrating two-dimensional plasmonic materials; analyzed and clarified elements affecting the operating mechanism of MPA-2DP hybrid models based on perfect impedance matching theory or electromagnetically induced transparency effect. Some notable applications of the models according to the dissertation's research direction include: electromagnetic shielding and sensing. However, studying integrated models may face some challenges such as: applying 2DP materials' plasmonic characteristics in simulation and experiment to control absorption spectrum amplitude and frequency; optimizing MPA-2DP model based on active control (mechanical-thermal, electrical, optical and doping); Evaluating the contribution of 2DP material layer's plasmonic characteristics to the absorption energy dissipation mechanism of MPA structures, etc.

CHAPTER 2: DESIGN METHODS, SIMULATION AND EXPERIMENTAL RESEARCH OF MPA-2DP

2.1. Theoretical calculation model for MPA-2DP

In the finite element theory calculations, to evaluate sample absorption, we need to determine scattering parameters through the relationship between incident waves and reflected and transmitted waves. Through the parameter characterizing reflection S_{11}, we can evaluate the position of impedance matching, and through S_{21} can evaluate the loss level of the environment. The absorption frequency of the structure can be predicted based on the equivalent RLC resonant circuit of that structure, depending on the unit cell structure design.

2.2. MPA-2DP Simulation Techniques

In simulation, the absorption mechanism and resonance properties of MPAs can be investigated based on commercial software (CST, COMSOL, ...) [93,94] licensed at the Digital Laboratory of the Graduate University of Science and Technology (GUST). These softwares allow obtaining electric field distribution, magnetic field, induced current and energy loss, etc. The explicit values of εeff and μeff parameters (or effective refractive index) can be determined through calculation techniques for isotropic MM environment (when using reflection and transmission coefficients).

2.3. Some MPA manufacturing techniques and 2DP material integration

2.3.1. MPA partially integrated with 2DP material

The fabricated sample can be included by 3 layers: metal/dielectric/metal, or 2 layers: metal/dielectric, depending on whether the two-dimensional plasmonic material is partially or fully integrated. The 2DP materials used to integrate into MPA samples will be: graphene, MoS_2. Initially, models optimized in simulation and calculation will be manufactured by conventional photolithography, then 2DP material will be integrated into the manufactured samples.

2.3.2. MPA with resonant structure using 2DP material

In the case of completely replacing the periodic metal layer with two-dimensional conductive ink, nano-graphene ink shot through a mask (with grooves being structures etched by laser) is a highly effective method with low cost.

Direct printing method is an effective tool helping to manufacture or integrate many different types of materials on MPA structure. The structure of the material will be created by conductive ink doped with metal components such as silver nanoparticles or graphene, MoS_2 and WS_2. This is a new simple, quick method and can integrate many types of 2DPs with large size without using masks. Therefore, this technology is completely suitable for manufacturing 2DP-MPA models in GHz frequency range.

2.4. Measurement and Experimental Investigation of Electromagnetic Properties of MPA-2DP

In the measurement process at GHz frequency range, in the anechoic chamber, we use the Hewlett-Packard ZNB20 system. In the free-space measurement method, two horn antennas with operating frequency range from 1.0 to 18 GHz are set up so that the incident angle of electromagnetic waves to the surface of MPA-2DP can be changed. Currently, this measurement process will be performed at the Institute of Materials Science (VAST) or at the Military Institute of Science and Technology - Vietnam.

CONCLUSION

To study the electromagnetic properties of MM materials integrated with two-dimensional plasmonic materials, theoretical calculation and simulation methods are first used to design MM with desired electromagnetic characteristics. The process of designing and simulating MM structures in GHz and THz regions is based on finite element technique (CST Studio and Comsol Multiphysics) or FDTD technique (Lumerical FDTD). Based on initial simulation results, we can evaluate the feasibility of using or integrating two-dimensional materials into MM structures operating in GHz or THz frequency ranges. After proposing and optimizing the structure and manufacturing feasibility, different structures to verify desired electromagnetic characteristics are fabricated based on photolithography technique. Finally, basic physical properties of materials are measured through devices such as Fourier transform infrared spectroscopy (FTIR), Vector Network Analyzer (VNA ZNB20), from which we will understand the operating mechanism of proposed integrated models.

CHAPTER 3: RESEARCH ON ABSORPTION CHARACTERISTICS OF METAMATERIALS INTEGRATED WITH PLASMONIC GRAPHENE

3.1. Investigation of graphene's influence on electromagnetic properties of metamaterial absorbers with direct near-field interaction effect (D-MPA)

3.1.1. Design - simulation of D-MPA

The unit cell of the proposed D-MPA is shown in Figure 3.1. This structure includes an FR-4 dielectric layer between two copper metal layers. The structural parameters have values presented in Table 3.1.

Table 3.1. Structural parameters of MPA

Parameter	a	d	g	l	r_1	r_2	s	t_d	t_m	w
Value (mm)	*22*	*1*	*1.5*	*11*	*7*	*1*	*0.5*	*1.6*	*0.035*	*2*

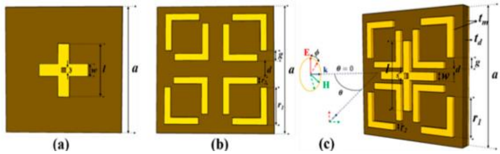


Figure 3.1. *Unit structure of proposed MPA: (a) CS structure, (b) SRR structure and (c) multi-resonant combined MPA structure.*

CS structure absorption can reach 98.4% at 6.85 GHz (Figure 3.2a), while SRR structure reaches 82.2% at 8.5 GHz (Figure 3.2b). By combining them together, MPA can create three absorption peaks at 6.85 GHz (92.2%) and two close absorption peaks at 8.37 GHz (98%) and 8.65 GHz (99.8%) (Figure 3.2d). Results show that exploiting EIT effect's NF interaction allows achieving multi-band absorption spectrum of proposed D-MPA operating based on "bright-bright" NF interaction. S-parameters S_{11}, S_{21}, S_{22} and S_{12} (Figure 3.2c) show proposed MPA only absorbs waves incident on the front structure layer and reflects all waves incident on a continuous metal layer. EIT effect due to NF interaction of bright-bright elements has caused D-MPA to create three absorption resonances at frequencies 6.85, 8.37 and 8.65 GHz. In other words, NF interaction plays important role in increasing number of resonances, thereby creating multi-peak absorption.

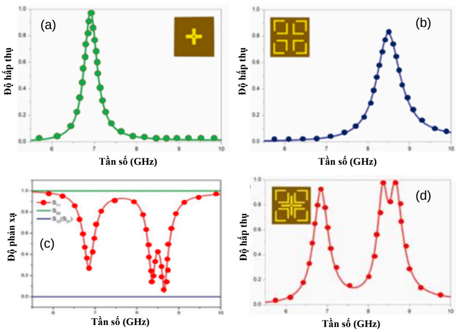


Figure 3.2. *Absorption spectra of (a) CS structure and (b) SRR structure. (c) S-parameters S_{11}, S_{22} và S_{12} (S_{21}), and (d) absorption spectrum of the proposed D-MPA.*

3.1.2. Investigation of Absorption Spectrum Variation According to Structural Parameters of D-MPA Structure

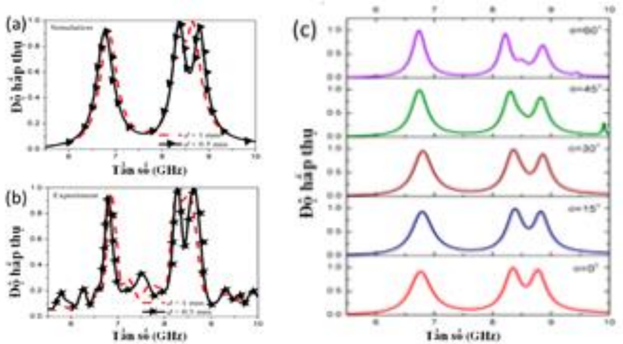


Figure 3.3. *(a) Simulated and (b) experimental absorption spectra dependent on distance d between SRR and CS structures and (c) incident angle.*

The absorption spectrum changes according to distance d between CS and SRR structures (characterizing the strength of NF interaction between resonances) of MPA have been studied (Figure 3.3). When *d* decreases from 1 to 0.5 mm, a small shift occurs at the first absorption resonance position (from 6.85 to 6.80 GHz) and the two high-frequency absorption peaks change more significantly. This result shows that parameter *d* plays an important role in controlling the NF interaction of the two resonant structures. When *d* is smaller, the NF interaction of SRR and CS becomes stronger, making the resonance peaks more distinct. It can be concluded that by controlling the structural parameter *d*, the resonance (absorption) position is flexibly controlled. Therefore, the absorption region from 8.28 to 8.75 GHz can be converted from broadband (at larger *d*) to dual-band absorption region (at smaller *d*).

Figure 3.6 shows simulated and experimental absorption spectra of the proposed MPA before and after graphene integration. When the surface resistance of graphene ink is 7.2 Ω/sq., the first absorption peak remains almost unchanged as this is the individual absorption resonance of the CS

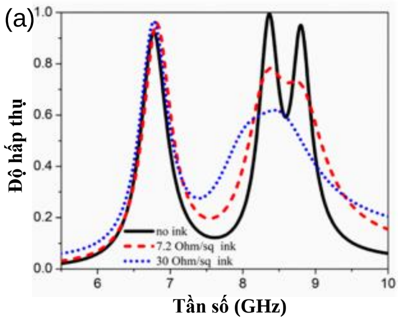


Figure 3.6. *(a) Simulated absorption spectra of initial plasmonic MPAs and after graphene integration with different resistances of graphene ink*

structure, thus not affected by the interaction of the graphene material layer. However, the distance between the second and third absorption peak positions is narrowed, the frequency range from 8.26 to 8.86 GHz shows absorption only reaching 70% to 80%. When the sheet resistance of graphene ink increases from 7.2 to 30 Ω/sq., first absorption peak remains almost unchanged. Meanwhile, the second and third absorption peaks transform into a single absorption peak at 8.4 GHz (60%).

The simulation results of surface current distribution indicate that the NF interaction of "bright-bright" resonant structures is weakened with the presence of the graphene material layer.

3.1.3. Fabrication and study of absorption characteristics of D-MPA integrated with plasmonic Graphene material

In fabricated sample, first, periodic metal structures on traditional FR-4/Polyimide substrate with 0.6-1.2 mm thickness (ε_r = 2,5-4.3, dielectric loss tanδ ~ 0,02) can be fabricated using photolithography technique. A layer of graphene ink (Resistivity: 0.003-0.005 Ω-cm) is coated on the surface of metal structures through direct inkjet printing method. The measured absorption spectra of D-MPA-2DP structure show similar trends to simulation results. Without graphene ink, absorption reaches above 90% at 3 peaks: 6.8 GHz, 8.4 and 8.8 GHz. When the resistance of graphene ink increases to 7.2 and 30 Ohm/sq., experimental spectra also show amplitude degradation (lowest absorption below 60%) at two peaks 8.4 and 8.8 GHz.

In the hybrid structure between D-MPA integrated with two-dimensional plasmonic, the graphene layer acts as a resistive layer weakening the individual resonance amplitude of SRR. Therefore, the near-field interaction of SRR and CS resonant structures is also weakened, causing the frequency separation between the second and third peaks to decrease. The obtained results indicate that the absorption characteristics of D-MPA can be flexibly controlled by integrating with low-conductivity material (similar to two-dimensional plasmonic material - graphene) in the desired operating frequency range.

3.2. Investigation of graphene's influence on electromagnetic properties of metamaterial absorbers with indirect near-field interaction effect (I-MPA)

3.2.1. Study of electromagnetic characteristics of metamaterial structure with indirect NF effect

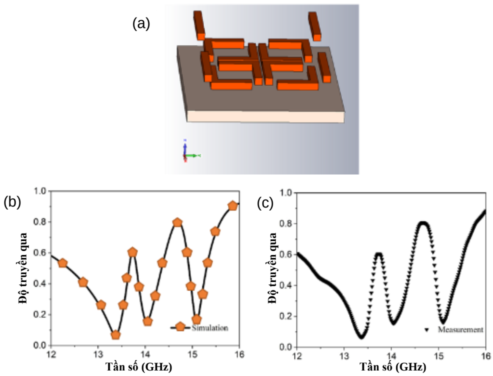
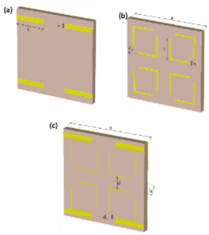


Figure 3.11. *(a) Optimized multi-peak EIT structure in GHz frequency range and (b) simulated and (b) experimental transmission spectra correspondingly.*

The I-MPA structure is designed including asymmetric split-ring resonator (SRR) and cut wire (CW) [Figure 3.11(a)]. Simulation results show two transmission maxima of 60% and 80% appearing at 13.7 and 14.7 GHz while transmission minima are at 13.3, 14 and 15 GHz respectively [Figure 3.11(b)]. When distance between CW and SRR is less than 0.5 mm, spacing between transmission minima positions is predicted to be most separated due to strongest NF interaction [95-97]. At *d* values larger than 0.5 mm, SRR and CW are considered symmetric so excitations due to NF interaction between SRR and CW are not clear, in other words can only create 1 dip in absorption spectrum (due to electric dipole induced on SRR). Additionally, when *d*=0.5 mm, position of SRR and CW becomes asymmetric and both "bright-dark" and "bright-bright" NF interactions between SRR and CW are created. Therefore, multi-band EIT effect appears due to these interactions.

3.2.2. Optimization of I-MPA Structure Model



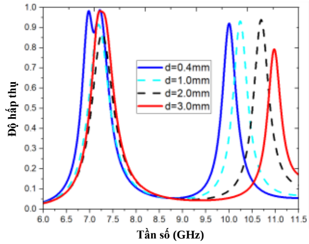


Figure 3.12. *Schematic diagram of 3-layer MA(Metal/dielectric/metal) structure: a) CW cut strip; b) SRR resonance ring; c) I-MPA structure.*

Figure 3.15. *Absorption spectra in GHz frequency range of MPA structures when reducing the distance value d from 3 to 0.4mm.*

To optimize and obtain absorption peaks with 90% absorption, we proceeded to design an additional continuous metal plate behind the CW-SRR structure (this structure is called I-MPA). When reducing the distance *d* between SRRs, there is a strong change at the absorption peaks of the structure. At lower frequencies, two absorption peaks appear at 6.97 (98.27%) and 7.22 GHz (98.75%) with $d < 0.4$ mm. The third absorption peak at 10 GHz increases from 79.25 to 92%. Observations indicate that parameter *d* plays an important role in controlling bright-dark and bright-bright NF interactions. When d is smaller, the near-field interaction of SRR becomes stronger, causing the resonance peak positions at lower frequencies to separate further. The absorption peak in the higher frequency region is due to CW resonance induced by the "bright-dark" NF interaction of SRR and CW. As the relative position of SRR and CW changes, the induced current distribution on CW also changes, thereby shifting the absorption frequency of this absorption peak.

3.2.3. Design and simulation of I-MPA structure integrated with plasmonic Graphene

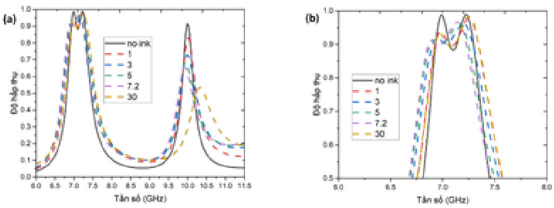


Figure 3.17. *(a) Absorption spectra of the structure when changing graphene ink resistance from 1 to 30 Ω/sq. and (b) observation of changes in two absorption peaks at low frequency.*

To expand the absorption spectrum control capability, we integrated low-conductivity graphene material into the I-MPA structure. Graphene ink was coated on CW and SRR. Different types of graphene ink with surface resistance varying from 1 to 7.2 Ω/sq were investigated with *d* = 0.4 mm. The absorption of two peaks at lower frequencies gradually decreases and tends to merge into one peak. The individual absorption peak at higher frequency (10 GHz) decreases from 92% to 62% when increasing graphene ink resistance value from 1 to 7.2 Ohm/sq. When graphene conductive ink resistance value increases to 30 Ohm/sq., the absorption spectrum still maintains 3 peaks around 6.9 GHz (90%), at 7.35 GHz (96%), and at 10 GHz (55%). This result confirms that the presence of 2D plasmonic material like graphene strongly affects the absorption peaks caused by NF interaction (at 6.9 GHz and 10 GHz), absorption amplitude decreases due to induced currents being cancelled on the surface of CW metal strip structures (Ohmic loss component is enhanced when surface resistance increases from 1 to 30 Ω/sq.). Conversely, the fundamental absorption frequency at 7.35 GHz is maintained due to the existence of magnetic resonance and inherent dielectric loss inside the MPA structure.

3.3. Control of absorption characteristics of MPA material integrated with plasmonic Graphene

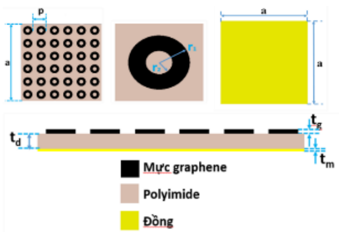


Figure 3.20. *Illustration of the MPA-2DP structure operating in the GHz region.*

Figure 3.20 describes the structure of MPA-2DP material using graphene ink layer. The material has a structure consisting of continuous copper metal layer at the back, flexible polyimide dielectric layer in the middle, and graphene ink layer with surface resistance of 7.2 Ω/sq created periodic resonant structures on the front. Simulation results show that the material has absorption reaching 99.99% at frequency 5.26 GHz in flat state. When bent, the simulated absorption spectrum of the material slightly decreases in amplitude and frequency undergoes blue shift as bending radius increases gradually. Specifically, when bent with bending radius R = 1000 mm, absorption reaches 98.8% at 5.26 GHz. Under conditions when R = 500 mm, 100 mm, and 50 mm, corresponding absorption reaches 98.99% (5.25 GHz), 99.88% (5.21 GHz) and 99.67% - 5.1 GHz (Figure 3.26). Thus, with the proposed material sample, the absorption spectrum does not depend much on the elastic properties of the material, it still maintains good absorption characteristics even when bent with bending radius R = 50 mm.

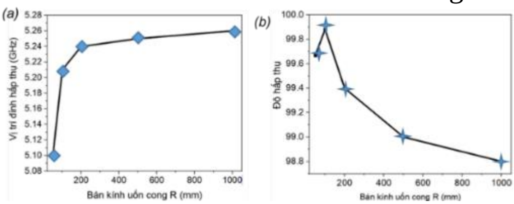


Figure 3.26. *Dependence of (a) position and (b) absorption peak intensity on curvature.*

**** Voltage-controllable MPA-2DP model for frequency and absorption***

To control frequency and absorption using voltage, a graphene layer is placed between the SiO_2 layer and the circular resonance structure (Figure 3.28). As the chemical potential increases, the absorption intensity of the MPA decreases while the position of the absorption peak shifts. When the chemical potential increases from 0 to 0.3 eV, the absorption peak shifts toward lower frequencies. If μ_c continues to increase, the absorption peak shifts toward higher frequencies. The changes in absorption amplitude and frequency can be explained by variations in induced charge density generated by different applied voltages. Comparing the energy dissipation ratio across material layers shows that the metal layer dominates due to Ohmic losses in the high-frequency region.

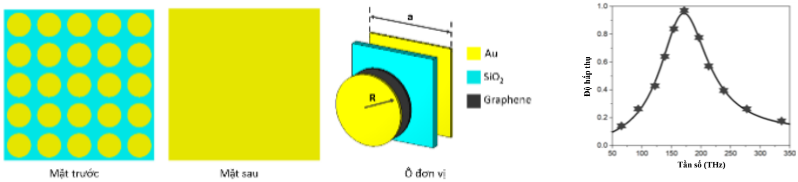


Figure 3.28. *Metamaterial structure integrated with graphene and simulated absorption spectrum for the corresponding MPA-2DP structure.*

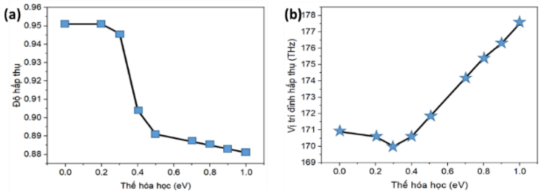


Figure 3.30. *Changes in (a) absorption intensity and (b) absorption peak position according to the chemical potential value of the graphene sheet.*

The decrease in absorption amplitude with increasing chemical potential up to 1.0 eV can be explained based on the position and intensity of graphene layer plasmon resonances that may shift to different frequency regions. Another factor could be the transition between interband and intraband regions of the graphene layer.

CONCLUSION

This chapter presents several key findings on multi-band absorption characteristics based on structures with direct (D-MPA) and indirect (I-MPA) NF interaction effects. Through the integration of two-dimensional plasmonic material (graphene), near-field interactions affecting the resonance of bright and dark mode elements lead to changes in absorption amplitude and frequency in the GHz frequency range. Specifically:

- D-MPA structure: strong NF interaction at d = 0.5mm distance between SRR and CS resonances, NF interaction decreases when integrating a partial graphene layer with surface resistance increasing from 7.2 Ω/sq. to

30 Ω/sq. Consequently, absorption maintains above 90% at 6.8 GHz and decreases below 60% at 8.2 GHz and 8.8 GHz.

- I-MPA structure: strong NF interaction at *d* = 0.4mm distance between SRR and CW resonances, absorption peaks due to NF interaction are strongly affected around 6.9 and 10.0 GHz, absorption maintains above 90% at 7.35 GHz when the surface resistance of graphene layer increases to 30 Ω/sq.

- The plasmonic characteristics of graphene affecting fundamental/higher-ordered resonances have been investigated in several deformed (curved state) structural models in the GHz frequency range and external voltage control model in the THz frequency range: in the GHz frequency range, the presence of graphene material creates Ohmic losses that play a primary role in maintaining high absorption peaks above 90% for curved deformation cases. In the THz frequency range, the plasmonic characteristics of the graphene layer controlled through chemical potential (from 0.2 to 1.0 eV) have affected the amplitude (absorption decreases from 95.1% to 88.1%) and absorption frequency (from 170.6 to 177.5 THz).

CHAPTER 4: INVESTIGATION OF ABSORPTION CHARACTERISTICS IN SOME METAMATERIAL MODELS INTEGRATED WITH MoS_2 MATERIAL

4.1. Research on absorption characteristics of MPA-2DP integrated with MoS_2 in GHz frequency range.

4.1.1. Design of MPA-2DP unit cell structure integrated with MoS_2 in GHz frequency range.

The MM is designed with a three-layer structure: continuous copper metal layer at the back, flexible dielectric layer made from polyimide in the middle, and MoS_2 ink layer consisting of simple square structures (Figure 4.1).

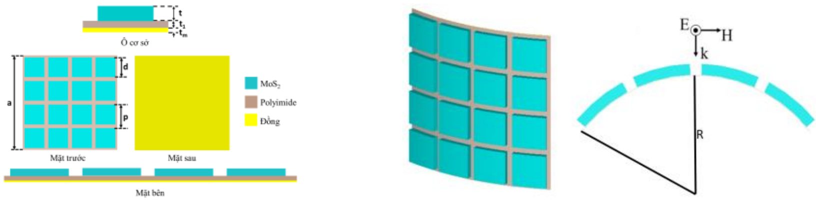


Figure 4.1. *Illustration of the unit cell structure and elastic state of MPA-2DP.*

4.1.2. Simulation of electromagnetic characteristics of MPA-2DP integrated with MoS_2 in flat State

Simulation results show that the material exhibits good absorption properties with absorption above 90% in the frequency range from 10.1 GHz to 17.6 GHz (Figure 4.3(a)). The ratio of energy dissipation in the MoS_2 layer and polyimide dielectric substrate to total energy absorbed by the material has been simulated showing that nearly 100% of absorption energy is dissipated on the MoS_2 ink layer, while energy dissipation on the dielectric layer accounts for less than 0.05%.

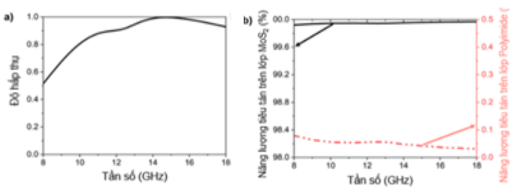


Figure 4.3. *(a) The absorption spectrum of the proposed MPA-2DP, (b) energy dissipation ratio in the material's structural layers.*

4.1.3. Analysis of Absorption Mechanism of MPA-2DP Integrated with MoS_2

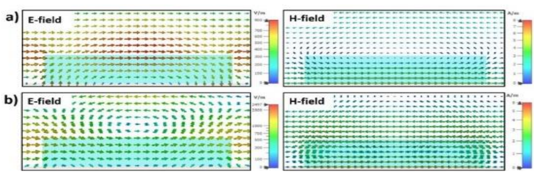


Figure 4.4. *Electric and magnetic field distributions at frequencies: a) 11.7 GHz and b) 15.3 GHz.*

Simulated results of electric- and magnetic-field distribution show that, at 11.7 GHz, a ring-shaped electric field is formed, creating a magnetic dipole concentrated in the MoS_2 block. Conversely, at 15.3 GHz, an electric dipole in the MoS_2 block can be observed formed due to ring-shaped magnetic field in the cube. From this point, we can conclude that the structure's wide bandwidth is achieved due to the combination of two Mie resonance modes, TE011 and TM011.

4.1.4. Influence of Incident angle and polarization angle on absorption characteristics of MPA-2DP integrated with MoS_2

Due to the symmetric nature of the structure, the absorption spectrum remains almost unchanged when the polarization angle varies from 0 to 80°. The material also demonstrates stability with wide incident angles; when the incident angle of electromagnetic waves increases up to 60°, the absorption spectrum maintains broadband absorption characteristics, with absorption reaching above 80% in the frequency range from 11.1 to 15.6 GHz.

4.1.5. Effect of bending deformation on absorption characteristics of MPA-2DP integrated with MoS_2

Simulation results show broadening of the absorption spectrum when bending with radius R = 100 mm, 50 mm, and 20 mm. When the bending radius decreases from 500 mm to 20mm, the FBW increased from 55.2% to 97.2%. The broadening of the absorption spectrum can be explained based on the structure's asymmetry. When the material is bent, electromagnetic waves reach the material surface at different incident angles. This leads to non-uniform distribution of electric and magnetic fields on the material surface. As the bending radius decreases, this non-uniformity increases, and will induce new resonances due to the electromagnetic asymmetry of the material. Therefore, when the bending radius decreases, the material's absorption spectrum broadens.

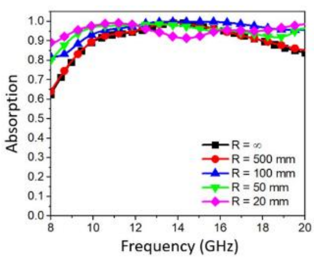


Figure 4.7. *Absorption spectrum of the material when bent with different bending radius.*

4.2. Control of amplitude and absorption frequency of MPA-2DP integrated with MoS_2 in THz frequency range.

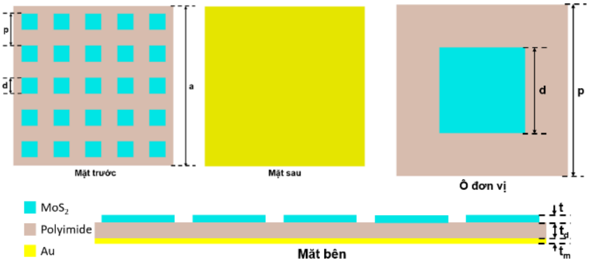


Figure 4.10. *MPA-2DP material model integrated with MoS_2 material*

The proposed metamaterial model has a three-layer structure: polyimide layer between continuous gold metal at the back. The two-dimensional MoS_2 material layer creates resonant structures on the front, having frequency-dependent complex dielectric constant calculated using the Drude model. Simulation results show that the material gives an absorption peak at frequency 2.2 THz with absorption reaching 99.7%.

When bent with different bending radii, we can control its absorption spectrum from single-peak state (when material is flat) to multi-peak absorption state (when material is bent with bending radius from 100 μm). By bending the material, both frequency and absorption of the material have been controlled. Through the bending deformation of resonant structures created by MoS_2, the local electromagnetic field distribution around them can excite new plasmonic resonance modes at different frequencies when THz waves are incident (different incident angles) on each resonant structure. This corresponds to the multi-resonance effect when the sample is bent as in references [99-102].

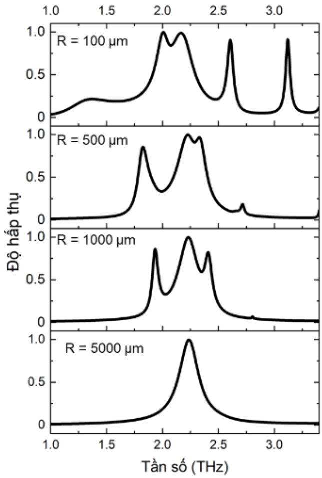


Figure 4.13. *Simulated absorption spectra of MPA-2DP integrated with MoS_2 material bent with R = 5000 μm, 1000 μm, 500 μm, and 100 μm.*

CONCLUSION

The chapter has been designed and studied through simulations for the MPA-2DP structure based on MoS_2 material, which has the ability to change resonance characteristics in the GHz-THz frequency range.

- In GHz frequency range: the material exhibits broadband absorption characteristics, with absorption reaching above 90% in the frequency range from 10.1 to 17.6 GHz. The absorption characteristics of the material in bent state show significantly increased absorption spectrum width.
- In THz frequency range: Single-peak or multi-peak resonance characteristics of the MoS_2-integrated MPA-2DP sample can be achieved based on adjusting sample curvature from 100-5000 μm. New absorption peaks are excited at 2.01 THz (99.6%), 2.17 THz (98.6%), 2.6 THz (90.6%), and 3.12 THz (90.5%).

GENERAL CONCLUSIONS

The dissertation has achieved its objectives and obtained several results regarding the study of multi-band absorption characteristics based on MPA-2DP structures that partially/fully integrate two-dimensional plasmonic materials (graphene and MoS_2):

(i) For MPA-2DP with partial graphene integration in directly excited resonant elements (D-MPA): The near-field (NF) interaction between direct SRR and CS resonances was investigated and evaluated to be strong at a distance d = 0.5mm between them. Simulation and experimental results confirm that NF interaction is reduced when integrating partial graphene layers with surface resistance increasing from 7.2 Ω/sq. to 30 Ω/sq. Consequently, absorption maintains above 90% at 6.8 GHz frequency while absorption decreases below 60% at 8.2 GHz and 8.8 GHz; For MPA-2DPs with partial graphene integration in I-MPA form: strong NF interaction at distance d=0.4mm between SRR and CW resonances, simulation results confirm absorption peaks generated by NF interaction are strongly affected around frequencies 6.9 and 10.0 GHz, while absorption maintains above 90% at 7.35 GHz when the surface resistance of the graphene layer increases to 30 Ω/sq.

(ii) Besides the flexible control of frequency and absorption amplitude due to NF interaction of the above D/I-MPA systems, the plasmonic characteristics of graphene affecting fundamental/higher-order resonances were investigated in several MPA-2DP structural models with full integration through deformation (bending) in GHz frequency range and external voltage control model in THz frequency range. Results show that in GHz frequency range, the presence of graphene material layer (fully integrated/replacing the resonant metal layer on surface) creates Ohmic losses playing a main role in maintaining high absorption peaks above 90% for bent deformation cases. In THz frequency range, the plasmonic characteristics of graphene layer controlled through chemical potential (from 0.2 to 1.0 eV) affected the amplitude (absorption decreased from 95.1% to 88.1%) and absorption frequency (from 170.6 to 177.5 THz).

(iii) Several MPA-2DP structures fully integrating MoS_2 material were investigated through their ability to change resonant characteristics in GHz-THz frequency range. In GHz frequency range: The structure shows broadband absorption characteristics, with absorption above 90% in frequency range from 10.1 to 17.6 GHz. The absorption characteristics of the material in bent state show significantly increased absorption spectrum width; In THz frequency range: single-peak or multi-peak resonant characteristics of MoS_2-integrated MPA-2DP samples can be achieved based on adjusting sample curvature from 100-5000 μm. New absorption peaks are excited at 2.01 THz (99.6%), 2.17 THz (98.6%), 2.6 THz (90.6%) and 3.12 THz (90.5%).

FUTURE RESEARCH DIRECTIONS OF THE DISSERTATION

Although basic research on the above MPA-2DP structures has achieved important initial results, in the next research phase several new challenges need to be addressed before using them in applications, including: Designing structures to increase the number of selective frequency bands, improve stability, increase flexibility and reduce unit cell size when operating at low frequencies; fully evaluate resonant characteristics of "hybrid" structures when integrating additional advanced materials/electronic components into MPA-2DP structures; Expand the ability to control amplitude and phase of electromagnetic waves of MPA-2DP when integrated with electromagnetic wave polarization converters.

Future applications of MPA-2DP can include: manufacturing metamaterials capable of directional electromagnetic shielding for military and healthcare applications; Developing energy storage structures using metamaterials for communications and energy applications; Developing sensors, environmental parameter monitoring/control systems integrated with metamaterials, oriented towards applications in high-tech agriculture and biomedical fields.

List of Published Works from the Dissertation

***. Journal indexed in SCIE**

1. **Man Hoai Nam**, Bui Son Tung, Bui Xuan Khuyen, Duong Thi Ha, Nguyen Van Ngoc, Manh Cuong Tran, Dac Tuyen Le, Vu Dinh Lam, Liangyao Chen, Haiyu Zheng, YoungPak Lee. "*Graphene-Integrated Plasmonic Metamaterial for Manipulation of Multi-Band Absorption, Based on Near-Field Coupled Resonators"*, Crystals, **12**, 525, 2022.

2. **Man Hoai Nam**, Vu Thi Hong Hanh, Nguyen Ba Tuong, Bui Son Tung, Bui Xuan Khuyen, Vu Dinh Lam, Liang Yao Chen and Young Pak Lee. *"Multi-Band Electromagnetically-Induced-TransparencyMetamaterial Based on the Near-Field Coupling of AsymmetricSplit-Ring and Cut-Wire Resonators in the GHz Regime"*, Crystals, **11**, 164, 2021.

3. Duong Thi Ha, **Man Hoai Nam**, Bui Son Tung, Bui Xuan Khuyen, Vu Dinh Lam and Quynh-Le Van. *"Ultra-broadband and flexible Metamaterial absorber based on MoS_2 cubes with Mie resonances*", Journal of the Korean Physical Society, Volume **82**, pages 1047–1054, (2023).

***. Proceedings of the National Conference**

4. Dương Thị Hà, Boudthaly Vankham, Đỗ Thùy Chi, **Mẫn Hoài Nam**, Lê Thị Hồng Hiệp, Phạm Thanh Sơn, Bùi Sơn Tùng, Nguyễn Thanh Tùng, Vũ Đình Lãm và Bùi Xuân Khuyến. "*Nghiên cứu mô phỏng vật liệu biến hóa hấp thụ sóng điện từ băng tần rộng dựa trên vật liệu plasmonic hai chiều MoS_2*", *Hội nghị Vật lý Chất rắn và Khoa học Vật liệu Toàn quốc – SPMS 2021*

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5. Pham Van Dien, Mai Duc Toan, Pham Quang Tuyen, Vu Minh Tu, Vu Dinh Lam, Nguyen Anh Dung**, Man Hoai Nam**, and Tran Manh Cuong. *"Multi-Element Unit Cell Metamaterial Absorber for the GHz Frequency Applications*", HNUE JOURNAl OF SCIENCE Natural Sciences 2023, Volume **68**, Issue 3, pp. 26-33.