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基于图案化石墨烯的宽带太赫兹超材料吸收器

冯越, 刘海, 陈聪, 高鹏, 罗灏, 任紫燕, 乔昱嘉

(中国矿业大学 教育部地下空间智能控制工程研究中心, 江苏 徐州 221116)

摘要:设计了一种基于石墨烯的新型可调谐宽带超材料吸收器。该吸收器采用经典夹层结构, 包含一个图案化的单层石墨烯超表面、介电层和金属底板。该石墨烯图案的结构由不同尺寸的石墨烯谐振器组成, 确保高吸收率的同时能拓宽吸收带宽。研究表明, 当费米能级 E_f 为 0.9 eV 时, 吸收器在光源垂直入射条件下能够实现 2.3~5.2 THz 波段 90% 以上的宽带吸收率, 同时通过控制石墨烯的费米能级可以灵活调节吸收器的带宽和吸收性能。此外, 基于单元结构的对称设计, 吸收器对偏振角度的变化并不敏感, 所设计的吸收器结构将促进石墨烯材料在太赫兹波段和新型吸波器件中的广泛应用。

关键词:超材料; 太赫兹; 时域有限差分法; 石墨烯; 吸收器

中图分类号: TN25

文献标识码: A

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0 引言

超材料(Metamaterial)作为一种具备超常物理性质的人工复合材料,近年来在电磁领域的研究中备受关注。超材料吸收器(Metamaterial Absorbers, MMAs)作为超材料功能器件的重要组成部分,如何在有限的亚波长尺寸内实现多个表面等离子体共振来增加吸收带宽,如何设计厚度小、结构简单、易于制作的高吸收率吸收器结构,已成为超材料领域的研究热点。自2008年LANDY L N等^[1]首次设计出一种吸收率接近100%的微波超材料吸收器之后,超材料吸收器实现完美吸收的波段逐渐拓展到可见光、红外以及太赫兹波段^[2]。其中,太赫兹技术^[3]属于交叉学科的前沿领域,已引起世界上众多国家的重视。目前太赫兹波段的功能器件相对较少,限制了太赫兹技术的进一步应用发展。超材料能够对太赫兹波的振幅、相位、偏振以及传播实现灵活多样的控制,从而提供了一种实现太赫兹功能器件的有效途径。超材料吸收器在实际应用过程中有两个缺点^[4]亟需解决。首先,大多数吸收器结构单元都是由金属构成,极大地限制了其工作带宽,另一个缺陷是很难对电磁或光学性质进行调控,无法满足可调谐的需求。因此,一种可调谐超材料^[5]对于新型吸收器的应用有着重要的意义。石墨烯作为一种零带隙半导体,具有可调谐性、超宽带光学响应、高载流子迁移率^[6-7]等非线性光学特性,在电子学、光学、磁学、生物医学和传感器^[8]等领域的应用前景广阔。石墨烯的载流子浓度和费米能级通过化学气相沉积或者静电掺杂等方式,在太赫兹和红外频率下能够大范围改变,实现对超材料光学特性的调制。PAPASIMAKIS N等^[9]的研究表明,厚度不到1 nm的石墨烯就可以使超材料的性质发生明显的变化,在共振波长附近单层石墨烯只损失2.3%的透过率,同时会极大地增强光与石墨烯的相互作用。此外,石墨烯独特的电子能带结构^[10-11]赋予它非常好的线性光学特性,如低损耗的极化控制和中红外到太赫兹频率范围内的探测。石墨烯优异的光学和电学性能,使其在可调完美吸收器、偏振器和滤波器^[12]等新型光学器件上的应用上具有得天独厚的优势。

本文设计了一种基于图案化石墨烯的三层超材料吸收器,由于结构的对称性,该吸收器是偏振无关的。研究表明,在优化后的结构参数下,当石墨烯的费米能级和弛豫时间分别设为0.9 eV和0.1 ps时,吸收

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第一作者:冯越(1998—),男,硕士研究生,主要研究方向为光纤光栅传感技术。Email: 17361775220@163.com

导师(通讯作者):刘海(1983—),男,教授,博士,主要研究方向为光电子科学与技术。Email: lhai_hust@hotmail.com

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器在 2.3~5.2 THz 的频率范围内可实现 90% 以上的吸收,其相对吸收带宽达到了 77.3%。此外,通过两个峰值处的电场分布并结合上下表面电流密度分布情况研究了相关吸收机制,仿真测试了结构几何参数、介质厚度、石墨烯费米能级以及入射角度对吸收的影响。该吸收器紧凑的三层结构和易于制造的单层石墨烯图案,正适用于隐身、滤波、光谱探测、太赫兹成像等应用领域。

1 结构与方法

图 1 为基于单层石墨烯的可调谐宽带超材料吸收器的单元结构示意图,其中单层石墨烯仅有约 0.34 nm,远小于入射波长,厚度可忽略不计。如图 1(a),该吸收器包括三层:顶层采用优化的液相化学气相沉积工艺^[13]制造的单层图案化石墨烯,介质层采用无损耗介质材料聚四氟乙烯(Polytetrafluoroethylene, PTFE),其相对介电常数设为 $\epsilon_{\text{PTFE}} = 2$ ^[14],厚度设为 $t = 14 \mu\text{m}$,基底选择电导率 $\sigma = 4.56 \times 10^7 \text{ S/m}$ 的金,厚度设为 $h = 0.2 \mu\text{m}$ 。为了实现宽带吸收,采用了多共振结构,保证它们之间能够产生强耦合,如图 1(b)所示,谐振结构由一个中心镂空十字形和四个直角石墨烯片组成,其他几何参数的取值分别为: $p = 25 \mu\text{m}$, $s = 8 \mu\text{m}$, $d = 3 \mu\text{m}$, $w_1 = 3 \mu\text{m}$, $w_2 = 2 \mu\text{m}$, $l = 13 \mu\text{m}$ 。

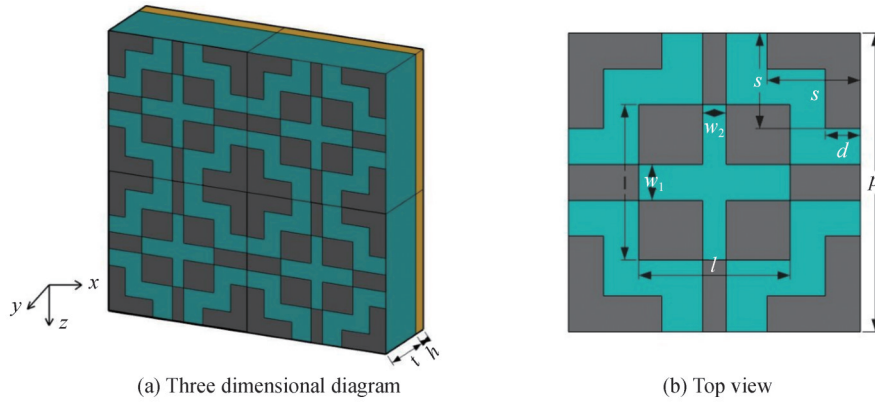


图 1 超材料吸收单元的结构示意

Fig. 1 Structural of metamaterial absorption unit

为了更好地研究该吸收器的吸收,首先要表征石墨烯的光学响应。由于石墨烯是一种二维材料,根据 Kubo 公式^[15-16],石墨烯的表面电导率可描述为

$$\sigma(\omega) = \sigma_{\text{intra}}(\omega) + \sigma_{\text{inter}}(\omega) \quad (1)$$

$$\sigma_{\text{intra}}(\omega) = \frac{iq_e^2 k_B T}{\pi(\omega + 2i\Gamma)\hbar^2} \left(\frac{E_f}{k_B T} + 2 \ln \left(e^{-\frac{E_f}{k_B T}} + 1 \right) \right) \quad (2)$$

$$\sigma_{\text{inter}}(\omega) = \frac{iq_e^2}{4\pi\hbar} \ln \left[\frac{2|E_f| - (\omega + i2\Gamma)\hbar}{2|E_f| + (\omega + i2\Gamma)\hbar} \right] \quad (3)$$

式中, $\sigma_{\text{intra}}(\omega)$ 和 $\sigma_{\text{inter}}(\omega)$ 分别表示带内跃迁贡献和带间跃迁贡献, q_e 为电子电荷常数, \hbar 为约化普朗克常数, k_B 为玻尔兹曼常数。 E_f 和 Γ 分别代表石墨烯的费米能级和散射率, ω 为入射波的角频率, T 为室温,设定为 300 K。本文所采用的调整石墨烯费米能级的方法是在石墨烯与底金属之间施加外部电压 v_g 。石墨烯的费米能级可表示为

$$E_f = \hbar v_F \sqrt{\frac{\pi\epsilon_0\epsilon_r V_g}{q_e t_d}} \quad (4)$$

式中, $v_F = 10^6 \text{ m/s}$ 为费米速度, ϵ_0 为真空介电常数, ϵ_r 为 PTFE 的相对介电常数。对于室温下的太赫兹频段($\hbar\omega \ll E_f$),由于入射光子的能量有限,石墨烯能带结构中电子激发的能带间跃迁的可能性非常小。这里石墨烯的带间跃迁很小,可以忽略不计,此时表面电导率可简化为 Drude^[17]模型

$$\sigma(\omega) = \frac{q_e^2 E_f}{\pi\hbar^2} \frac{i}{\omega + i\tau^{-1}} \quad (5)$$

为了获得最佳性能,利用FDTD Solutions光学软件进行仿真分析,采用渐进式仿真^[18]对结构参数进行优化,综合考虑后获取最优值。由于采用的是单层石墨烯,所以选择其材料库中的2维石墨烯材料。这里要严格说明的是,此方法仅对单层石墨烯正确表示。在边界条件设置中,将单元格的 x/y 方向定义为周期性边界条件,而在 z 方向使用完美匹配层以满足吸收边界条件^[16],网格精度分别设置为 $dx = dy = 0.4 \mu\text{m}$, $dz = 0.2 \mu\text{m}$,该值保证了计算结果的收敛性和可靠性,最后,基于时域有限差分法获得数值仿真结果。另外,利用频域场和功率监测器采集透射率 T 和反射率 R ,吸收率 A 可由公式 $A = 1 - T - R$ 计算得出。由于底部Au材料的厚度远大于电磁波的趋肤深度,因此监视器得到的透射率几乎为零。最终该吸收器的吸收公式可简化为: $A = 1 - R$ ^[19-20]。

值得注意的是,绝大多数文献中是通过改变石墨烯的弛豫时间(Relaxation time)和费米能级(Fermi level)来达到调控目的,其中弛豫时间量纲是秒(s),但在FDTD软件中石墨烯的基本参数之一为散射率(scattering rate),量纲单位为eV,因此涉及到两者之间的量纲转换。从Drude模型可以推导出弛豫时间 τ 与散射率 Γ 存在二倍倒数关系: $\tau = \frac{1}{2\Gamma}$,而 $\tau = \frac{\mu E_f}{ev_f^2}$,其中 μ 是载流子迁移率,进一步推算关系式,最终得出两者的关系为: $\Gamma = 0.5 \times \text{hbar} / \tau$,其中 $\text{hbar} \approx 6.58 \times 10^{-16} \text{eV} \cdot \text{s}$ 。在整个仿真过程中,石墨烯的费米能级设为0.9 eV,弛豫时间为0.1 ps(即散射率等于0.003 3 eV)。

2 结果与讨论

首先研究了两种常见的基本结构在正入射条件下的吸收性能,分别为一个孤立的十字结构和四个矩形组成的阵列,结果如图2。由吸收频谱可以看出,这两个结构都无法满足宽带要求且吸收效率不高,接着考虑将两种结构进行适当的拆解组合,最终设计出由四个直臂和四个矩形组成的、中间为镂空十字的结构,从绿色曲线可以看出该组合结构表现出了宽带吸收,其低频吸收峰和高频吸收峰和上述两个结构的吸收峰相互对应,但吸收性能依旧较低,因此在其四周增加了四个直角石墨烯结构,通过共振效应来提高吸收效率,最终经过仿真和参数优化得到了完美吸收曲线。从黑色曲线可以看出,吸收率在90%以上的带宽达到了2.9 THz,对应的相对带宽达到了77.3%。同时,可以看到有两个吸收峰在 $f_1 = 2.56 \text{ THz}$ 和 $f_2 = 4.6 \text{ THz}$ 频率处的吸收率分别达到了97%和98.9%。基于以上结论,充分证明利用石墨烯图案间的耦合效应可极大地提升吸收器的吸收性能,进而实现宽带吸收。

对于提出的太赫兹频率范围,我们讨论了不同结构参数对吸收特性的影响。当 $p = 25 \mu\text{m}$, $s = 8 \mu\text{m}$, $d = 3 \mu\text{m}$, $h = 0.2 \mu\text{m}$ 时,只改变 w_1 、 w_2 、 l 和 t 。首先,当十字的宽度 w_1 在2~5 μm 之间以1 μm 的间隔变化时,另外三个参数分别设为 $w_2 = 2 \mu\text{m}$, $l = 13 \mu\text{m}$, $t = 14 \mu\text{m}$ 。从图3可以看出,两个吸收峰的形成直接或间接地与沿着两个臂端激发的共振偶极子有关,当 $w_1 = 3 \mu\text{m}$, $w_2 = 2 \mu\text{m}$ 时局域等离子体共振达到最强。除了交叉臂的宽度外,交叉臂的长度也会对吸收光谱产生影响,主要体现在吸收效率上,由图3(b)可知,吸收效率先增大后减小,此外,随着中心十字长度的增加,吸收峰经历了一条类似山峰的路径。最后一个参数是介质层的厚度,在 $t = 14 \mu\text{m}$ 的吸收光谱中,中心频率为3.75 THz,对应于80 μm 的波长。此外,PTFE的折射率为 $n = \sqrt{\epsilon}$,即介质层对应的波长接近56.6 μm 。因此,吸收的物理机制可以用干涉对消原理来解释,由于介质厚度为14 μm ,接近有效光路的1/4,满足反射与入射之间的干涉对消条件,反射波减小。因此,当介质层厚度为14 μm 时,吸收器表现出较强的吸收效应。实际上,仿真中发现介质层的厚度是非常重要的,在第一个界面通过相消干涉^[21]时,它和介质层共同耦合入射波的电磁分量,同时可以独立调制电场和磁场响应,使结构阻

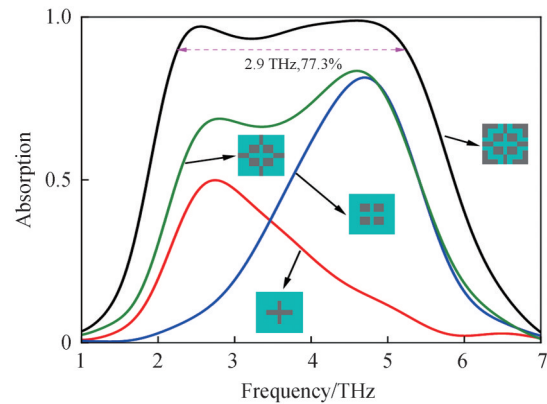


图2 不同石墨烯结构的吸收光谱

Fig. 2 Absorption spectra of different graphene structures

抗与自由空间相匹配,从而达到完美吸收。

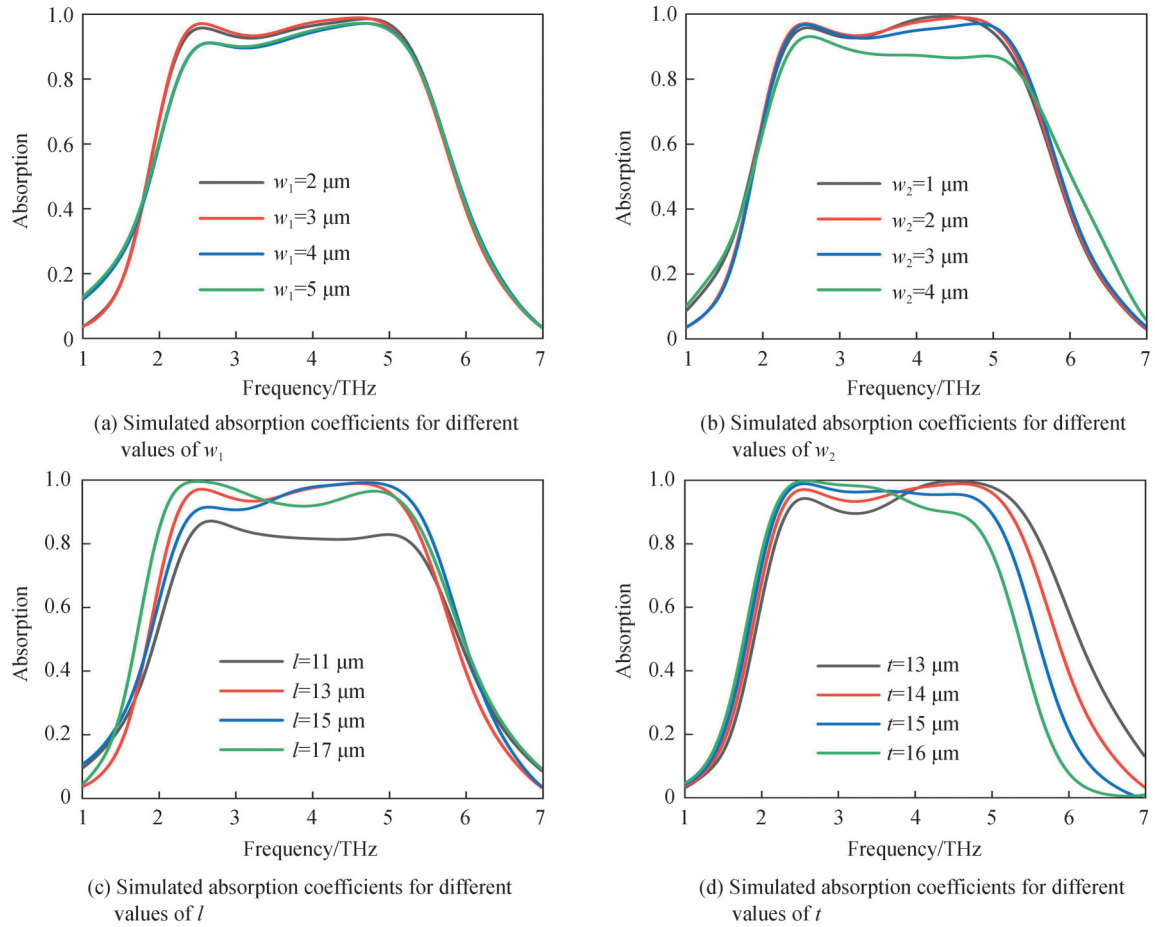
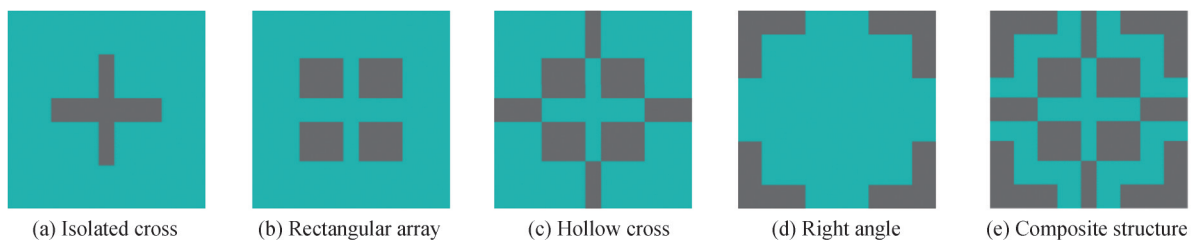


图3 正入射下不同参数的模拟吸收系数

Fig. 3 Simulated absorption coefficients of different parameters under normal incidence

为了探究给定几何参数下的横向吸收机制,首先通过监视器观察两个独立结构的吸收峰对应的电场分布。从图4可以看出,孤立十字结构的电场分布在水平轴两端,而在矩形阵列中主要集中在四个矩形石墨烯片的左右两侧并在中轴形成强耦合,经过重新组合后的结构如图4(c)所示,可以看出在第一个吸收峰($f_1=2.8$ THz)处,入射光激发石墨烯的电子横向振荡,导致电场在中心十字的左右臂端集中,当 $f_2=4.6$ THz时,电场集中在四个矩形石墨烯片上。因此,局部表面等离子体共振和等离激元晶格共振是此组合结构实现宽带吸收的原因。但由图2的吸收谱可以看出,此时依然无法实现高效吸收,通过分析前面的电场图,认为主要原因是由于晶格的纵向缺少共振,因此加入了直角结构,通过图4(i)~(l)发现,在两个吸收峰处,强电场集中在晶格的上下两端。将两者结合起来得到最终的复合结构,如图4(e)所示,在第一个吸收峰时,电场集中在晶格的纵向以及横向臂端的左右两侧,在另一个吸收峰处,电场则增强分布在四个矩形石墨烯片上。通过上述电场分析,可以总结出复合结构的完美吸收主要归因于镂空十字结构的偶极等离子体模式、四个直角石墨烯片水平的四极共振和近场耦合复合结构的叠加。



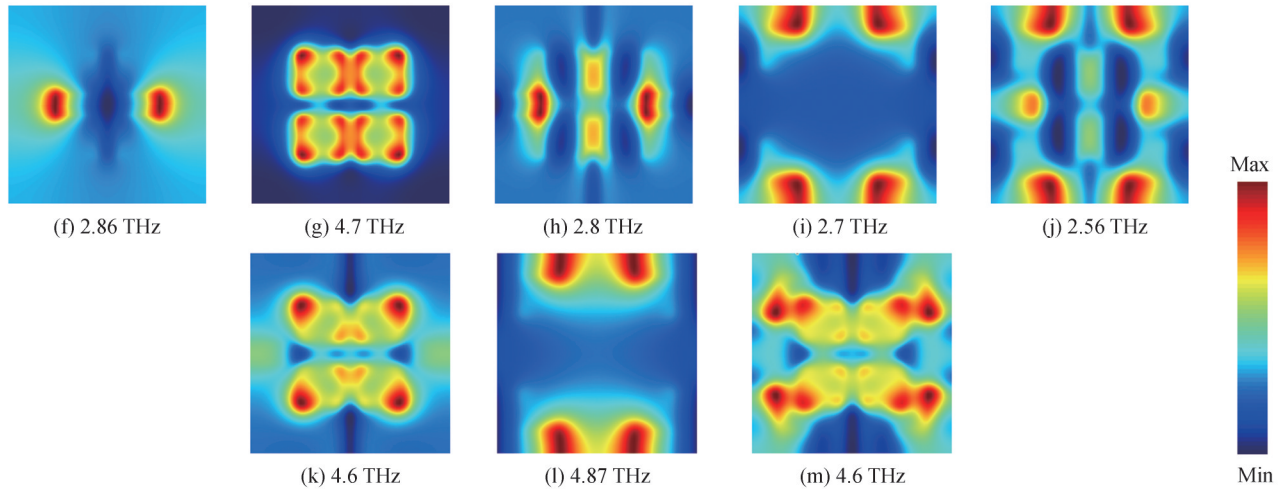
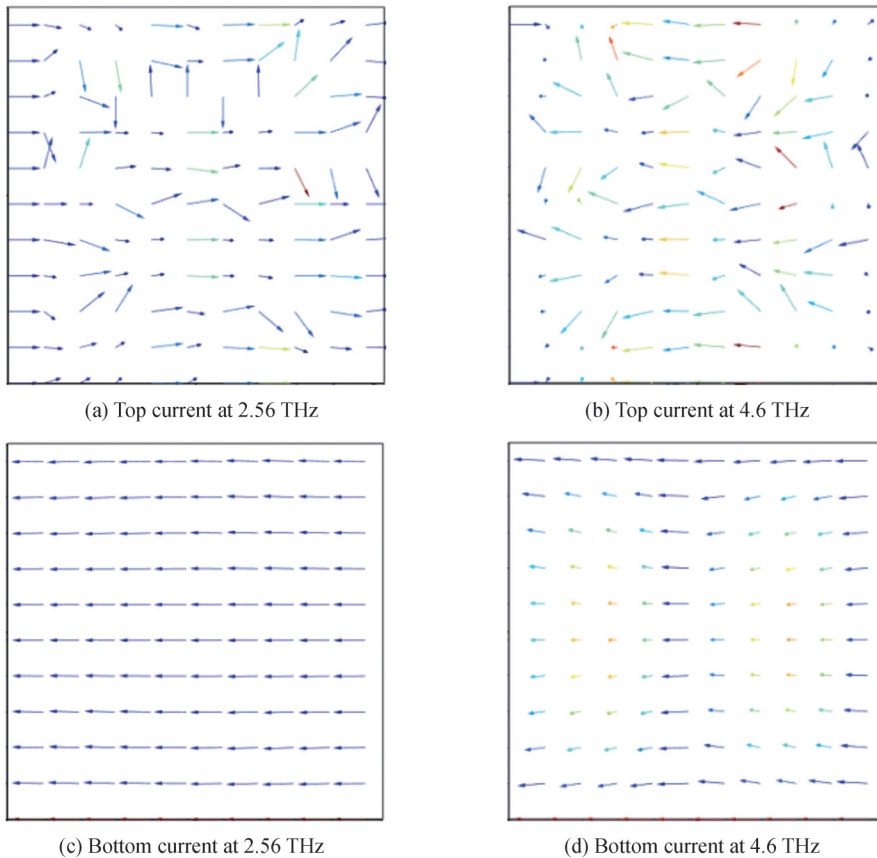
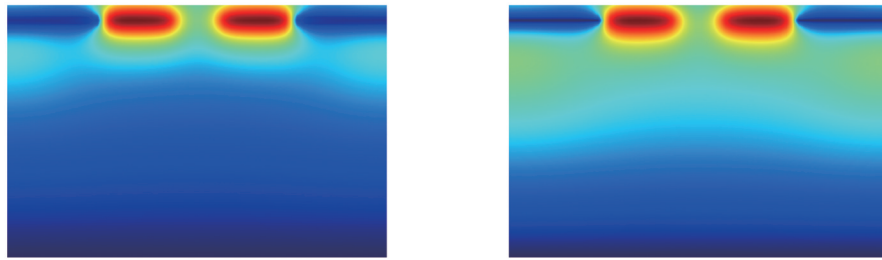


图4 不同石墨烯结构的吸收峰处的电场分布

Fig. 4 Electric field distribution at absorption peaks of different graphene structures

图5为两个共振频率处的表面电流分布以及 yo z平面处的电场分布,更深入地揭示了吸波原理。通过图5(a)可以发现,当 $f=2.56$ THz时,顶层的表面电流流向为自左向右,而底部金属层的电流方向则与之相反,此时在整个结构中产生了一个等效的电流环,从而导致磁耦合共振^[22]。而在4.6THz处,上下表面形成了同向电流,其产生的极化磁场可以与入射波的磁场发生相互作用,同时超材料吸收器中的强电磁共振会将入射的电磁能量限制在结构内部并消耗掉,从而实现入射电磁波的有效吸收^[23],极大地增强了光的吸收效率。从图5(e)可以看出,在 $f_1=2.56$ THz时,电场主要集中在十字的左右臂端和相邻单元结构的间隙处,当 $f_2=4.6$ THz时,电场主要集中在电介质和镂空十字的边缘处。这与文献中阐述的现象一致^[24-26],也进一步验证了图4中不同模式下的吸收性能。





(e) Electric field distribution on yoz plane at 2.56 THz (f) Electric field distribution on yoz plane at 4.6 THz

图5 吸收器在两种不同共振频率下的表面电流分布和 yoz 平面的电场分布

Fig. 5 Surface current distribution and electric field distributions of the absorber in yoz plane of absorber at two different resonance frequencies

进一步研究了石墨烯的可调谐特性。如前文所述,石墨烯作为一种可调谐材料,在可调太赫兹与光学超材料研究中有着重要应用,其费米能级的变化将直接影响表面电导率,对石墨烯的吸收起着决定性作用。因此,本文利用优化后的结构参数,计算了不同费米能级下对正入射吸收光谱的影响。图6为在正入射波下的频率和不同费米能级的函数关系。结果表明,当费米能级从0逐渐增加到0.9 eV,步长为0.3 eV时,峰值吸光度从12%增加到98%以上,同时,随着费米能量的增加,工作带宽也随之变宽。因此,这意味着通过改变石墨烯的费米能级,在特定频率范围内可使所提吸收器的工作状态在反射与吸收之间自由切换^[12],大大增强了其实用价值。

在实际应用中,除了要求高吸收率,所设计的结构对入射波的偏振角和斜入射角度的稳定性同样不可忽视。图7为不同偏振角 ϕ 以 20° 步长从 0° 变化到 80° 时的吸收光谱,可以发现随着偏振角度的改变,所提吸收器的吸收性能几乎没有发生变化,吸收率仍然高于90%。实际上,几何结构的对称性^[27-28]是吸收器能够产生完美偏振无关的根本原因。图8显示了在TE和TM模式下吸收对斜入射角度的依赖关系,随着入射角的增大,吸收器的性能必然降低。在仿真中,对入射角从 0° 到 60° 的范围进行了扫描分析,从图8(a)可以看出,TE模式下,在3.3~5.1 THz的频率范围内,当入射角达到 38° 时,吸收率依旧保持在90%以上且吸收带宽基本不变,有趣的是,虽然在TM模式中入射角仅能达到 26° 才可以维持此标准,但吸收带宽却增大了,这是由斜入射与镂空十字形谐振腔的有效耦合引起的。需要注意,随着入射角的增加,四个直角石墨烯片与中心镂空十字形石墨烯片之间的耦合会变弱,产生共振频率的蓝移。

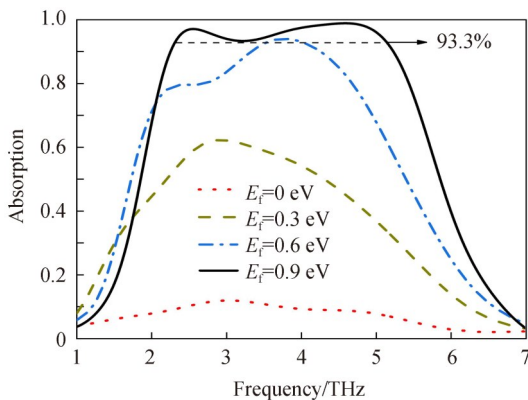


图6 吸收器在不同费米能级下的吸收光谱

Fig. 6 Absorption spectra of absorber under different Fermi levels

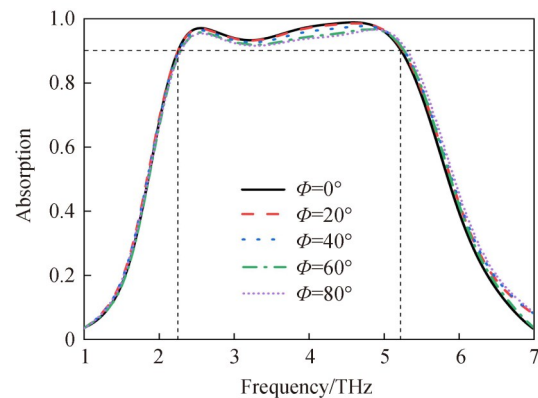


图7 不同偏振角下的吸收光谱

Fig. 7 Absorption spectra under different polarization angles

另外,比较了部分基于单层图案化石墨烯的宽带吸收器在吸收率大于90%的相对带宽,并对模型的几何尺寸进行了比较,计算结果如表1。可见,本文设计的超材料吸收器具有更宽的相对吸收带宽和更紧凑的几何尺寸,潜在的应用价值较高。

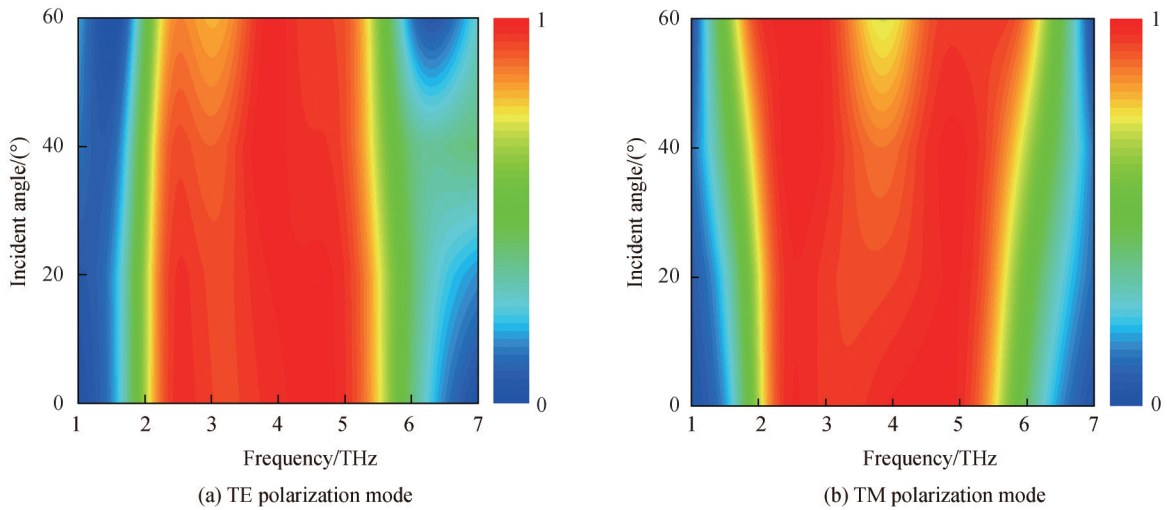


图8 吸收光谱在不同极化模式下对入射角的依赖关系

Fig. 8 Dependence of absorption spectrum on incident angle under different polarization modes

表1 所提吸收器性能参数与参考文献比较

Table 1 Comparison of performance parameters of the proposed absorber with references

Ref.	Absorption > 90% Frequency band/THz	Relative bandwidth	Thickness/ μm
[3]	1.93~3.18	48.9%	15.3
[12]	5.88~10.01	52%	5.1
[16]	1.5~3.0	66.7%	20.2
[24]	1.38~3.12	77.3%	22.2
[26]	2.67~4.84	57.78%	11.2
This work	2.3~5.2	77.3%	14.2

3 结论

本文设计了一种基于石墨烯的宽带可调谐超材料吸收器。该吸收器采用传统的三层夹层结构,由石墨烯、PTFE介质层和金属底板构成。研究表明,在2.3~5.2 THz范围内的相对吸收带宽达到77.3% (90%以上的吸收),这意味着近乎完美吸收,可以解释为2.56 THz和4.6 THz下两个石墨烯等离子体共振的相互作用。在这两个频率下,峰值吸收分别可以达到97%和99%。特别是通过调节石墨烯的费米能级和弛豫时间,能灵活地调节石墨烯的吸收率。由于吸收器的对称性,该吸收器对偏振不敏感,通过分析谐振频率处的电场分布,验证了其宽带吸收、结构紧凑的优势。

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Broadband Terahertz Metamaterial Absorber Based on Patterned Graphene

FENG Yue, LIU Hai, CHEN Cong, GAO Peng, LUO Hao, REN Ziyang, QIAO Yujia
(*Underground Space Intelligent Control Engineering Research Center, Ministry of Education, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China*)

Abstract: The frequency of terahertz radiation is 0.1~10 THz, which is between microwave and infrared. In terms of energy, it is between electrons and photons. Terahertz, like X-rays and sound waves, can penetrate the surface of objects for imaging. In addition, the terahertz frequency is very high, so its spatial resolution is also very high; it also has very high temporal resolution because of its short pulse (picosecond order). Terahertz radiation has been widely used in safety inspection because different chemicals can absorb terahertz radiation at different frequencies to different degrees, showing unique frequency characteristics. In addition, it shows unique advantages and broad application prospects in many fields such as radar, communication and nondestructive testing. It can be predicted that terahertz technology will be one of the major emerging fields of science and technology in the 21st century. However, there are relatively few functional devices in the terahertz band at present. The fundamental reason is that most natural materials can only exhibit weak electromagnetic response when interacting with Terahertz waves, which limits the further development of terahertz technology. The absorption of terahertz waves, especially the complete absorption, has great potential application value in electromagnetic stealth, thermal sensing and thermal imaging. Therefore, the search for an absorbing material that can perfectly absorb the terahertz band has become a major research topic in the direction of materials science in various countries. Traditional absorbing materials are mostly designed based on the principle of Salisbury absorption screen. Its typical weakness is that it is very thick and bulky. With the increasing demands on the performance of absorbing materials in the fields of communication and stealth, traditional absorbing materials can no longer meet the needs of civil and military applications. Therefore, the development of more lightweight and miniaturized new wave absorbing devices has become an urgent task at present. With the discovery and research of metamaterials, it provides an effective way to realize terahertz functional devices, especially terahertz absorption devices. Metamaterials are artificial electromagnetic structures typically made of subwavelength metals on dielectric or semiconductor substrates. Compared with traditional materials, metamaterials have some special properties, such as changing the normal properties of light or electromagnetic waves, and such effects can not be achieved by traditional materials. The research shows that using the exotic electromagnetic properties of metamaterials can not only improve the performance of antennas and microwave devices, develop new equipment, but also provide a new technical means for the development of new absorbing materials. Most of the metamaterial absorbers proposed so far are composed of precious metals such as gold and silver. Once the size is determined, it is difficult to adjust the resonance frequency and absorption intensity. At present, the research on terahertz metamaterials at home and abroad is more focused on the application of metamaterials in the realization of tunable functional devices in the terahertz band, especially the realization of tunable terahertz functional devices. Therefore, the development of tunable metamaterial absorbers will be of great significance for the application of metamaterials. Fortunately, the medium on which processing is relied also plays a role in the electromagnetic properties of metamaterials. Studies have shown that the control of the optical properties of metamaterials can be achieved very effectively by combining metamaterials with media with tunable optical properties such as graphene, liquid crystal and phase change materials. Among them, graphene, as a band-free semiconductor, has attracted much attention in the field of materials science in recent years due to its unique electrical and optical properties. Graphene also has important applications in the research of optically tunable metamaterials. Through chemical or electrostatic doping, the carrier concentration and Fermi level of graphene can be changed, which is very wide on the terahertz frequency range, effectively change the position of the resonance peak and can be used to implement the infrared to the terahertz frequency range adjustable perfect absorber, polarizer, filters and other optical components. Based on the selected topic background and the research status, this paper aims at the research of tunable dielectric metamaterials and the design and optimization of broadband terahertz metamaterial absorber structures, mainly on the optical

properties and tunability of graphene in metasurfaces. The main research content is to take graphene two-dimensional planar metasurface as the research object, design a patterned graphene broadband metamaterial absorber model, and conduct simulation analysis through FDTD solutions optical simulation software. The modulation of graphene on the optical properties of metamaterials and the control of the amplitude, polarization and propagation of terahertz waves by metamaterial are studied based on the FDTD method and principle of surface plasmon resonance, and the structural parameters are optimized by progressive simulation. The absorber adopts a classical sandwich structure containing a patterned single-layer graphene metasurface, a dielectric layer, and a metal backplane. The structure of the graphene pattern consists of graphene resonators of different sizes, ensuring a high absorption rate while broadening the absorption bandwidth. The results show that when $E_f=0.9$ eV, the absorber can achieve a broadband absorption rate of more than 90% in the 2.3~5.2 THz band under the condition of normal incidence of the light source. Meanwhile, the bandwidth and absorption performance of the absorber can be flexibly adjusted by controlling the Fermi energy level of graphene. In addition, based on the symmetrical design of the unit structure, the absorber is not sensitive to the change of the polarization angle, and the designed absorber structure can promote the wide application of graphene materials in the terahertz band and new absorbing devices.

Key words: Metamaterials; Terahertz; Finite difference time domain method; Graphene; Absorbers

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