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Research Progress of Terahertz Nonlinearity on Graphene Materials

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Abstract: *Terahertz waves are located between microwaves and infrared in the electromagnetic spectrum, and have unique properties, such as high penetrating ability, low energy loss, and the advantages of matching the intrinsic resonance frequency with molecules and materials, which make teraherz waves have great application potential in materials science, communications, medical imaging, materials analysis, security and other fields. However, due to the weak transmission capacity of traditional materials in the terahertz band, THz technology faces challenges such as weak nonlinear effects and low signal conversion efficiency. As a two-dimensional material, graphene has become a research hotspot in recent years due to its unique electronic structure of Dirac cone, ultra-high carrier mobility and strong nonlinear optical response, and its characteristics in the terahertz band are particularly interesting. In this paper, we review the research progress of graphene in the field of terahertz nonlinear optics in recent years, focusing on its enhancement strategy, experimental results and technical applications.*

Keywords: *Terahertz, Graphene, Nonlinear Optics*

1. Introduction

Since the 90s of the last century, terahertz technology [1]has developed rapidly, especially the research of teraherz sources [2]and detectors [3]has made remarkable progress. Nowadays, with the emergence of new materials such as graphene [4]and topological insulators [5], the research of teraherz technology has entered a new stage. By taking advantage of the nonlinear optical effects of these materials, the generation, regulation [6], and detection of terahertz waves become more efficient and flexible.

Terahertz radiation [7]generally refers to electromagnetic waves with frequencies between 0.1 and 10 THz, with longer wavelengths and lower energies. In this frequency band, electromagnetic waves can penetrate many non-metallic materials, so they have a wide range of applications in materials science [8], imaging technology [9], and communications.

The nonlinear optical effect [10]refers to the nonlinear relationship between the polarization response of the material and the electric field when the electric field strength is large. Common nonlinear effects include second harmonic generation [11], optical parametric amplification [12], and cubic nonlinear effects [13]. In the terahertz band, the nonlinear response of the material is particularly important, as it enables frequency conversion, amplification, and modulation of terahertz waves.

Graphene, as a single-layer honeycomb structure formed by the arrangement of carbon atoms in a sp^2 hybrid manner, has a special electronic structure that makes it have extremely high carrier mobility and strong optical nonlinear response. These properties make graphene an important place in the study of terahertz nonlinearity [4]. The conductivity and transparency of graphene make it unique in the absorption, emission and regulation of terahertz bands, and it has become a potential material for the next generation of terahertz light sources and detectors.

2. Nonlinear response in the terahertz band

2.1 Research progress of graphene materials in the field of terahertz

Graphene is a two-dimensional (2D) material consisting of carbon atoms arranged into a honeycomb lattice structure, exhibiting a linear electron band structure. The carrier dynamics of graphene are characterized by massless quasiparticles, described by the Dirac equation [14-16]. In solid-state materials with linear dispersion, the momentum scattering rate, a key factor describing carrier dynamics, has been reported to be significantly lower than in parabolic systems, limiting most of the interaction volume to the terahertz frequency range [17]. This property enables a strong interaction between terahertz light and graphene, promoting terahertz nonlinearities such as higher harmonic generation, Pauli blocked [18] saturation absorption [19], and four-wave mixing [20]. As a result, graphene has become a versatile material platform for optoelectronic devices operating in the terahertz frequency range, including terahertz detectors, emitters, and modulators.

Graphene's high carrier mobility allows it to exhibit strong absorption characteristics in the terahertz band. Wang, BK et al. [21] proposed a graphene-based hybrid surface with quasi-continuous bound states (BIC) for the regulation of terahertz wave absorption. Through the strategic application of terahertz element surface structure perturbation, the symmetry-protected BIC is transformed into a quasi-BIC. The introduction of graphene satisfies the critical coupling condition well, which is conducive to obtaining the theoretical maximum absorption of 0.5 in the quasi-BIC in the terahertz band. The quasi-BIC has a Q value of up to 2755. In addition, by changing the asymmetry parameters and Fermi levels, the metasurface exhibits a unique ability to tune and efficiently absorb terahertz waves. This study provides a promising strategy for controlling the absorption of terahertz waves.

Terahertz waves have strong penetrating power, and radars operating in the terahertz band can detect targets protected by various stealth means. Terahertz metamaterial absorbers can efficiently absorb terahertz waves, which is an effective means to deal with terahertz radar in the field of electromagnetic stealth. However, the absorption performance of traditional absorbers cannot be arbitrarily tuned, and some tunable absorbers have complex structures and are affected by environmental factors such as light and temperature, which greatly limits the application and development of absorbers. Chen, JY et al. [22]designed a perfect absorber of dual-band tunable terahertz metamaterials based on patterned graphene, which has an absorption rate of 99.9% at 0.5968THz and 1.7488THz, and the absorption rate and resonance frequency can vary with the change of graphene Fermi level. The absorber can effectively reduce the radar cross section of the target and can be applied to the field of electromagnetic stealth.

The response characteristics of photoexcited graphene (semiconductor-type positive response or metal-type negative response) depend on the Fermi level position, which can be adjusted by gate regulation, doping, and growth processes. This bipolar photoconductive response has potential applications in ultrafast optical modulators that regulate light transmission. However, it is still challenging to achieve a high switching ratio in photoexcited graphene due to the low absorption rate of electromagnetic waves and the limited change in photoconductance. Choi, G et al. [23]experimentally confirmed the negative-responsive high-switching ratio ultrafast terahertz modulation through a graphene/metal nanoslit antenna structure. When graphene covers the nanoslit antenna, the terahertz waves are completely blocked (off-state). This perfect extinguishing source absorbs in the strong local field-enhanced graphene band near the nanogap. However, when the optical pump acts on the graphene/nanoslit antenna, the terahertz transmission recovers in a resonant manner (conduction state) due to the phototranslucent effect of graphene resulting in a significant transition from the off state to the resonant state. In addition, the fast relaxation process induced by carrier redistribution driven by strong terahertz field is a key mechanism to achieve ultrafast modulation of transient terahertz transmission. The research results open up a new way for the application of negative-responsive terahertz modulation with high switching ratio and ultrafast time scale.

By adjusting the applied electric field or chemical doping of graphene, dynamic tuning of the dual-frequency formant position can be realized. The mechanism of the sensor can be elucidated by the excitation of graphene plasmon resonance. In 2024,Fu, MX et al. [24]proposed a tunable dual-band terahertz sensor based on graphene. The sensor consists of a metal bottom layer, an intermediate dielectric layer, and a single layer of graphene etched with four ribbons on top. The numerical simulation results show that the sensor presents two significant absorption peaks at 2.58 THz and 6.07 THz, and the corresponding absorption rates are as high as nearly 100% and 98%, respectively. Its figure of merit (Q value) is 11.8 at 2.58 THz and 29.6 at 6.07 THz. In order to verify the practical application performance of the device, the terahertz response characteristics of different types and thicknesses of the DUT were studied and analyzed, and the frequency shift of the two resonance peaks was observed. The sensor has important potential for terahertz applications in materials characterization, medical diagnostics and environmental monitoring.

A typical terahertz isolator requires a magnetic field to break the time reversal symmetry. Xu, CR et al. [25]proposed a microwave isolator in the terahertz range based on non-reciprocal graphene plasmon operating in a reflection configuration. The bias voltage creates a drift current in graphene, which breaks the time reversal symmetry and causes non-reciprocal reflections. The isolator device has a high isolation of more than 20 dB and an insertion loss of less than 3 dB. In addition, bandwidths with isolation of more than 20 dB can be broadened by a factor of 5 to 1.7 THz by adjusting the carrier density. The isolation, insertion loss and bandwidth of the isolator have a strong correlation with the drift velocity, mobility and air gap thickness of graphene. The study shows the great potential of emerging terahertz technologies, where tunable and electrically tunable isolators are still lacking.

2.2 Terahertz nonlinear enhancement strategy

The electronic structure of graphene has a linear dispersion relationship, and the speed of electrons is close to the speed of light, so its nonlinear response is stronger than that of conventional semiconductor materials. The carriers of graphene accelerate and decelerate rapidly under the action of an applied electric field, resulting in a strong nonlinear effect in the terahertz band.

In 2022, Wang, B. K. [26]et al. proposed a unique scheme to effectively improve terahertz third harmonic generation (THG). By using the high-order mode resonance of the graphene plasma grating, the off-plane transverse local field can be suppressed, so that the electromagnetic energy is tightly concentrated and significantly enhanced in the single-layer graphene. At the same time, the high-density in-plane hot spots can be excited to form a quasi-continuous enhanced field distribution, called a hot surface. This will bring about a huge nonlinear enhancement of the interaction between the terahertz field and graphene, greatly promote the enhanced teraherz field effect in single-layer graphene, and break through the limitations of various patterned metamaterials. Graphene plasma gratings have broad application prospects in the fields of nonlinear terahertz spectroscopy, imaging, and communication.

Metamaterials can significantly amplify the nonlinear response of graphene through the local field enhancement effect. Graphene metamaterials are widely used in optoelectronic devices, optical modulators, and chemical sensors due to their excellent tunability and optical responsiveness in the terahermeth (THz) band. Zhou, SY [27]et al. designed, fabricated, and modulated tunable THz metamaterial absorbers based on patterned graphene. The proposed metamaterial absorber consists of patterned graphene arrays and aluminum (Al) films separated by polyimide (PI). Different THz absorption spectra can be obtained by changing the pattern of graphene. To verify the simulation results, a series of tests were performed using a terahertz time-domain spectrometer (THz-TDS) system. The proposed absorber is not sensitive to the angle of the incident wave. In addition, the chemical doping method was used to change the Fermi level of graphene, and with the increase of Fermi level, the absorption performance of graphene also improved. The experimental results show that there is a resonance between the simulated results and the experimental results. This work will provide further steps in the development of high-performance terahertz devices, including tunable absorption devices, sensors, and electro-optical switches.

The nonlinear response of optical materials provides an important way for optical switching and nonlinear frequency conversion. However, bulk crystals typically have a weaker nonlinear response and require higher pumping energies. Yan, DX et al. [28]investigated the significant enhancement effect of graphene band-assisted ultrathin nonlinear supergrating metasurfaces on third harmonic generation (THG) in the terahertz band. In this structure, the incident pump light achieves strong local field enhancement along the graphene surface due to the generation of strong localized surface plasmon resonance. Taking advantage of graphene's remarkable third-order nonlinear conductance properties, efficient THG conversion can be achieved even at low pump strengths. The results show that the third harmonic conversion efficiency can reach about 2.8% at a low incident intensity of 10 kW/cm^2. The effects of the structure parameters of the supergrating, the graphene Fermi level, the incident angle and the pump light intensity on the working characteristics of THG were systematically explored. This study provides an effective way to greatly improve the THG conversion efficiency in the terahertz band, and opens up a new direction for the development of emerging compact frequency converters and modulators in the field of terahertz chip integration and nonlinear devices.

In 2025, Chao, CTC et al. [29]proposed a technique for achieving high third-harmonic generation (THG) conversion efficiency using bilayer graphene/dielectric/graphene superstructure in the terahertz (THz) range. The enhanced THG mechanism utilizes gap and cavity plasmon resonance at resonant frequencies to result in efficient localization and significant amplification of electromagnetic (EM) waves on graphene surfaces and gap regions. This is due to the induction of interstitial and cavity plasmon resonance. The introduction of a metal substrate under a bilayer structure narrows the resonant response bandwidth, resulting in zero transmittance and the formation of oscillating Fabry-Perot (FP) waves within the cavity. This field enhancement, combined with the high nonlinear conductivity of graphene, increases the THG conversion efficiency (CE) by several orders of magnitude, achieving -24.905 dB at a relatively low fundamental frequency (FF) input intensity. The device is expected to be used in a variety of nonlinear optical and terahertz integrated circuit applications, including terahertz switches and modulators.

3. Conclusion

Although graphene has shown great potential for terahertz nonlinear applications, it still faces many challenges. First of all, the preparation of graphene is still a technical problem, especially the high cost of large-scale, high-quality graphene films, which affects its promotion in practical applications. Secondly, the nonlinear response of graphene is limited by the defects and impurities of its materials, and the nonlinear effect of graphene may decay under some high-intensity terahertz excitation. In addition, the generation and detection efficiency of terahertz waves are still low, especially under low power conditions, and how to improve the sensitivity and output power of terahertz light sources and detectors is still a difficult research point.

Future research will further focus on the optimization and modification of graphene materials. For example, the nonlinear optical properties of graphene can be improved by means of doping, layer number control, surface modification, etc., so as to improve its application efficiency in the field of terahertz. With the development of nanotechnology, the design of micro-nano structures of graphene will provide more innovative ways for the regulation of terahertz waves. In addition, the study of multi-material composite structures will also be an important direction in the future, for example, combining graphene with other two-dimensional materials (such as transition metal sulfides, black phosphorus, etc.) to achieve more efficient terahertz nonlinear responses by taking advantage of heterostructures.

In terms of terahertz communication [30], the nonlinear properties of graphene are expected to provide new solutions for high-speed wireless communication. Through the development of new graphene-based terahertz sources and detectors, the bandwidth and transmission distance of communication frequency bands can be greatly improved. In addition, the application of graphene in terahertz imaging and detection will also have a wide range of application prospects in biomedicine, security and other fields. In general, the research of graphene in the field of terahertz nonlinearity not only promotes the progress of fundamental physics, but also provides the possibility of future application technology.

Graphene, with its unique electronic structure and nonlinear optical properties, occupies a pivotal position in teraherz technology. In this paper, we review the latest research progress of graphene materials in the field of teraherz nonlinearity, including their applications in teraherz absorption, nonlinear light sources, detectors and signal regulation. Although graphene still faces challenges in terms of fabrication process, material quality, and efficiency in practical applications, its huge potential makes it one of the core materials for future terahertz technology. Future research will focus on the optimization of graphene materials, the design of heterostructures, and the combination with other novel materials. With the deepening of research, the application of graphene in the field of terahertz nonlinearity is expected to be significantly improved in the next few years, bringing revolutionary progress to the fields of terahertz communication, imaging, and medical diagnosis.

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