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Abstract

Graphene plasmons have attracted a lot of attention due to large confinement and small mode volume. However, the graphene-based plasmonic devices are still limited in the practical applications due to relatively small light absorption of graphene and limited light–matter coupling efficiency in general excitation strategy. Here, this work reported a strong plasmonic coupling effect observed in a novel graphene-Bi₂Te₃ heterostructure on the top of silicon gratings. It is interesting to find that the extinction spectra of the graphene-Bi₂Te₃ heterostructure has shown three times greater magnitude than that of graphene. This observation is mainly attributed to two factors: first, the coupling efficiency between the graphene and Bi₂Te₃ second, the higher light absorption in the graphene-Bi₂Te₃ heterostructure. Moreover, the plasmonic resonance peak of the graphene-Bi₂Te₃ heterostructure can be easily tuned by changing the grating period just like what happens in the graphene film. In all, this work utilizes the simple silicon grating to couple the light into the graphene-Bi₂Te₃ heterostructure, and further explores the hybridized Dirac plasmons in the graphene-Bi₂Te₃ heterostructure. We believe it will stimulate the interest to study the variant plasmonic heterostructure and trigger new terahertz device applications.

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Highly efficient plasmon excitation in graphene-Bi₂Te₃ heterostructure

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Graphene plasmons have attracted a lot of attention due to large confinement and small mode volume. However, the graphene-based plasmonic devices are still limited in the practical applications due to relatively small light absorption of graphene and limited light-matter coupling efficiency in general excitation strategy. Here, this work reported a strong plasmonic coupling effect observed in a novel graphene-Bi₂Te₃ heterostructure on the top of silicon gratings. It is interesting to find that the extinction spectra of the graphene-Bi₂Te₃ heterostructure has shown three times greater magnitude than that of graphene. This observation is mainly attributed to two factors: first, the coupling efficiency between the graphene and Bi₂Te₃; second, the higher light absorption in the graphene-Bi₂Te₃ heterostructure. Moreover, the plasmonic resonance peak of the graphene-Bi₂Te₃ heterostructure can be easily tuned by changing the grating period just like what happens in the graphene film. In all, this work utilizes the simple silicon grating to couple the light into the graphene-Bi₂Te₃ heterostructure, and further explores the hybridized Dirac plasmons in the graphene-Bi₂Te₃ heterostructure. We believe it will stimulate the interest to study the variant plasmonic heterostructure and trigger new terahertz device applications. © 2016 Optical Society of America

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1. INTRODUCTION

Surface plasmons (SPs), which are collective oscillations of electrons localized at the conductor/dielectric interface, have been discovered in atomically thin layered materials such as graphene [1]. Due to a number of favorable properties, such as ultrahigh confinement and electrical/chemical tunability, graphene plasmonic structures are widely studied. More importantly, the lifetime of SP modes in graphene can reach hundreds of optical cycles, which potentially circumvents one of the major bottlenecks faced by noble-metal plasmonics [2,3]. The graphene surface plasmon polariton has been verified by both infrared transmission spectroscopy and scanning near-field optical microscopy in real-space [4,5]. In order to achieve efficient light-plasmon coupling, Ju *et al.* explored plasmon excitations by engineering graphene film into microribbon arrays, and found that graphene plasmon resonance peak can be tuned over

a broad terahertz frequency range by changing microribbon width and *in situ* electrostatic doping [6]. However, it is noteworthy that the single-layer graphene supports relatively low resonance frequency/magnitude dependence on carrier concentration. Thus, Yan *et al.* fabricated graphene/insulator multilayer stacks to effectively enhance the plasmonic magnitude, which is based on the Dirac fermions' carrier density scaling law [7]. Similarly, Zhang *et al.* used atomically thin hexagonal boron nitride films as spacers to enable strong plasmon coupling between adjacent graphene layers in the vertical dimension, which produces two times upshift of plasmonic resonance frequency and three times enhancement of its magnitude [8].

The surfaces of topological insulators (TIs) and graphene share similar low-energy electronic structures in which the conduction and valence bands touch at Dirac points. Dirac fermions also occur in the two-dimensional (2D) electron gas that

forms at the surface of TIs as a result of the strong spin–orbit interaction existing in the insulating bulk phase [9]. The spin-plasmons, collective modes originated from the transverse spin fluctuations caused by density fluctuations, have been theoretically predicted in TIs [10]. Pietro *et al.* reported the first experimental evidence of Dirac plasmon excitations in a TI (Bi_2Se_3 thin microribbon arrays) [11]. The plasmonic resonance in the TI is attributed to the bounded oscillation of Dirac quasiparticles of the conducting 2D edge state. According to theoretical studies [12], the plasmon in graphene will interact and hybridize strongly with itself by inserting a thin dielectric spacer between two layers of graphene to form a “metal–dielectric–metal” sandwich structure. Though the graphene and TI plasmons are considered to have the same origin [13], the experimental observation of hybridization between graphene plasmon and other Dirac plasmons has not been reported.

In this work, we fabricated a new graphene- Bi_2Te_3 heterostructure by growing Bi_2Te_3 on the graphene template. With the help of silicon gratings underneath the heterostructure, the normal incident light can successfully excite the plasmon resonance in the graphene- Bi_2Te_3 heterostructure [Fig. 1(a)]. It is interesting to find that the extinction spectra of the graphene- Bi_2Te_3 heterostructure has shown $3\times$ greater magnitude of that of the graphene, which is attributed the plasmon coupling between the plasmon at the graphene surface and the top surface of the Bi_2Te_3 thin film. Moreover, the extinction peak of the plasmonic resonance of the graphene- Bi_2Te_3 heterostructure can be easily tuned in a broad terahertz (THz) frequency range by varying grating periods just like what happens in the graphene film. Since plasmon in graphene and plasmon in Bi_2Te_3 share a similar origin, the study of the hybridization between them will stimulate the interest to study variant plasmonic heterostructures and trigger new terahertz device applications.

2. RESULTS AND DISCUSSION

Bi_2Te_3 is a small bandgap (~ 0.145 eV) material with rhombohedral crystal structure, in which a sequence of Te-Bi-Te-Bi-Te along the *c*-axis is followed. Each of the smallest vertically repeated units forms one quintuple layer (QL). It is covalently bonded within the QLs, while attached together via van der Waals force between the QLs. In the *x*–*y* plane, Bi_2Te_3 shares a similar hexagonal lattice with graphene. The periodicity of Bi_2Te_3 along graphene carbon atoms is 4.38 Å, and the length of graphene C–C bond is 1.42 Å. Thus, the lattice mismatch between Bi_2Te_3 and graphene is 2.7% (0.12 Å), which is ideal for van der Waals epitaxial growth [Fig. 1(b)] [14–17]. This work utilizes the chemical vapor deposition (CVD) method to fabricate the graphene- Bi_2Te_3 plasmonic heterostructure. First, monolayer graphene film was grown on copper substrates. Then, the Bi_2Te_3 nanocrystals were deposited on the graphene film. Subsequently, they were transferred onto silicon grating substrates with the help of a thin poly-(methyl methacrylate) (PMMA) layer. Finally, the PMMA layer was removed via thermal annealing in the vacuum.

The scanning electron microscopy (SEM) images of graphene- Bi_2Te_3 heterostructure films with different thicknesses are shown in Figs. 2(a) and 2(b). The reaction times for

Fig. 1. Schemes showing (a) the graphene- Bi_2Te_3 heterostructure on the silicon diffraction grating and (b) lattice mismatch between graphene and Bi_2Te_3 .

Fig. 2. Characterizations of the graphene- Bi_2Te_3 heterostructure. (a),(b) SEM images and (c),(d) AFM images of graphene- Bi_2Te_3 heterostructures with different Bi_2Te_3 thicknesses. The Bi_2Te_3 thicknesses in (a) and (c) are around 20 nm; in (b) and (d) are around 50 nm. Scale bars in (a) and (b) are $2\text{ }\mu\text{m}$. Scale bars in (c) and (d) are $1\text{ }\mu\text{m}$. (e) TEM image of the graphene- Bi_2Te_3 heterostructure. The inset of SAED pattern was taken at the red square area in the TEM image. Scale bar is $1\text{ }\mu\text{m}$. (f) HRTEM images taken at the yellow square area in TEM image [Fig. 2(e)]. Scale bar is 2 nm .

growing Bi_2Te_3 samples shown in Figs. 2(a) and 2(b) are about 30 min and 60 min, respectively. The direct consequence of the deposition times is the variation in the Bi_2Te_3 thickness. As revealed in the atomic force microscopy (AFM) image [Figs. 2(c) and 2(d)], the thicknesses of Bi_2Te_3 in the heterostructure shown in Figs. 2(a) and 2(b) are 20 and 50 nm, respectively. The microstructure of the graphene- Bi_2Te_3 heterostructure film was further characterized by transmission electron microscopy (TEM) [Fig. 2(e)]. Both graphene and Bi_2Te_3 nanocrystals are revealed, in which the latter shows a quasihexagonal shape. The regular shape and sharp edges indicate an excellent crystallinity of Bi_2Te_3 [18]. The selected area electron diffraction (SAED) pattern [inset of Fig. 2(e)] exhibits a clear hexagonal symmetry, which can be indexed to along $[001]$ zone axis of graphene- Bi_2Te_3 . This further confirms the single crystalline nature of Bi_2Te_3 . The atomic structure of Bi_2Te_3 was characterized using high-resolution transmission electron microscopy (HRTEM) at 200 keV. Figure 2(f) clearly resolves the hexagonal lattice fringes. In particular, the lattice fringes are structurally uniform with a spacing of 0.21 nm. It is in good agreement with that of the $(11\bar{2}0)$ planes of the rhombohedral phase of Bi_2Te_3 [19].

Fig. 3. Fabrication of graphene-Bi₂Te₃ plasmonic devices. (a) Optical microscope image of the silicon grating. Scale bar is 10 μm . (b) SEM image showing the graphene-Bi₂Te₃ heterostructure on the Si grating. Scale bar is 10 μm . (c) Tilted SEM image showing the graphene-Bi₂Te₃ heterostructure on a 2 μm deep silicon grating. Scale bar is 5 μm . (d) Raman spectrum of the graphene-Bi₂Te₃ heterostructure measured at 633 nm excitation.

One-dimensional gratings with the different periods (i.e., 4, 8, 12, and 16 μm) were fabricated with intrinsic silicon. Figure 3(a) shows an optical microscope image of a silicon grating with a period of 8 μm . After the wet transfer, PMMA-supported graphene and graphene-Bi₂Te₃ films are very well suspended on silicon gratings. The frames of the gratings have been revealed in the SEM image in Fig. 3(b). A clear SEM snapshot was taken from a tilted angle in Fig. 3(c). In order to avoid the mechanical damage made to graphene from solvents, high temperature annealing rather than acetone rinsing was used to remove the PMMA layer. In this way, silicon grating supported graphene and graphene-Bi₂Te₃ films are ready for spectroscopy measurements. Raman spectra were measured at room temperature with the excitation of a 633 nm laser. Sample damage is avoided by controlling the power below 0.5 mW. Figure 3(d) shows a typical Raman spectrum from 20 to 3000 cm^{-1} , which covers the interested spectrum ranges of both the TI and graphene. In the low-frequency range, the Raman spectrum exhibits five characteristic of Bi₂Te₃ crystal at 39, 69, 92, 101, 117, and 137 cm^{-1} , which correspond to the E_g^1 , A_{1g}^1 , A_{1u}^1 , E_g^2 , A_{1u}^2 , and A_{1g}^2 vibrational modes of single-crystalline Bi₂Te₃, respectively. At the same time, two characteristic peaks of graphene are observed at 1598 cm^{-1} (G band) and 2698 cm^{-1} (2D band). Compared with the Raman spectrum of monolayer graphene, it is interesting to observe the blueshift of the G band and significant intensity reduction of the 2D band in the graphene-Bi₂Te₃ heterostructure. This is also an indication of the doping effect on the graphene film [20].

The plasmon resonance in the graphene-Bi₂Te₃ heterostructure was studied using Fourier transform infrared (FTIR) spectroscopy. The extinction spectra of graphene and graphene-Bi₂Te₃ on various periods were shown as solid lines in Figs. 4(a) and 4(b). It is found that the plasmon resonance

Fig. 4. Experimental and simulated spectra of the graphene and graphene-Bi₂Te₃ heterostructures. The experimental extinction spectra of (a) graphene film and (b) graphene-Bi₂Te₃ on silicon gratings with different periods, where incident light is vertically polarized to the grating axis. (c) Simulated frequency shift of graphene film (red line) and graphene-Bi₂Te₃ heterostructure (black dashed line) with various Si grating periods. (d) The extinction spectra of graphene-Bi₂Te₃ on the Si grating with polarized incident infrared light. Polarizer at 90 deg refers to the polarization perpendicular to the grating (red trace) and polarizer at 0 deg refers to the polarization parallel to the grating (blue).

peaks for both graphene and graphene-Bi₂Te₃ heterostructures shift to lower wavenumbers when the grating periodicity is reduced. In particular, the resonance peak shifts from 57.7 to 118.9 cm^{-1} when the grating period changes from 16 to 4 μm for graphene [Fig. 4(a)]. In the graphene-Bi₂Te₃ case, the resonance peak shifts from 52.3 to 97.4 cm^{-1} when the grating period changes from 16 to 4 μm [Fig. 4(b)]. In comparison, the fundamental mode plasmon resonance peak of the graphene-Bi₂Te₃ heterostructure blueshifts by 21.5 cm^{-1} than that of graphene (on a 4 μm grating). It has also been noticed that the resonance tuning range of the graphene-Bi₂Te₃ is smaller than that of the graphene alone. This is caused by the addition of the more dielectric Bi₂Te₃ layer on the top of graphene. Moreover, the resonance of graphene-Bi₂Te₃ under similar incident conditions is enhanced by more than three times. This significant change can be concluded by several reasons: first, the additional Bi₂Te₃ layer enhances the light absorption in the heterostructure; second, the addition of the Bi₂Te₃ layer also introduces the plasmon coupling in graphene and Bi₂Te₃ layered structures. These characteristics attest to the strong plasmonic hybridizing effect between graphene and Bi₂Te₃.

To better understand the plasmonic resonance in graphene-Bi₂Te₃ heterostructures, numerical simulation is performed. In the simulation, the dynamic conductivity of the graphene-Bi₂Te₃ film follows the Drude mode, like graphene [2]. Only the intraband contributions of the spin-plane wave vector for the graphene-Bi₂Te₃ heterostructure have been considered. The simulated extinction spectra of the graphene and graphene-Bi₂Te₃ heterostructures are shown as dashed lines in

Figs. 4(a) and 4(b). It is found that the resonance peak position of the graphene and graphene-Bi₂Te₃ heterostructures matches well with those in the experiment results. The resonant frequency in both the graphene and graphene-Bi₂Te₃ heterostructures increases as the period of the silicon grating decreases, which is consistent with the experimental results. Comparing Fig. 4(a) with Fig. 4(b), it is also noticed that the tuning range of the graphene-Bi₂Te₃ heterostructure has been decreased from 61.2 cm⁻¹ (graphene) to 45.1 cm⁻¹ (graphene-Bi₂Te₃ heterostructure). This is attributed to the increase of dielectric constants in the graphene-Bi₂Te₃ heterostructure. It is also noticed that magnitude of the experimental graphene extinction spectra is much weaker than that of the simulated graphene extinction spectra, which could be explained by the possible defects in the real samples. However, the intensity weakening in the graphene-Bi₂Te₃ heterostructure is less obvious than that in graphene, which indicates the robustness of the graphene-Bi₂Te₃ heterostructure.

In order to have a complete picture about grating-coupled plasmon resonance in the graphene-Bi₂Te₃ heterostructure, we plotted the plasmon resonance of different materials as a function of grating period in Fig. 4(c). The experimental data points were obtained by fitting the experimental spectra in Figs. 4(a) and 4(b) with Lorentzian function. Both simulation and experimental results confirmed that the plasmon resonance in the graphene-Bi₂Te₃ heterostructure can be effectively tuned by changing the grating period. While the period of the silicon grating is fixed, the extra Bi₂Te₃ layer on graphene will cause redshift in plasmon resonance due to the change of dielectric constant. It is interesting to see that thicker Bi₂Te₃ crystals on graphene induces further redshift of the resonance frequency.

Figure 4(d) depicts the extinction spectra ($-\Delta T$) of the graphene-Bi₂Te₃ heterostructure under randomly polarized and linearly polarized illuminations. Under randomly polarized illumination and 0° polarized illumination (polarization direction parallel to the gratings), both spectra (black line and blue line, respectively) have shown a broad bump with a slight peak at 63 cm⁻¹ (~1.89 THz). While, under 90° polarized illumination (polarization direction perpendicular to the gratings), the peak at 63 cm⁻¹ is much stronger than that under 0° polarized illumination. This is because the light perpendicular to the grating axis ($\theta = 90^\circ$) can excite bounded electron oscillation [11].

3. EXPERIMENTAL RESULTS

A. Material Synthesis

Graphene films were grown on 25 μm thick copper foils (Alfa Aesar, item no. 13382) in a typical CVD system and transferred onto the silicon grating. The graphene film is p-doped as a mixed solution of FeCl₃ and HCl was used to etch away copper during the transferring process. In order to maintain the integrity of the graphene-Bi₂Te₃ heterostructures, Bi₂Te₃ nanoplatelets were epitaxially grown on the transferred graphene film on silicon gratings by the physical vapor deposition method in a separate tube furnace. Specifically, high purity Bi₂Te₃ powder (Alfa Aesar, purity: 99.999%) and silicon grating substrate with graphene were placed in the center facing downstream of the furnace, respectively. Then the furnace was heated up to 500°C

and maintained for 90–300 s with argon (with a gas flow rate of 100 sccm) as carrier and protective gas to transport Bi₂Te₃ vapor onto the graphene film. Then the furnace was cooled down naturally to room temperature.

B. Material Characterizations

The morphologies and microstructures of the silicon grating and graphene-Bi₂Te₃ heterostructure were investigated by SEM (FEI Quanta 200 FEG, acceleration voltage: 5–30 kV) and TEM (FEI Tecnai F30, acceleration voltage: 200 kV). Raman spectra were recorded by a micro-Raman system (Horiba JobinYvon, HR800) with an excitation wavelength of 633 nm. The laser spot of the Raman system is focused to <2 μm with ×100 object lens. A Bruker Vertex 80 V FTIR system covering the far-IR (40–700 cm⁻¹) wavelength range was used to investigate the transmission of the graphene-Bi₂Te₃ heterostructure. As for the far-IR characterizations, a liquid nitrogen cooled mercury-cadmium-telluride detector and a far-IR deuterated triglycine sulfate detector were used in conjunction with potassium bromide and polyethylene terephthalate beam splitters, respectively. The reference used in the spectroscopy measurement is the silicon grating without the graphene and TI layer.

C. Fabrication of Silicon Grating

Standard semiconductor fabrication processes were used to fabricate the grating on an intrinsic silicon wafer. UV lithography was applied to define the grating pattern following with dry etching to obtain the gratings.

D. Numerical Simulation

All the numerical simulations were carried out with Lumerical FDTD. Periodic boundary conditions were used along the x-axis and perfect matched layers were used along the z-axis. Linearly polarized light was normally incident on the structures. The refractive index of Si was set to be 3.42. The conductivity of single-layer graphene was calculated with Kubo formula [21], where the chemical potential and scattering rate were set to be 0.5 and 0.005 eV, respectively, to match the experimental results. A layer of Bi₂Te₃ covers the graphene layer with complex refractive index $n = 8.1 + 2.6i$ [22].

4. CONCLUSION

In summary, we have successfully demonstrated a new robust graphene-Bi₂Te₃ heterostructure. This graphene-Bi₂Te₃ heterostructure has shown three times greater magnitude of that of the graphene due to higher light absorption and strong graphene and Bi₂Te₃ coupling. The resonance peak of the graphene-Bi₂Te₃ heterostructure has been proved to be tunable by changing the grating periodicities, theoretically and experimentally. This work opens up new possibilities for the applications of graphene-Bi₂Te₃ heterostructures in various terahertz photonic devices.

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