Bilayer MoS$_2$ on silicon for higher terahertz amplitude modulation

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Abstract

The terahertz (THz) amplitude modulation has been experimentally demonstrated by employing bilayer molybdenum disulfide (MoS$_2$) on high-resistivity silicon (Si). The Raman spectroscopy and x-ray photoelectron spectra confirm the formation of bilayer MoS$_2$ film. The THz transmission measurements are carried out using a continuous wave (CW) frequency-domain THz system. This reveals the higher modulation depth covering wide THz spectra of 0.1–1 THz at low optical pumping power. The modulation depth up to 72.3% at 0.1 THz and 62.8% at 0.9 THz under low power optical excitation is achieved. After annealing, the strong built-in electric field is induced at the MoS$_2$–Si interface due to $p$-type doping in MoS$_2$. This improves modulation depth to 86.4% and 79.7%, respectively. The finite-difference time-domain (FDTD) based numerical simulations match well with the experimental results. The higher modulation depth at low optical power, broadband response, low insertion losses, and simplicity in the design are the key attributes of this THz modulator.

1. Introduction

In the area of terahertz technology, the rapid development of compact and commercial THz system require miniaturized components such as sources, modulators, and detectors [1–5]. In particular, the THz modulator is one of the vital elements for many commercial applications such as wireless communication, imaging, and security scanning [6, 7]. Based on the driving mechanism, the THz modulators are generally classified into four categories that are electrical, optical, mechanical, and thermal [8–10]. In particular, modulation of the THz regime by optical pumping has proven to be an efficient and liable approach because of high modulation depth, low insertion losses, and high ON-OFF switching speed. The operating principle is to change the conductivity of the semiconductor by generating charge carriers with pumping light and hence change in transmission of THz radiations [11, 12]. In recent years, the optical modulation depth can be well meliorated when the silicon substrate is incorporated with metamaterials [13], plasmonic structures [14], organic materials [15], graphene, and transition metal dichalcogenides (TMDs) such as MoS$_2$, WS$_2$, etc. [16, 17]. Until now, very few reports have been presented on TMDs based THz modulators particularly on MoS$_2$ [18–20]. Most of the modulators have shown low modulation depth at large pumping laser power. For graphene and TMDs, laser-induced damage due to the thermal and non-thermal effects have been observed [21, 22]. The high laser intensity can gradually burn out the film. Therefore, in the case of optically pumped TMDs based THz modulators, the modulation characteristics and speed are improved to a great extent, but a conflict between the higher modulation depth and limitation of pumping laser power is the main drawback to resolve.

Herein, an optically driven THz modulator based on bilayer MoS$_2$ on high-resistivity Si substrate working up to 1 THz frequency has been reported. The ultrafast THz photoconductivity, strong dielectric response in THz and visible spectrum make this material suitable for fast optoelectronics and photonic devices [23–25]. The
higher carrier mobility, density of states, and long-term stability, and stronger light–matter interaction of bilayer MoS2 as compared to monolayer MoS2 show greater potential for DC and high-frequency optoelectronics devices [26–28]. It is also accounted that effective p-type doping is possible in MoS2 after annealing in the presence of air, as reported previously [29, 30]. The oxygen atoms incorporated in the MoS2 layer behave as p-type dopants. Therefore, the MoS2–Si samples are annealed at 300 °C for 2 h under an ambient environment [29]. The modulation depth is increased after annealing on account of the strong built-in electric field at the MoS2–Si interface and reaches up to 79.7% at 0.9 THz. The numerical simulations have shown that the photogenerated carrier density around the MoS2–Si interface is more prominent as compared to bare Si, which contributes to higher modulation depth.

2. Experiment and characterization

Bilayer MoS2 is formed on commercially available 500 μm thick high-resistivity Si substrate having resistivity around 5000 Ωcm. The uniform and continuous film of MoS2 as shown in the inset of figure 1(a) is formed through sulfurization of ~1 nm molybdenum (Mo) film [31, 32]. The Raman spectra show two sharp and large peaks at 384.7 cm⁻¹ and 406.5 cm⁻¹, which corresponds to E₁₂g and A₁₇g modes of MoS2, respectively, as given in figure 1(a). The difference between the obtained two modes is 21.8 cm⁻¹, which demonstrates the formation of bilayer MoS2 on silicon [33]. X-ray photoelectron spectra (XPS) of the MoS2 film are given in figures 1(b) and (c) for Mo and S elements, respectively. The stoichiometry (S/Mo) is calculated using the area under the respective peaks and considering the relative sensitivity factor, is found ~2.

Figure 1(d) depicts the schematic diagram of the THz modulator. The quasi-continuous-wave laser of 800 nm wavelength is used as an optical pumping source. The different THz measurements are carried out by using the frequency-domain THz system (TOPTICA’s Terascan 1550) [34, 35] in air atmosphere. It consists of photoconductive antennas as continuous wave THz source and Schottky diode as THz receiver. The emitted THz beam is collimated and focused by four off-axis parabolic mirrors. The sample is then placed at the focus of the beam after second mirror so that the beam spot size remain constant throughout the experiment. The
diameter of the incident THz beam and the laser is 3 mm and 4 mm, respectively. The measured THz spectra of air up to 1 THz frequency are taken as reference.

3. Results

The measured photocurrent of the detector after the incidence of THz and laser beam simultaneously on the samples is shown in figures 2(a) and (b). The values of photocurrent for THz spectra is lowering down for MoS2-Si and anneal-MoS2-Si samples as compared to bare Si. This indicates higher absorption and reflection of THz beam from MoS2 layers on Si. It has been also observed that the photocurrent values at all frequencies decrease gradually with increasing laser power. This is because of the varying conductivity of the samples due to the extra charge carriers generated after absorbing laser power.

The measured THz transmittance of the samples at frequency $\nu$ is:

$$ T(\nu) = \left[ \frac{I_{\text{sample}}(\nu)}{I_{\text{reference}}(\nu)} \right]^2 $$

Where $I_{\text{sample}}(\nu)$ is THz photocurrent with sample and $I_{\text{reference}}(\nu)$ is THz photocurrent without sample. The transmittance plot of the bare high-resistivity Si substrate is depicted in figure 3(a). It shows the characteristic Fabry–Perot oscillations undergo multiple reflections at the sample surface and within the sample due to the difference of refractive index. The measured transmittance spectra of MoS2-Si and annealed-MoS2-Si samples illuminated by different laser power are given in figures 3(b) and (c). It has been observed from these plots that a change in conductivity brings a reduction of THz transmission with increasing laser power. The THz transmission at higher frequencies is very small due to the high absorption coefficient of the silicon substrate and, both the photo mixer output and the detection efficiency of the Schottky diode decrease towards higher frequencies [34, 35]. In the transmission plots, the change in the oscillations is observed after the formation of MoS2 on the Si substrate. There could be several possible reasons such as MoS2 film on the Si sample acts as an antireflection layer to inhibit small THz wave reflections [36, 37]. The changing absorption coefficient of MoS2 film with THz frequency results in a change in transmission with frequency, and further the plasmonic resonance response of the nanometer MoS2 film can also contribute to the oscillations in the transmittance spectra [24, 38].

The modulation depth is computed using:

$$ \text{MD} = \frac{(T_0 - T_P)}{T_0} $$

Where $T_0$ and $T_P$ are a THz transmittance through the samples without and with an incidence of laser power $P$. The modulation depth of samples at 0.1 THz, 0.4 THz, and 0.9 THz frequencies, respectively is shown in figure 4. The bare Si, MoS2-Si, and annealed-MoS2-Si samples are having modulation depth of 8.07%, 14.18%, 21.4%, respectively at 0.1 THz frequency under 0.1 W cm$^{-2}$ low optical excitation power. It is further increased to 47.6%, 72.3% and 86.4% respectively at 1 W cm$^{-2}$. At 0.9 THz frequency, the bare Si, MoS2-Si, and annealed-MoS2-Si samples are having modulation depth of 2.2% 9.9% and 16.5% under 0.1 W cm$^{-2}$, which is improved to 21.2%, 62.8%, and 79.7%, respectively at 1 W cm$^{-2}$ power. It is found that the modulation depth
increases with an increase in pumping power. The reason is an increase in conductivity due to more photogenerated charge carriers with an increase in pumping power, which contributes to a decrease in transmission, as given in figure 3(d). It is also observed that the modulation depth is improved by the annealing process. The maximum modulation depth of 86.4% is accomplished at 0.1 THz frequency for the annealed-MoS2-Si sample. The change in modulation depth with THz frequencies is presented in figure 4(d) for all the samples. The modulation depth of bare Si declines sharply with an increase in frequency, while the MoS2-Si and annealed-MoS2-Si samples show a decline in modulation depth at a slower rate. Therefore, the MoS2 layer on the silicon substrate improves the modulation depth of THz modulators with broadband modulation response.

The dynamic modulation experiment, as given in figures 5(a) and (b), respectively, before and after annealing of MoS2-Si samples in the presence of modulated laser beam at 0.1 Hz frequency. The change in the THz photocurrent is higher in annealed-MoS2-Si samples in comparison to MoS2-Si samples under the same incident laser power. This suggests more THz attenuation in the annealed-MoS2-Si samples with pumping power. The modulation speed is also an important parameter to characterize the performance of the THz modulators. The modulation speed typically depends on the rise time and fall time of the device. The rise time is determined as the time interval taken for 10% to 90% of the maximum THz photocurrent value, and the fall time is determined as the time interval taken for 90% to 10% of the maximum THz photocurrent value. The enlarged view of the response of the MoS2-Si and annealed-MoS2-Si samples at 0.1 THz frequency to the laser beam modulated at 1 kHz frequency is shown in figures 5(c) and (d). The rise time and fall time are calculated as 76 μs, and 130 μs for MoS2-Si samples, respectively. While in the case of annealed-MoS2-Si samples, the obtained rise time and the fall time are 20 μs, and 22.7 μs, respectively, which is comparable to other reported TMDs based THz modulator [39–41]. The steep rise and fall edges of the photocurrent response entail the fast response of the THz modulator.

To further examine the behavior of the MoS2-Si modulator, the numerical calculations have been carried out using the Lumerical software tool based on the FDTD method. The MoS2 layer having 0.6 nm thickness is considered on 5 μm Si acting as a photoconductive layer. The maximum number of photogenerated carriers are
generated in the bulk Si substrate, not in the nanometer-thick MoS$_2$ layer. This brings a change in the conductivity of Si to a great extent with pumping laser power. This conductivity change is equivalently replaced with the number of charge carriers induced in Si after optical excitation. The photogenerated carriers produced in the silicon substrate per unit area per unit time is

\[ n = \frac{P(1 - R)\alpha\tau}{\hbar v d} \]  

(3)

Where P is the optical pumping power, \( \alpha \) is responsivity as 20\%, R is reflection losses as 30\%, \( \tau \) is carrier lifetime as 10 \( \mu \)s, \( v \) is laser frequency, and d is the photoconductive silicon thickness. The obtained number of charge carriers after calculations is, \( n = 1.12 \times 10^{16} \) cm\(^{-1} \).

The conductivity of the silicon substrate is

\[ \sigma = n(\mu_p + \mu_n) \]  

(4)

Where \( n \) is the electron charge, \( \mu_p \) and \( \mu_n \) are hole and electron mobilities respectively. The calculated value of conductivity for silicon is 351.5 Sm\(^{-1} \), equivalent to 1 W laser power. The pumping laser power is equivalently replaced with this change in conductivity of the Si for carrying out simulations. We have considered the conductivity of Si substrate as 80 S m\(^{-1} \), 150 S m\(^{-1} \), 250 S m\(^{-1} \), and 400 S m\(^{-1} \) for pumping power of 0.25 W cm\(^{-2} \), 0.5 W cm\(^{-2} \), 0.75 W cm\(^{-2} \), and 1 W cm\(^{-2} \) respectively after doing numerical calculations and series of further simulations \(^{18, 42} \). It is found that the photogenerated carrier density is higher near the MoS$_2$-Si interface than bare silicon, as presented in figures 6(a) and (b). It suggests higher concentration of charge carriers as the built-in electric field is high at the interface of MoS$_2$ and Si. This leads to accomplish higher modulation depth due to the significant change in conductivity. The modulation depth is simulated for 0.4 THz and 0.9 THz frequencies which matches well with the experimental results.

The normalized electric field profile is given in figure 7. It is observed that the electric field profile is changing with the change in pumping power due to a change in conductivity. There is more change in the electric field.
file with pumping power at a lower frequency, i.e., 0.4 THz in comparison to the higher frequency, i.e., 0.9 THz. This is owing to more change in THz transmittance and conductivity for lower THz frequencies.

4. Discussion

In the case of optically driven THz modulators, the change in conductivity of semiconductors with the incidence of optical laser power brings variation in the THz reflection and transmission and hence the modulation. In the silicon substrate, the extra charge carriers are induced after illuminated by CW laser, and these carriers affect the conductivity of the substrate. But the variation in the conductivity is small in bare Si substrate because these carriers recombine quickly due to long carrier lifetime and low mobility in Si as compared to GaN, and GaAs substrate. Whereas in the case of the MoS2-Si sample, MoS2 forms a heterostructure with Si [43]. Because of the potential barrier and built-in electric field at the interface of MoS2 and Si, there is a separation of charge carriers, which reduces the recombination rate. Thus, there is more change in the conductivity of the MoS2-Si sample as compared to bare Si. But the small difference in Fermi energy level, as shown in figure 8(b) leads to drift fewer number of charge carriers due to the small potential barrier and electric field. After annealing, MoS2 becomes p-type, and the large number of holes generated in MoS2 leads to move Fermi energy level near to valence band maximum [30, 44]. The significant difference in Fermi level induces a large built-in electric field causes to drift more number of charge carriers. And the difference in concentration of charge carriers also leads to diffusion of carriers on both silicon and MoS2 side. This separation of charge carriers contributes to more changes in conductivity and improves the modulation efficiency. Hence, the heterostructure formed with a thin MoS2 film, and Si enables more change in carrier density near the interface and hence improves modulation of THz waves. The energy band diagram before and after annealing of the MoS2-Si sample is depicted in figure 8. Among various reported TMDs based modulators, modulators described in this work, based on bilayer MoS2 have the highest modulation performance under low optical power excitation of 1 W cm$^{-2}$. The comparison of different reported THz modulators based on TMDs is given in table 1.

Figure 5. The measured THz photocurrent at 0.1 THz frequency of (a) MoS2-Si and (b) annealed-MoS2-Si samples irradiated by laser pulse at 1 Hz frequency. The enlarged view of the detected photocurrent for (c) MoS2-Si and (d) annealed-MoS2-Si samples at 0.1 THz frequency to the laser beam modulated at 1 kHz under 1 W cm$^{-2}$ optical power.
Figure 6. The simulated carrier generation rate at (a) 0.4 THz and (b) 0.9 THz frequency. The experimental and simulated modulation depth at (c) 0.4 THz and (d) 0.9 THz frequency.

Figure 7. The simulated normalized electric field at 0.4 THz frequency for (a) 0.1 W cm$^{-2}$ and (b) 1 W cm$^{-2}$ optical power and for (c) 0.1 W cm$^{-2}$ and (d) 1 W cm$^{-2}$ at 0.9 THz frequency.
5. Conclusion

In conclusion, we have experimentally manifested that uniform thin film of MoS$_2$ on silicon can be utilized to manipulate and control the amplitude of THz waves. The annealing process of MoS$_2$ film on silicon further improves the modulation performance. The maximum modulation depth of 81.4% at 0.1 THz and 79.7% at 0.9 THz frequency are achieved after annealing under 1 W cm$^{-2}$. The FDTD simulation results have shown that photogenerated charge carrier density and modulation depth are higher in MoS$_2$-Si samples in comparison to bare silicon. The obtained experimental results of our modulator have exhibited excellent performance, which is comparable to other reported TMDs based THz modulators. This work suggests the applicability of MoS$_2$ and other TMD materials for the development of different efficient and high performing THz components.

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![Figure 8. Schematics of the energy band diagram of (a) MoS$_2$ on Si before contact, (b) MoS$_2$ on Si after contact, and (c) After annealing of MoS$_2$ on Si sample.](image-url)
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**Data availability statement**

All data that support the findings of this study are included within the article (and any supplementary files).

**Appendix 1**

The numerical calculations have been carried out using the Numerical software tool based on the FDTD method. The silicon substrate having 5 μm thickness as the photoconductive layer and refractive index of 3.4 is considered. The thickness of the MoS2 film is 0.6 nm and conductivity assumed to be around 80 Sm⁻¹. The perfectly matched layer (PML) boundary conditions are used in all three axis to minimize the reflections at the boundaries. A small mesh grid size is used for MoS2 film to get higher accuracy. The mesh grid size is 0.1 nm in X and Y-axis and 0.05 nm in Z-axis direction. The plane wave source of frequency from 0.1 to 1 THz is used.

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