Recent advances of terahertz (THz) science and technology are witnessing the rapid development of disruptive applications in THz wireless communication and radars, intelligent sensing, and imaging among others. However, the lack of sufficiently multifunctional devices, such as THz modulators, is impeding the proliferation of THz applications, expecting breakthroughs in new materials, structures, and phenomena. Herein, for the first time, anomalously enhanced THz transmission phenomenon in vertically standing molybdenum disulfide (MoS$_2$) nanoplates integrated onto silicon (Si) substrates under active tailoring of photoheating and photodoping is reported. The formation of MoS$_2$/Si heterojunction enables a heat-induced broadband amplitude improvement exceeding 10%, while photodoping reduces the THz absorption by ≈70% in the measurement frequency region of 0.2–2.0 THz. Through systematically discriminating the transmitted and reflected THz responses from MoS$_2$/Si heterojunction and those from Si substrates, the photothermally decreased THz conductivity can be attributed to the scattering effects, while photodoping-induced enhancement originates from the decreased carrier density. Further verification experiments are implemented on a 100 GHz video imaging system, and the THz transmission spots become brighter under both stimuli. The observations not only enable an impellent understanding of low-dimensional light–matter interactions but also provide possible routes for developing novel, integrated, multifunctional THz optoelectronic devices.

1. Introduction

The rapid development of ultrafast laser technology has witnessed the boosting progress of terahertz (THz) science and technology, covering spectroscopy,[11] imaging,[15] (bio)sensing,[3,4] and most recently, high-speed wireless communication.[5,6] These researches have spurred massive strides in the growth of essential THz components, such as efficient THz sources,[7–12] highly sensitive detectors,[13,14] as well as flexible multifunctional devices.[15,16]

Toward future faster applications, THz functional components are meant to take advantage of those materials that can respond at a high speed, especially for THz wave modulation.[17,18] Conventional THz modulation materials concentrate on semiconductors, metamaterials, phase transition materials, magnetic materials, strain materials, and their compounds.[19–23] In parallel to these recent advances at the intersection of THz and material sciences, there have been major progress in 2D materials.[24] For instance, tunable perfect THz absorbers were demonstrated with near-zero reflection using integrated metallic gratings into the classical Salisbury screen structure based on graphene.[25] Highly tunable solid-state graphene/quartz modulators based on the effect of tunable Brewster angle were successfully developed.[26] Besides, various transition metal dichalcogenides (TMDs) (WS$_2$, WSe$_2$, and MoSe$_2$, and so on) have also revolutionized the field of THz modulation with their unusual physical properties. Among all of them, molybdenum disulfide
employing MoS$_2$ incorporating with classic semiconductors has nontrivially tunable bandgaps (a direct bandgap of 1.8 eV for monolayer, while an indirect bandgap of 1.3 eV for bulk)\textsuperscript{[27]} More importantly, the light absorption of MoS$_2$ spans widely on a large scale, which can be regulated from visible to the near-infrared spectral region (350–950 nm).\textsuperscript{[28]} All these unique properties feature in a well-defined boundary between its on and off states, which could be altered via the common methods, such as optical and thermal means, and thus are beneficial for THz devices.\textsuperscript{[29]} While MoS$_2$ has been considered as a promising candidate for THz modulators, more interesting observations are expected in the light-driven interaction between THz wave and MoS$_2$, such as the attenuated THz conductivity induced transmission enhancement effect. From the phenomenological point of view, the attenuated conductivity can indicate the conductivity of a material is lowered under external excitations. This means the material is to release the part of THz energy absorbed by itself, which will, in turn, enhance the THz transmission signal while the reflection remains almost unvaried. Actually, there has been a fraction of signs about probing the THz conductivity attenuation in some novel quantum materials, 2D materials, and even in monolayer MoS$_2$\textsuperscript{[10–32]} However, such experiments require either ultrafast time-resolved methods or else low-temperature circumstances. For practical applications, on one hand, operation under normal ambient conditions is of great relevance. These harsh conditions are hard to be satisfied simultaneously due to the high cost as well as complicated configurations. And external excitations could further influence the component performances, such as modulation depths, gain efficiency, response time as well as operation bandwidth. On the other hand, to further optimize the device performances, employing MoS$_2$ incorporating with classic semiconductors has gained more attention in recent years, as the constructed heterojunction structures often obtain excellent performances by improving band energy alignment and interface properties.\textsuperscript{[13]} Therefore, an available measurement method, as well as an effective structure design, is highly essential to yield deep insights into the performance of TMDs-based THz functional devices.

Here we observe, for the first time, the two anomalous THz transmission behaviors in MoS$_2$/Si heterojunction via a straightforward optical pump-THz probe technique at room temperature. Optically driving out of equilibrium with a continuous-wave (CW) laser results in the nonlinear THz amplitude modulation. By photothermal pumping, the THz transmission through the MoS$_2$/Si heterojunction increases which can be ascribed to the surface conductivity attenuation. We also observe that the heterojunction can hinder the all-optical THz modulation, which is opposite from the previous optical pumping reports. Experimental results from optical pump-THz video imaging reproduce the spectroscopic evolution process, explicitly corroborating that MoS$_2$/Si heterojunction could act as a promising device structure for THz modulation and even in the next-generation THz functional devices.

2. Results and Discussions

2.1. Anomalously Enhanced THz Transmission Induced by Heat

To investigate the THz conductive responses in MoS$_2$/Si heterojunction, a custom-designed angle-resolved THz time-domain spectroscopy (THz-TDS) is integrated with a CW modulating laser (wavelength of 808 nm, oblique incidence angle of 45°) as an external stimulus. Compared with the conventional time-resolved femtosecond laser pulse pump-THz probe method, CW laser pumping directly prepares the material to the excited states, which are deposited onto the transmitted THz probing signals. This method is more suitable for the research of application-oriented materials and structures. The THz pulses, generated and coherently detected by a pair of InGaAs photoconductive antennas driven by a femtosecond fiber laser, are normally incident onto the samples, as illustrated in Figure S2a, Supporting Information. For light-induced heat modulation, the modulating laser irradiates first onto a piece of iron. Along with the temperature increase on the iron (highest temperature of $\approx$180°C for 54 W cm$^{-2}$), the heat flows to samples, resulting in indirect heat excitation for the samples, as illustrated in Figure 1a. To ensure that the film is not damaged under such high-energy infrared continuous laser, we increase the continuous laser power step by step. We fix the delay line at the THz peak position and observe the THz signal value as well as the sample variation at each pumping power. THz signal value basically has not changed, and the sample shows no obvious damage.

Under this circumstance, we fix the modulating laser fluence at 54 W cm$^{-2}$, and record the transmitted THz pulses from the MoS$_2$/Si sample with and without photothermal excitations. As exhibited in Figure 1b, an enhanced THz signal is obviously probed through the heat excitation. To evidence this improved temporal modulation, we subtract the unheated sample waveform from the heated sample waveform and obtain a boosted differential curve ($\Delta E$). From this curve, we can see that neighboring spectral weights near the main pulse peak are positive, which unambiguously manifests that the light-induced heating leads to the increase of THz transmission. To further investigate the anomalous transmission behavior, we conduct THz transmission and reflection measurements under 30° oblique incidence via light-induced heating (see Figure 2b and S4, Supporting Information). Reflected THz signals are basically invariant while transmitted THz signals are obviously improved, further manifesting the experimental validity of the reduced THz absorption. To check whether this anomalous phenomenon is served for broad- or narrow-band, we transform time-domain signals to frequency-domain amplitude spectra. As plotted in Figure 1c, these spectral distributions clearly indicate that the THz signal amplification is a broadband behavior, covering the whole frequency range (0.2–2.0 THz) of the measurement system.

2.2. Heating Power Dependence

To further systematically investigate the influence of the heating power on anomalous THz transmission, we adjust the
modulating laser power to control the heating impact on samples. When we increase the modulating pump fluence, nonmonotonic behaviors are observed in both MoS$_2$/Si heterojunction and Si samples, as respectively, depicted in Figure 2a,b. To distinctly observe this interesting behavior, the extracted THz peak amplitudes as a function of the heat power are plotted in Figure 2c. In both cases, anomalously enhanced THz transmission tendencies are induced by continually increasing the heat power till $\approx 13$ W cm$^{-2}$. Below this critical fluence, the enhanced THz transmission disappears. There is an exceptional point of $\approx 2.5$ W cm$^{-2}$, below which the THz peak amplitudes reduce as the heat power increases, while above it, the THz peak signals follow a monotonously increasing tendency. In the case of the Si substrate, the THz enhancement saturates at $\approx 7.5\%$, defined as a modulation depth, where denotes the modulated THz peak signal, while presents the THz peak signal without any external stimulus. However, for MoS$_2$/Si heterojunction, the enhanced THz transmission behavior keeps a monotonic linear increasing relationship, and there is no obvious saturation tendency. The maximum modulation depth for MoS$_2$/Si is $\approx 12.5\%$ in our experiments due to the limitation of the modulating laser power. Furthermore, another special pump fluence appears at $\approx 34$ W cm$^{-2}$, where the competitively enhanced THz transmission behaviors of the two samples are reversed. Below this point, the phenomenon in Si is more distinct than that from MoS$_2$/Si heterojunction, and vice versa. In the static measurements, the maximum value of the THz electric field for the MoS$_2$/Si heterojunction is $\approx 2\%$ smaller than that of the Si substrate, as shown in Figure S3a, Supporting Information. This indicates that MoS$_2$/Si heterojunction attenuates more THz energy than the pure Si substrate due to the enhanced absorption of MoS$_2$ plates. Upon increasing the heat power, THz enhancement behaviors appear in both samples. However, under weak heat power, extra THz attenuation of MoS$_2$ plates cannot be neglected in MoS$_2$/Si heterojunction. This effect induces a weaker transmission than the Si substrate. Under strong heat power closing to the switching point at 34 W cm$^{-2}$, the THz transmission enhancement effect in MoS$_2$/Si heterojunction becomes dominant.

Another intuitive phenomenon is that for the case of heat driving 100 GHz real-time imaging evolution at corresponding

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**Figure 1.** Anomalous THz transmission in MoS$_2$/Si heterojunction. a) Schematic diagram of the enhanced THz transmission behavior in MoS$_2$/Si heterojunction under photothermal excitations. The vertically standing MoS$_2$ nanoplates (with a thickness of $\approx 50$ nm) were integrated onto n-type Si substrates (thickness of $\approx 500$ μm, area of $1 \times 1$ cm$^2$) with a 1.2 nm thick native SiO$_2$ layer. b) Normalized THz temporal waveforms transmitted from MoS$_2$/Si heterojunction without (gray lines) and with photothermal excitations at 54 W cm$^{-2}$ (red lines), respectively. The bottom blue curve exhibits the differential THz electric field. c) Corresponding THz spectra manifest a broadband-enhanced transmission behavior.
modulating pump fluences. As illustrated in Figure 2d–i, when the photogenerated heat is injected into our samples, THz transmission is enhanced in MoS\textsubscript{2}/Si heterojunction as well as in Si. Compared with Si (Figure 2g–i), a slightly higher enhancement is presented in MoS\textsubscript{2}/Si heterojunction (Figure 2d–f). Figure 2j illustrates the overall brightness contrast obtained by image processing. It can be seen from these data that the anomalous transmission phenomenon can still be observed for 100 GHz electromagnetic waves in the case of photothermal excitation. Those visual results are expected to reproduce the spectroscopy feature by the frequency shift of anomalous transmission enhancement, also indicating the potential THz imaging applications.

2.3. Light-Induced THz Enhancement in MoS\textsubscript{2}/Si Heterojunction

Encouraged by the results of heat-induced anomalously enhanced transmission, we continually study the light-induced effects in the MoS\textsubscript{2}/Si heterojunction as well as in the Si substrate. The normally weakened transmission appears in two samples. The THz temporal profiles of MoS\textsubscript{2}/Si heterojunction (Figure 3a) decay slowly while the results of the Si substrate (Figure 3b) fade away promptly upon initiating the value of the pump fluence. In Figure 3c, the THz peak amplitudes are also extracted to further elucidate the observed THz transmission varying with the modulating pump fluence. Under the equilibrium state of 0 W cm\textsuperscript{-2}, the THz peak amplitude of MoS\textsubscript{2}/Si heterojunction is slightly lower than that of the Si substrate. However, when we turn on the modulating laser and keep increasing its pump fluences, different modulation behaviors emerge between the Si substrate and MoS\textsubscript{2}/Si heterojunction, consequently up to >70% difference of modulation depths at the modulating fluence of \( \approx 20 \) W cm\textsuperscript{-2}.

Analogous to the measurements under heat excitations, more explicit images are also captured to observe THz transmission enhancement of MoS\textsubscript{2}/Si heterojunction under the light forcing...
(Figure 3d–i). The brighter THz spots of MoS₂/Si heterojunction (Figure 3d–f) unambiguously enable the higher antireflection than that of the pure Si substrate (Figure 3g–i) at each pump fluence. The overall image weights are drawn in Figure 3j, evidently indicating that it is also effective to extend the transmission enhancement behavior to 100 GHz under direct photoexcitation. This demonstrates an efficient modulation control of MoS₂/Si heterojunction.

3. THz Transmission Enhancement Effect

The physical origin of the two intriguing effects can be understood by considering the variation of the system conductivity. The detailed interpretations of photoheating and photodoping behaviors in both the Si substrate and the MoS₂/Si heterojunction are exhibited in the following parts:

First, we focus on explaining the enhanced THz transmission behavior in Si substrates under photothermal excitation, which can be safely attributed to carrier scattering. When extrinsic heat spreads to the n-type Si substrate, the carrier concentration may increase appropriately by initiating a lower heat power, which induces the increase of the conductivity, and THz transmission decreases corresponding to the dip of the THz peak amplitude at 2.5 W cm⁻² as shown in Figure 2c. However, thermally induced increase of carrier density is restricted, heat excitation would not produce a sustained scaling-up carrier concentration. Upon continually increasing the heat power, the carrier scattering behavior may also contribute to the enhanced THz transmission signal. Normally, the thermal effect can distinctly influence the carrier transition by incorporating the scattering process in doped materials, which is explained using an established Drude model (see S8, Supporting Information). When an additional thermal field is exerted, the lattice periodic potential field is disturbed, and the carriers are scattered. Therefore, the effective charge carrier density decreases as well as the average scattering rate of the carriers increases. This possibility is supported by the experimental results of intrinsic high-resistivity Si under the same photothermal conditions (see S6, Supporting Information). Photoheating excited refractive index results illustrated in S7, Supporting Information, can further support this lattice periodic potential field transformation. As a result, the conductivity decreases as the scattering effect enhances, resulting in experimentally observed enhanced THz transmission phenomenon.
Based on the illustrations of the conductivity variation in Si substrates, the physical origin of the enhanced conductivity attenuation in MoS2/Si heterojunction can be further extrapolated. The introduction of MoS2 to Si substrates can induce stronger scattering effects, leading to the reduced conductivity in MoS2/Si heterojunction. For a more comprehensive quantitative analysis, the frequency-dependent conductivities of the MoS2/Si heterojunction as well as that of the Si substrate with varying pump fluences are extracted following the detailed description in Methods and are plotted in Figure 4. It should be noted that the conductivity of MoS2/Si heterojunction (Figure 4a), as well as that of the Si substrate (Figure 4c), are sensitive to the pump fluence in the frequency range from 0.2 to 0.7 THz, while the conductivities of various pump fluences are extracted and fitted with the Drude model (see S8, Supporting Information). As illustrated in Figure 5, the frequency-dependent conductivity of MoS2/Si heterojunction (Figure 5a) shows weaker sensitivity to pump fluences than the Si substrate (Figure 5c). Therefore, the conductivities of the MoS2/Si heterojunction increase more slowly upon the increase of the pump fluence at specific frequencies of 0.5, 0.7, and 0.9 THz, as shown in Figure 5b,d.

Next, we try to shed light on the THz enhancement effect in the MoS2/Si heterojunction under the case of direct photocarrier injection. For the Si substrate, photoexcited carriers are the main factor for the conductivity promotion, which degrades the THz transmission drastically. Normally, if the Si substrate is integrated with 2D materials, this kind of heterojunction can further increase the modulation depths (degrades the THz transmission further). This phenomenon does not exist in our experiments. Although, the light pumping induces that the conductivity of the sample increases and THz transmission decreases. The decrease extent is rather low because the thick MoS2 nanoplates absorb most part of the pumping energy, and the carriers are almost generated in MoS2. The Si could not provide electrons and holes due to the 1.2 nm SiO2 barrier.

Like the photothermal quantitative analysis method, the conductivity for the MoS2/Si heterojunction as well as that of the Si substrate under various pump fluences is extracted and fitted with the Drude model (see S8, Supporting Information). As illustrated in Figure 5, the frequency-dependent conductivity of MoS2/Si heterojunction (Figure 5a) shows weaker sensitivity to pump fluences than the Si substrate (Figure 5c). Therefore, the conductivities of the MoS2/Si heterojunction increase more slowly upon the increase of the pump fluence at specific frequencies of 0.5, 0.7, and 0.9 THz, as shown in Figure 5b,d.
4. Conclusion

We successfully demonstrated actively tunable THz enhancements in MoS2/Si heterojunction via externally applied CW modulating laser. Through comprehensive comparative investigations on the photothermal effect induced enhanced THz transmission responses between MoS2/Si heterojunction and Si substrates, we observe boosted THz conductivity attenuation in MoS2-based heterostructures. Further comparative investigations on direct photocarrier injection in MoS2/Si heterojunction and Si substrates unveil that the existence of MoS2 can obviously reduce the THz absorption. The principle confirmatory imaging application experiments manifest that the photothermal conductivity attenuation effect and the optical-induced THz absorption reduction effect observed here are expected to develop some potential application scenarios. For example, high-resistance Si is a kind of commonly used material in some THz optical systems. One of its functions is optical filter, which can stop pumping light but let THz waves pass through. However, when optical pump power gets stronger, the photogenerated carriers in Si from two/multi-absorption effects would apparently decrease the THz transmission. This proposed MoS2/Si heterojunction can attenuate the optical pump light and promise high THz transmission. Our findings suggest that efficient enhancement conversion of THz signals pave the way for exploiting Si/TMDs heterojunction toward conceivable applications such as future THz wireless communications requiring accessible, engineerable, and integrable THz modulators and other multifunctional devices.

5. Experimental Section

Sample Fabrication and Characterization: The vertically standing MoS2 nanoplates were prepared on the n-type Si (≈500 μm thick) substrate to form the n-MoS2/n-Si homotype heterojunction using magnetron sputtering. The resistivity of the Si substrate is 1–10 Ω cm (room temperature), and a 1.2 nm thick SiO2 layer was naturally produced on the surface. Before the deposition of MoS2 nanoplates, the wafer was carefully cleaned based on standard cleaning techniques to remove ions and organic impurities. A composite MoS2 target (99.998%) was selected as the sputtering source. During the preparation process, the reaction chamber was first evacuated to 1.0 × 10^5 Pa, and the substrate temperature was raised to 500 °C, then the argon gas was introduced by maintaining a 25 sccm flow rate and a 1.0 Pa pressure. Finally, the ≈50 nm MoS2 nanoplates were prepared after adding the sputtering power of 50 W and keeping a 2.0 nm min^-1 deposition rate for about 25 min.

CW Laser Pumping-THz Probing Experimental Setup: The measurements of the photoheating and photodoping THz responses were performed on a custom-designed angle-dependent THz-TDS coupled with CW pumping laser in both transmission and reflection geometries. The schematic diagram is shown in Figure S2a, Supporting Information. Ultrafast THz pulses were acquired with a 33.3 fs time-resolution over a 20 ps time-window by varying the time delay between the THz signal and the optical probing pulses. For each measurement, multiple time-domain traces were collected and averaged. The described THz experimental setup provided a 60 dB signal-to-noise ratio over a 2 THz noise-equivalent-power.
bandwidth. All measurements were performed at room temperature without purging or pumping the system.

100 GHz Video Imaging Experiments: To distinctly investigate the anomalously enhanced THz transmission phenomenon, THz spot images were recorded under external excitations by employing a self-assembled 100 GHz video imaging system as shown in Figure S2b, Supporting Information. During the experiment, the commercialized 100 GHz (TeraSense) signal was first collimated and then focused normally onto the samples, while the CW modulating laser had a tilting incidence angle of $\approx$45$^\circ$. The modulated 100 GHz signals were collimated and then imaged onto a THz camera (TeraSense, 32 x 32 pixels). Upon varying the modulating fluence, the influenced 100 GHz images were recorded at ambient atmosphere.

Extraction of THz Conductivity: The THz time-domain electric field transmitting through the sample as $E_{\text{sam}}(\omega)$, and that of the reference signal as $E_{\text{ref}}(\omega)$ was determined. Using fast Fourier transform (FFT), THz frequency-domain electric fields, $E_{\text{sam}}(\omega)$ and $E_{\text{ref}}(\omega)$, can be obtained. The complex transmission ratio $T(\omega)$ can be calculated as \[ T(\omega) = \frac{A(\omega)e^{i\phi(\omega)}}{E_{\text{ref}}(\omega)} = \frac{4\varepsilon_0\varepsilon_\parallel(\omega)}{\varepsilon_0(\omega) + 1} e^{i\phi(\omega)} \] where $A(\omega)$ denotes the amplitude of the transmitted electric field, $\phi(\omega)$ presents the phase of the transmitted electric field, $\varepsilon_\parallel(\omega)$ is the complex refractive index, $d$ is the sample thickness, and $c$ is the speed of light (in vacuum). The MoS$_2$ nanosheets on the Si substrate, forming a heterostructure was treated as an equivalent medium. The complex refractive index of the sample can be calculated following these equations

$$\varepsilon_\parallel(\omega) = n_\parallel(\omega) + i\kappa_\parallel(\omega)$$

$$n_\parallel(\omega) = 1 + \frac{\phi(\omega)c}{\omega d}$$

$$\kappa_\parallel(\omega) = \frac{\omega d}{c} \ln \left( \left( \frac{n_\parallel(\omega) + 1}{4\varepsilon_0\varepsilon_\parallel(\omega)} \right) A(\omega) \right)$$

$$\alpha_\parallel(\omega) = \frac{2\omega\kappa_\parallel(\omega)}{c}$$

where $n_\parallel(\omega)$ is the refractive index, $\kappa_\parallel(\omega)$ denotes the extinction coefficient, $\alpha_\parallel(\omega)$ presents the absorption coefficient, and the imaginary part of the dielectric constant is expressed as

$$\varepsilon_\parallel(\omega) = 2n_\parallel(\omega)\kappa_\parallel(\omega)$$

The real part of the complex conductivity reflects the material electric conduction capability, and it can be calculated with the following relationship from the imaginary part of the dielectric constant

$$\sigma_\parallel(\omega) = \varepsilon_\parallel(\omega)\varepsilon_0\omega$$

where $\varepsilon_0$ is the dielectric constant (in vacuum). Hence, the real part of the conductivity can be directly calculated with a complex refractive index

$$\sigma_\parallel(\omega) = 2n_\parallel(\omega)\kappa_\parallel(\omega)\varepsilon_0\omega$$

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

anomalous THz transmission, molybdenum disulfide, THz modulators

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