

Research Article

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All-optical modulation based on MoS₂-Plasmonic nanoslit hybrid structures

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Abstract: Two-dimensional (2D) materials with excellent optical properties and complementary metal-oxide-semiconductor (CMOS) compatibility have promising application prospects for developing highly efficient, small-scale all-optical modulators. However, due to the weak nonlinear light-material interaction, high power density and large contact area are usually required, resulting in low light modulation efficiency. In addition, the use of such large-band-gap materials limits the modulation wavelength. In this study, we propose an all-optical modulator integrated Si waveguide and single-layer MoS₂ with a plasmonic nanoslit, wherein modulation and signal light beams are converted into plasmon through nanoslit confinement and together are strongly coupled to 2D MoS₂. This enables MoS₂ to absorb signal light with photon energies less than the bandgap, thereby achieving high-efficiency amplitude modulation at 1550 nm. As a result, the modulation efficiency of the device is up to 0.41 dB μm^{-1} , and the effective size is only 9.7 μm . Compared with other 2D material-based all-optical modulators, this fabricated device exhibits excellent light modulation efficiency with a micron-level size, which is potential in small-scale optical modulators and chip-integration applications. Moreover, the MoS₂-plasmonic nanoslit modulator also provides an opportunity for TMDs in the application of infrared optoelectronics.

1 Introduction

Photonic chip integration is an indispensable technology for information processing and communication. Critical components of this technology, such as lasers, detectors, and modulators, have been gaining increasing attention [1–3]. Among them, the all-optical modulators can completely convert and modulate signals in the optical domain which are essential for on-chip interconnection, short-distance communications [4, 5]. To implement their use in such applications, smaller-sized all-optical modulators are required, while sufficient modulation depth should be maintained. However, the all-optical devices based on traditional three-dimensional semiconductors, such as Si [6], lithium niobate [7], and III–V compound semiconductors [8], are usually limited in size to a few millimeters or centimeters due to the inherent bulk material properties.

Owing to the atomic-level thickness, CMOS compatibility, and excellent optoelectronic properties, two-dimensional (2D) material-based hybrid nanophotonic components are a promising platform for realizing integrated optoelectronic or photonic devices. Among the emerging possibilities, all-optical modulators based on 2D materials have shown great potential [9–12]. In recent years, all-optical modulation with 2D materials such as graphene [13–19], transition metal dichalcogenides (TMDs) [20–23], and black phosphorous [24, 25] have been successively demonstrated based on nonlinear light saturation absorption effect [26, 27]. However, due to the low optical absorption of 2D materials arising from their atomic thickness, a high-power density and large light-matter interaction area are required to ensure that the device can achieve sufficiently large modulation depths (2–17 dB) [14–25], but that limits the modulation efficiency (only 10^{-2} – 10^{-4} dB μm^{-1}). Furthermore, the strong light-matter interaction in most 2D materials (i.e., TMDs) lies in the visible range, restricting the applications in optical

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communication wavelength bands [28]. Recently, TMDs that exceed the bandgap to achieve optical modulation have been individually reported owing to sub-bandgap transition [29, 30] caused by crystallographic defects or edge states [11, 25]. However, in such cases, the low transition efficiency results in insufficient light absorption and low modulation efficiency. A solution may be found in plasmonic nanoslit [31–34] which squeezes the light into a narrow mode with high electromagnetic confinement, not only ensures strong coupling between light and 2D materials but also compresses the device volume. In the previous work of graphene-slit waveguides, the modulation efficiency has been initially improved [34].

Herein, we fabricated an all-optical modulator with a monolayer MoS₂ and a plasmonic nanoslit hybrid structure. The strong field confinement at the nanoslit, for both the signal and modulation lights, is used to enhance the light–matter interaction dramatically and extend the operating wavelength range. With an effective size of only 9.7 μm , is successfully fabricated. Experimentally, the 532-nm modulation light facilitates effective control of the 1550 nm signal light, and the modulation efficiency is 0.41 dB μm^{-1} . This study expands the application of 2D TMDs materials with a large intrinsic bandgap in infrared optoelectronics, offering an opportunity for developing high-efficiency all-optical modulators based on 2D TMDs materials.

2 Main text

A schematic diagram of the MoS₂-plasmonic nanoslit all-optical modulator is shown in Figure 1a. The signal light (1550 nm) in the fiber is coupled to the Si waveguide and then converted to surface plasmon polaritons (SPPs) in the Au nanoslit due to the strong field confinement. The monolayer MoS₂, as a light-absorbing layer, is transferred to the surface of nanoslit to realize the strong interaction with the excited SPPs. Different from signal light, the modulation light (532 nm laser) is illuminated onto the MoS₂-plasmonic nanoslit hybrid structure in free space. Figure 1b shows the working mechanism of the proposed all-optical modulator. The off and on state of modulation light is depicted by the blue curve, while the corresponding changes of the signal light are shown in the red curve. When the 532 nm modulation light is in the OFF state, the signal light SPP cannot be absorbed by MoS₂ because its energy is less than the MoS₂ band gap (Figure 1b OFF state and Figure S1a). However, in the ON state of the modulation light, a large number of electron-hole pairs can be

generated in MoS₂ (Figure 1b, ON state, i). The excited electrons can further interact with the signal light SPP and undergo an intraband transition (as shown in Figure 1b, ON state, ii) [35, 36]. As a result, the output intensity of signal light can be modulated effectively by switching the 532 nm laser.

According to the intraband transition theory and plasmon effect [37], the light absorption coefficient of the signal light SPP can be expressed as

$$\alpha = \frac{ne^2}{m^* \eta c \omega^2 \epsilon_0 \tau}. \quad (1)$$

where m^* is the effective mass of the electron, η is the refractive index of the MoS₂ film, ϵ_0 is the vacuum dielectric constant, ω is the signal light angular frequency, τ is the electron relaxation time, e is the electron charge, and n is the carrier concentration (see Supplementary Note 2 for detailed formula derivation).

According to Equation (1), the absorption coefficient α is proportional to the square of the signal light wavelength and the carrier concentration. The carrier concentration depends on the intensity of the signal and modulation light. By changing the intensity of the modulation light, the light absorption coefficient of MoS₂ for the signal light SPP can be changed. Therefore, the transmission output of the signal light can be modulated by controlling the intensity of modulation light.

The influence of nanoslit width on the signal and modulation light confined fields was analyzed by the finite element analysis method. Figure 1c depicts the cross-sectional view of the MoS₂-plasmonic nanoslit structure in the yz direction. The plasmonic nanoslit is composed of Au and Si and has a width of W . In our previous study, this design meant that the refractive indexes of the upper and lower layers of the Au nanoslit were symmetrical (both the upper and lower layers are air), enabling the electromagnetic field to be better confined in the nanoslit [34]. Figure 1d shows the electric field distribution at the signal light in the nanoslit, which corresponds to the schematic structural diagram of Figure 1c (the white dashed line marks the position of the Au layer). The confined electric fields of the signal SPP are mainly distributed at the surface of Au nanoslit, which ensures that the MoS₂ can sufficiently interact with the electromagnetic field. Another beam of modulation light irradiates the nanoslit in the xy plane vertically, generating strong LSP resonance [38, 39], which further enhances the local electric field of the modulation light at the nanoslit (Figure 1e).

The electric field energy at the contact interface for the plasmon formed by the two beams of light and MoS₂ is plotted as a function of slit width in Figure 1f (electric field

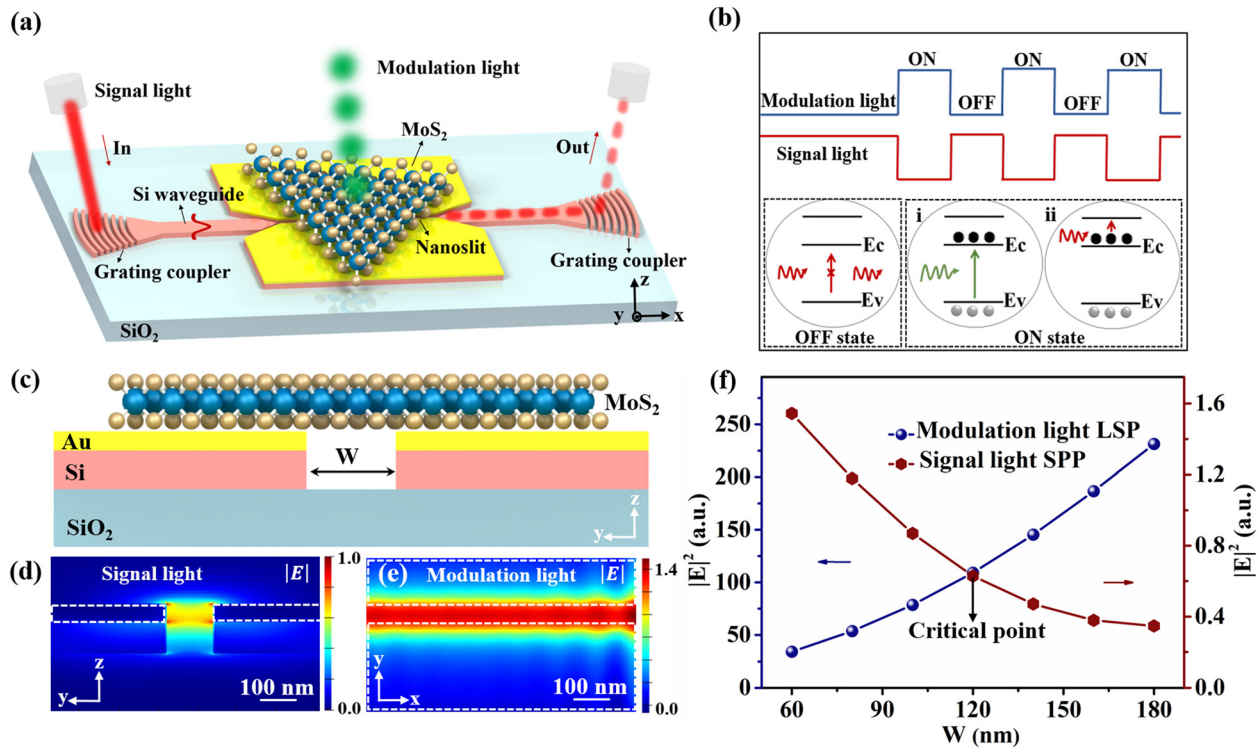


Figure 1: MoS₂-plasmonic nanoslit all-optical modulation.

(a) Schematic diagram of the device structure. (b) Working principle of the all-optical modulation. (c) Cross-sectional view of (a) (in yz plane). (d)–(e) Confined electric field distribution of signal and modulation lights. The slot width is 120 nm in each case, and the Au and Si layer thicknesses are 80 and 140 nm, respectively. Scale bar, 100 nm. The wavelengths of the signal and modulation lights are set to 1550 and 532 nm, respectively. And the TE mode is selected. (f) Electric-field energy of signal light SPP and modulation light LSP as functions of plasmonic nanoslit width.

energy: $E^2 = \int |E_i|^2 dw$, where E_i is the electric field intensity at the point of contact between the nanoslit and MoS₂, and W is the width of the nanoslit). The trends for the two curves are exactly opposite: the electric field intensity of the signal light SPP decreases exponentially with nanoslit width, while the modulation light LSP gradually increases. This is due to the different resonance frequencies and excitation mode of the modulation and signal light. It is revealed that for the signal light, the smaller nanoslit has a strong confined field, which can improve the SPP–MoS₂ interaction. For the modulation light, a large nanoslit width can ensure that a larger proportion of LSP are absorbed by MoS₂. In addition, the electric field energy of the modulation light LSP is much higher than that of the signal light SPP, which means that the influence of the modulation light LSP on the device performance will be dominant. To satisfy the requirement for sufficient interaction between the plasmon and MoS₂, a trade-off is required to select a suitable nanoslit width. The 120-nm slit width is considered as the critical point, at which the electric field intensities of the signal and modulation light

plasmon are completely balanced. Therefore, this is regarded as the optimal slit width.

The material properties of MoS₂ and the prepared MoS₂-plasmonic nanoslit device were characterized. A micrograph of the MoS₂ grown on the SiO₂ substrate is shown in Figure 2a. The MoS₂ film is triangular and has a uniform thickness. The length of this MoS₂ section is approximately 100 μm (white dotted line) and can be positioned to completely cover the nanoslit by use of the directional transfer method. Scanning transmission electron microscopy (STEM) allowed observation of the regular atomic structure of MoS₂ (Figure 2b). The selected area electron diffraction (SAED) pattern (the inset in Figure 2b) reveals the single crystal characteristics of MoS₂. Figure 2c depicts the Raman spectrum of MoS₂, with peaks assigned to the in-plane E_{2g}^1 mode and the out-of-plane A_{1g} mode at 384.6 and 404 cm⁻¹, respectively. The frequency difference between the two modes is approximately 20 cm⁻¹, indicating that the MoS₂ forms a monolayer on the surface [40]. The photoluminescence (PL) spectrum of MoS₂ (the inset in Figure 2c) shows that its optical band gap is 1.84 eV [41],

consistent with the theoretically reported monolayer MoS₂ band gap (1.8 eV). A false-color SEM image of the MoS₂-plasmonic nanoslit device is given in Figure 2d (detailed device preparation is given in Supplementary Note 3). The MoS₂ film, highlighted by a dark green color, completely covers the plasmonic nanoslit region. The length of the nanoslit is 9.7 μm , and the width is 120 nm. The propagation loss of the plasmonic nanoslit is estimated to be approximately 5 dB. The atomic force microscopy (AFM) amplitude map shows that the MoS₂ film on the nanoslit is undamaged (Figure 2e), and the height scan of the white dashed area shows the MoS₂ is suspended on the nanoslit, ensuring that the MoS₂ can fully interact with the confined electric field in the nanoslit. In addition, the typical Raman peak intensities for different slit widths are shown in Figure 2f. The wavelength of the laser light source used is 523 nm and the microscope magnification is 100 \times . The results show that the Raman typical peak intensity is highest when the slit width is 120 nm. This reveals that with 523-nm excitation, the larger the nanoslit width, the stronger the electromagnetic enhancement caused by LSP resonance. This conclusion verifies the simulation results for the MoS₂-plasmonic nanoslit with modulation-light excitation.

The MoS₂-plasmonic nanoslit all-optical experiment was conducted to verify the modulation ability. The optical test system used in the experiment is shown in Figure 3a.

The output wavelength of the tunable continuous laser (TFL-C-20) is 1550 nm (power, 1 mW), and this is used as the signal light. The fiber polarization controller is used to select TE-mode light as the signal light and couple it to the device through a tapered fiber. At the other end of the output, the spectrum analyzer is connected to the optical fiber (AQ6370D, wavelength range: 600–1700 nm) to receive the signal light. The insertion loss of the device was 42 dB, mainly from the fiber-waveguide coupling and the signal light SPPs propagation in nanoslit. A 532-nm laser is used as the modulation light with a 160- μm spot diameter, and the maximum output power is 80 mW. By adjusting the rotation angle of the attenuator, modulation light output with different powers can be realized. Figure 3b depicts the signal light transmission output ($T = 10 \times \log_{10}(P_{\text{out}}/P_{\text{in}})$), where P_{in} is the signal light input power and P_{out} is the signal light output power as a function of the modulation light power. When the modulation is in the OFF state, the signal light output intensity is -48.4 dBm. As the modulation light power increases, the signal light output intensity gradually decreases, indicating that the absorption of signal light by MoS₂ increases.

The blue curve in Figure 3c is the signal light transmission output at 1550 nm and this decreases linearly as the power increases. This trend is exactly the opposite of that for light saturation absorption in all-optical

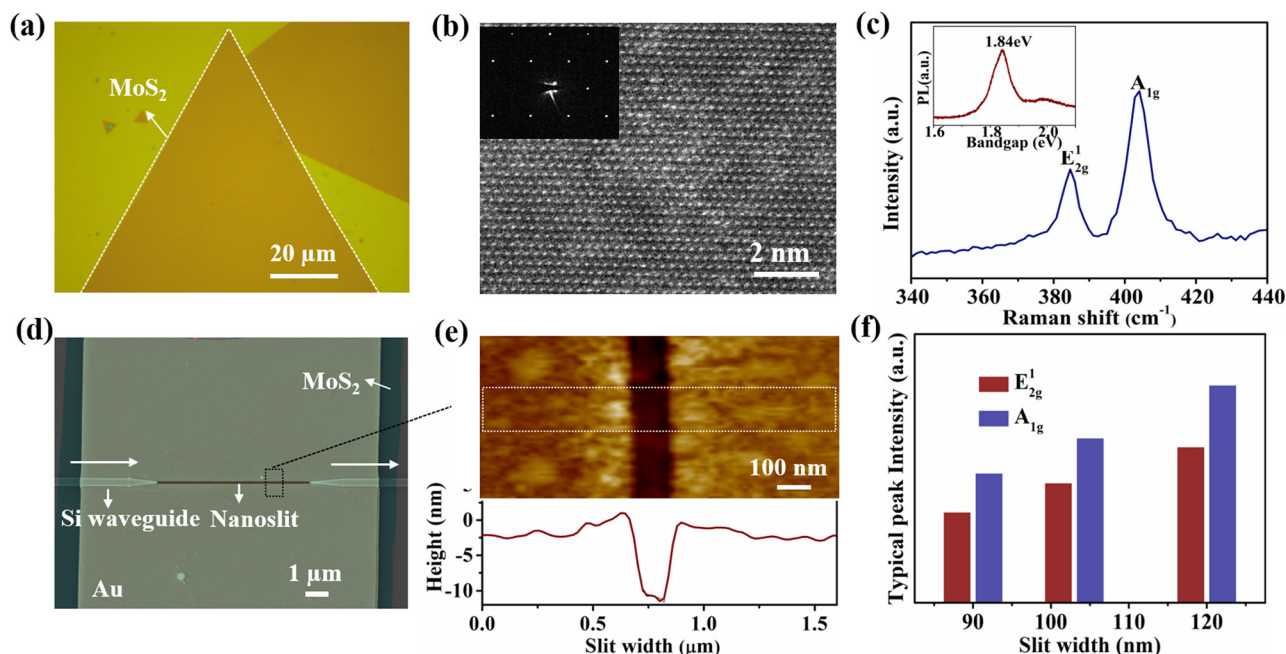


Figure 2: MoS₂ material and MoS₂-plasmonic nanoslit device characterization.

(a) Microscopy image of MoS₂ thin film grown on SiO₂. (b) STEM image of MoS₂ crystal; the inset shows the SAED pattern. (c) Raman and PL spectrum of monolayer MoS₂. (d) False-color SEM image of the device. (e) Amplitude scan of MoS₂-plasmonic nanoslit area by AFM; the height difference between the MoS₂ film at the nanoslit and the Au surface is approximately 7 nm. (f) Typical Raman peak intensities of MoS₂ with slit widths of 120, 100, and 90 nm.

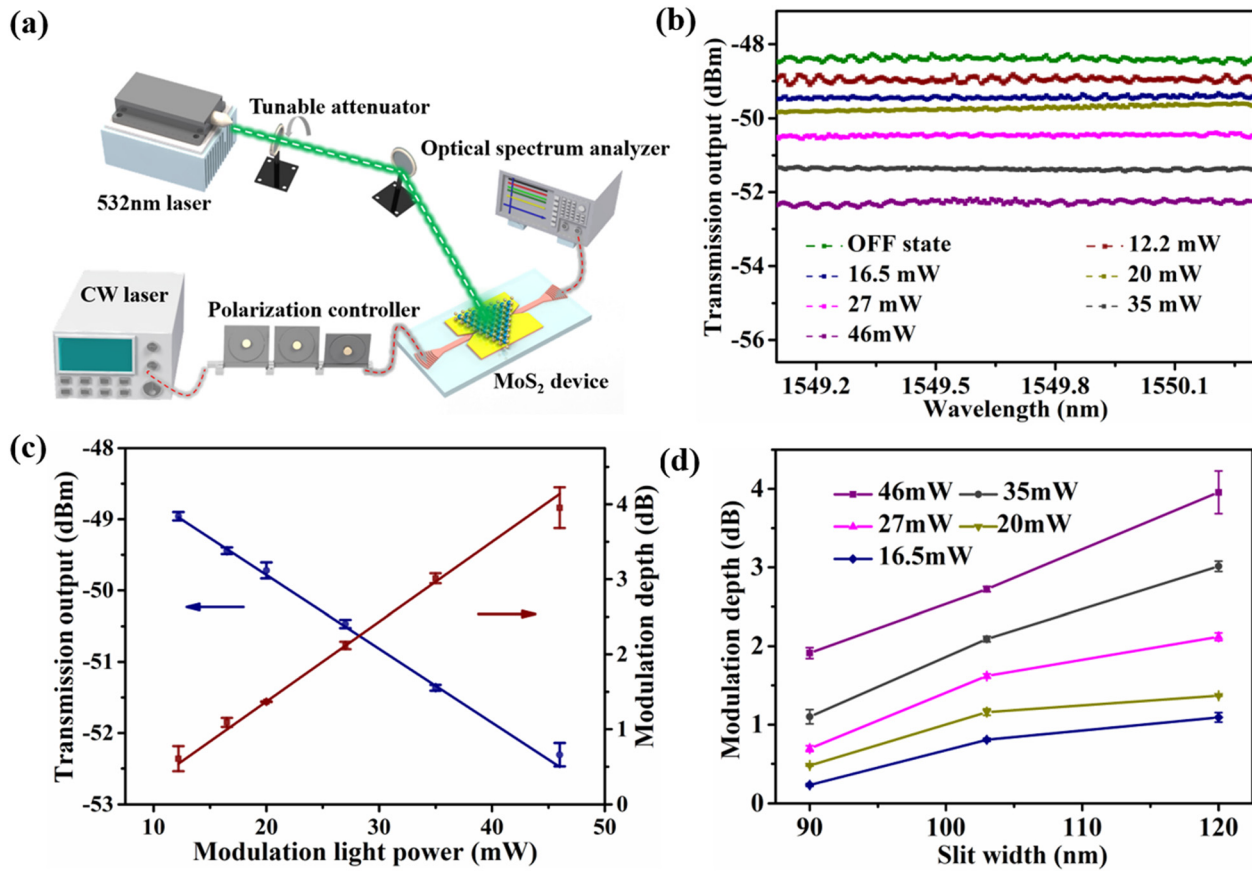


Figure 3: MoS₂-plasmonic nanoslit device all-optical modulation.

(a) All-optical modulation test system. (b) Change in signal light output with modulation light power. (c) Transmission output ($T(\text{dBm}) = 10 \times \log_{10}(P_{\text{out}}/1 \text{ mW})$) and modulation depth of the signal light. (d) Modulation depth as a function of slit width; the slit widths are 90, 110, and 120 nm.

modulation because the increase in modulation light power stimulates MoS₂ to generate more localized electron-hole pairs, leading to intraband electron transitions that require the absorption of a larger amount of signal SPPs. However, for optical saturable absorption modulators, Pauli blocking causes photon competition between the signal and modulation light. Consequently, the absorption of the signal light decreases when the power of modulation light increases [13]. The modulation depth, represented by the red curve in Figure 3c, increases linearly as the modulation power since it is proportional to the modulation light-induced absorption coefficient. The maximum modulation depth is 3.95 dB, and the modulation efficiency (modulation efficiency = modulation depth/effective contact length) is 0.41 dB/ μm . When the modulation light power increased, the output of signal light would increase correspondingly, and eventually tend to be saturated. This is caused by the energy threshold of the material.

The modulation depths of the devices with nanoslit widths of 90 and 103 nm were also tested (Figure 3d). Under

the same laser power, their maximum modulation depths are 1.9 and 2.7 dB, respectively. Among the devices with nanoslit widths of 90, 103, and 120 nm, the 120-nm-width device has the largest modulation depth (3.95 dB). This reveals that the influence of the nanoslit width on the modulation light LSP plays a dominant role in determining the modulation depth of the device, within a certain width range, which verifies the theoretical simulation results. It is also demonstrated that the device modulation is not caused by direct excitation of the signal light SPP, nor is it a result of a thermal effect of the signal light irradiation. In addition, we also fabricated a normal silicon waveguide device with MoS₂, where the area of MoS₂ was the same as that of the nanoslit device. Under the maximum power excitation (46 mW), the experimental result shows that the light-induced absorption at 1550 nm is almost weak (as shown in Figure S4).

To further verify the MoS₂ all-optical modulation mechanism excited by the plasmonic nanoslit, an experiment based on the double-layer MoS₂ all-optical modulator

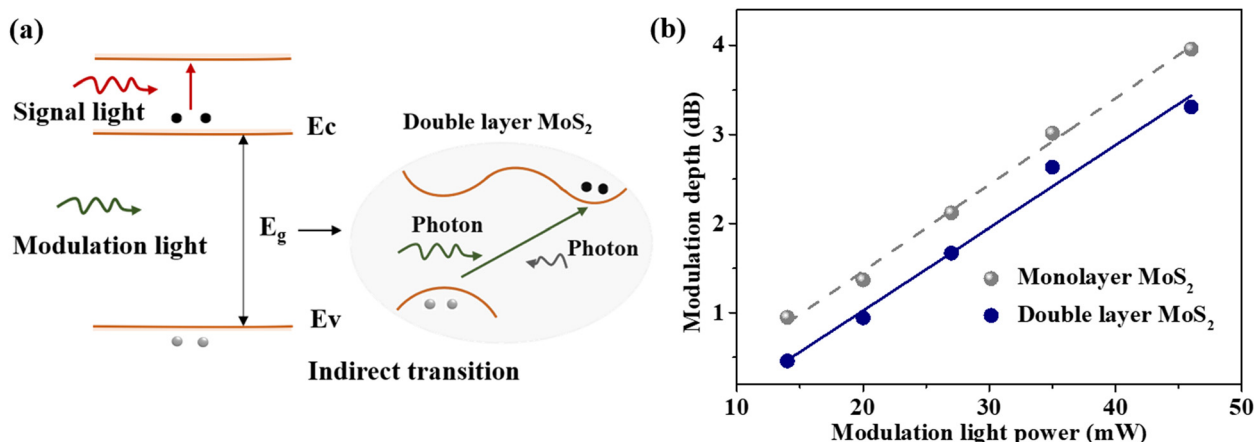


Figure 4: Double-layer MoS₂-plasmonic nanoslit all-optical modulation.

(a) Schematic of working principle for the double-layer MoS₂ device. (b) Modulation depth of monolayer and double-layer MoS₂ devices versus modulation light power.

was performed. Unlike monolayer MoS₂ which has a direct bandgap, double-layer MoS₂ is an indirect-band-gap material. The interband transition not only requires the interaction of photons and electrons but also depends on the participation of phonons (Figure 4a). This leads to a transition probability lower than that of a direct transition when the modulation light LSP is excited [42, 43]. Therefore, the number of electron–hole pairs generated by the double-layer MoS₂ via excitation by the modulation light LSP is significantly reduced, leading to a reduction in the signal light absorption. The double-layer MoS₂-plasmonic nanoslit device was prepared with a slit width of 120 nm (other parameters are the same as mentioned above). The results in Figure 4b show that the modulation depth of the double-layer MoS₂ device is obviously lower than that of the single-layer device, at the same modulation light intensity. The modulation depth of the double-layer MoS₂ device is 3.3 dB, and the modulation efficiency is 0.34 dB μm^{-1} . This result further confirms the modulation mechanism of the device and also proves that it is not caused by a thermal effect of the metal.

The dynamic response mechanism of the proposed modulator is characterized and analyzed next (Figure 5). For the MoS₂-plasmonic nanoslit hybrid structure, the work function mismatch between MoS₂ and Au structure leads to the Fermi level of MoS₂ shift (Figure 5a i–ii) [44–46] and the formation of a homojunction of MoS₂ with a symmetrical built-in electric field (Figure 5a ii–iii). When the modulation light is illuminated, the excited electrons in the depletion region will move to the center of nanoslit (Figure 5a iii) and interact with the signal light SPP to undergo an intraband transition. The output signal light intensity thereby decreases gradually until the internal

electric field of MoS₂ reaches dynamic balance. When the modulation light is turned off, the photogenerated carriers will overcome the built-in electric field and gradually recombine to the initial equilibrium state. Therefore, the response time of the proposed modulator depends on the photocarrier dynamics in the MoS₂-plasmonic nanoslit hybrid structure.

The response time test of the proposed modulator is carried out, and the experimental setup is shown in Figure S5. A square-wave control light (wavelength is 532 nm) with a modulation frequency of 10–5000 Hz and optical power of 30 mW is coupled into the MoS₂-plasmonic nanoslit hybrid structure. The output signal is received by a 5 GHz photodetector and a 100 MHz oscilloscope. Figure 5b shows the waveforms of signal output at the modulation frequency of 10 Hz. The results signify that the rise time is about 11 ms and the fall time is about 14 ms (Figure 5c). The change of output voltage ($V_{\text{max}} - V_{\text{min}}$) at different frequencies is shown in Figure 5d. The 3 dB bandwidth [47] of the MoS₂-plasmonic nanoslit modulator is estimated to be 700 Hz. Compared with most TMDs' all-optical modulators, our modulator shows an excellent frequency response [20, 30].

For the proposed device structure, both the signal and modulation light can interact with MoS₂ by means of strong electromagnetic confinement of plasmonic nanoslit. Meanwhile, the drive of the built-in electric field in the homojunction makes the light–matter interaction area extend effectively. As a result, the proposed modulator exhibits higher modulation efficiency and smaller device size compared to the currently reported 2D material-based all-optical modulators (Table 1). In addition, the response time in the millisecond range is consistent with the currently reported TMDs based all-optical modulators

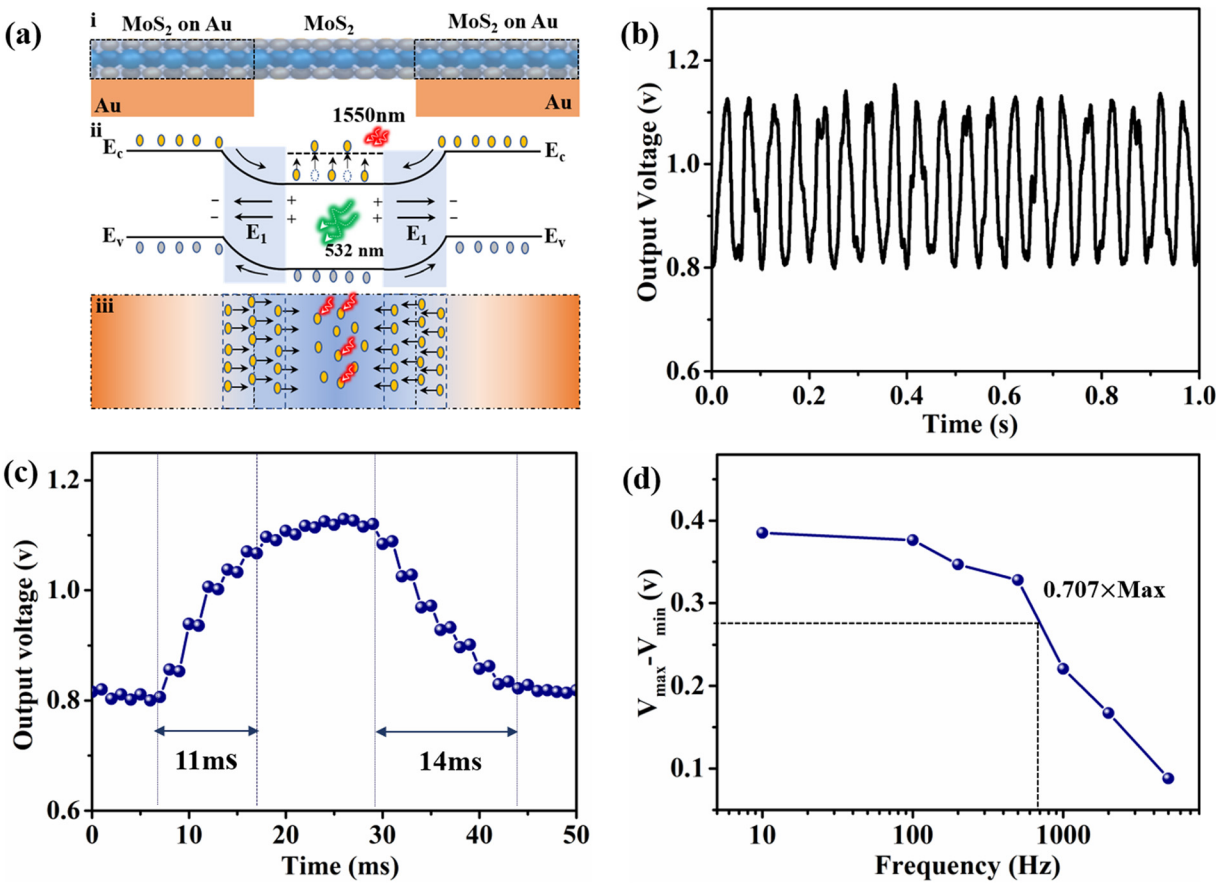


Figure 5: Frequency response of MoS₂-plasmonic nanoslit modulator. (a) Schematic of dynamic response mechanism. (i) On both sides of the nanoslit, MoS₂ is in contact with the Au layer; on the nanoslit, MoS₂ is suspended. (ii) Energy band diagram of MoS₂ homojunction. (iii) Excited electron movement driven by a built-in electric field. (b) Measured signal light waveform changes with time. (c) Rise time and fall time of proposed modulator. (d) Output voltage changes at different frequencies.

[20, 30], even if the carrier dynamics in the homojunction cause the response time to be limited. Energy band engineering [48] or interface optimization technology [49] is expected to further balance the response time and the modulation efficiency, achieving a fast and high efficiency all-optical modulator.

Table 1: 2D material-based all-optical modulators.

Devices	Signal light (nm)	Modulation depth (dB)	Interaction length (μm)	Modulation efficiency (dB μm ⁻¹)	Rise time/fall time	Ref.
MoS ₂ -plasmonic nanoslit	1550	3.95	9.7	0.41	11/14 ms	This work
Optically induced MoS ₂ nanosheets	632.8	7	–	–	–	[22]
WS ₂ /silicon-nitride waveguide	640	2	~50	~0.04	0.5/0.5 ms	[21]
MoSe ₂ -microfiber	1550	2	2.5 × 10 ⁴	0.0012	400/600 ms	[30]
WS ₂ -microfiber	1520–1620	2.71–3.99	–	–	20.5/19.6 ms	[20]
Stereo graphene-microfiber	1550	7.5	12 × 10 ³	6.3 × 10 ⁻⁴	Ns	[15]
Graphene-clad microfiber	1550	38%	16	–	2.2 ps	[14]
Graphene-plasmonic slot	1550	2.1	10	0.21	–	[34]
Graphene plasmon-enhanced	1550	3.5	12	0.28	30–120 fs	[18]

3 Conclusions

In conclusion, we have realized the all-optical modulation using MoS₂ at 1550 nm via highly confined plasmonic nanoslit excitation. The maximum modulation efficiency can reach 0.41 dB μm^{-1} and the 3 dB bandwidth is about 700 Hz. Plasmonic nanoslit excitation in MoS₂ can be further extended to other 2D material optoelectronic devices. Furthermore, the small-scale MoS₂ optical modulator operates at the communication wavelength and is compatible with CMOS technology, which is suitable for the development of high-density integrated chips. This has great application prospects in on-chip optical interconnection and smart sensing.

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Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

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