Abstract

All-optical modulation has been regarded as an effective method to solve the electrical bandwidth bottleneck problems existing in the current telecommunication network. By taking advantage of high photothermal conversion of two-dimensional (2D) MXene, a high-performance all-optical modulator is demonstrated. The polarization-dependent all-optical modulator exhibits a broadband intensity modulation behavior with a modulation depth of 15 dB. The response time of such all-optical modulator is approximately ten times than that of fiber-type MZI/MI-assisted all-optical modulator based on the thermal-optic effect. Besides, an all-optical information loading with a bit rate of 400 bit s⁻¹ is successfully achieved. Therefore, it is anticipated that the all-optical modulator with the advanced optical structures and excellent 2D materials has extraordinary potentials for future optical information processing.

1. Introduction

All-optical signal processing has attracted considerable attention as the processing speed of the current communication system gradually reaches the limitation [1]. Owing to excellent optical properties of two-dimensional (2D) materials, such as broadband absorption and ultrafast optical response, all-optical modulators based on 2D materials have made many remarkable achievements in the past decade. Until now, various all-optical modulators have been demonstrated by taking advantage of the saturable absorption [2–6], Kerr effect [7–10], and thermo-optic effect [11–16] of 2D materials. In 2014, Li et al reported an ultrafast all-optical graphene-based modulator with a response bandwidth of ~200 GHz and a modulation depth of 38% [2]. The response speed of this modulator is much faster than that of current electronic devices, suggesting that all-optical modulators based on 2D materials have abilities to realize high-speed information processing. However, its modulation depth needs to be further enhanced to satisfy the requirement of practical applications [17]. Taking advantage of phase modulation brought by Mach–Zehnder interferometer, Yu et al significantly improved the modulation depth of a high-speed all-optical modulator with 4.6 times enhancement [9]. After that, all-optical modulators based on interferometer structures (such as Mach–Zehnder interferometer and Michelson interferometer) are widely reported [13, 18–21]. However, as for this kind of all-optical modulators, their response time of these all-optical modulators is mainly restricted to the millisecond level, hardly meeting the demands of practical applications. Therefore, it is worthy to explore novel high-speed all-optical modulator to overcome these problems.

Besides, selecting an appropriate optical material is of importance to promote the response time of all-optical modulators. Owing to the great thermal conductivity and the ultra-fast carrier response time of...
Recently, MXene, a graphene-like layered material, has drawn more concerns for its broadband optical absorption from ultraviolet to the infrared region [27–29], high conductivity (∼10,000 S cm⁻¹) [30], and ultrafast optical response (∼fs level) [31]. Various MXenes (over 30 types) have been successfully obtained experimentally [32], such as Ti₃C, Ti₃C₂ and Mo₂C, which provide many opportunities to explore the 2D materials with excellent electrical and optical properties. Notably, MXene Ti₃C₂Tx possesses large photothermal conversion efficiency (∼100%) [33] and a great absorption coefficient (15 ∼ 40 g⁻¹ cm⁻¹), indicating that MXene Ti₃C₂Tx is a potential optical material in terms of light absorption and energy conversion. Moreover, the thermal conductivity of Ti₃C₂Tx and Ti₃C₂Tₓ/PVA composite are 55.8 W m⁻¹ K⁻¹ and 47.6 W m⁻¹ K⁻¹ [34], respectively, which are greater than that of other 2D materials, such as MoS₂ [35] and black phosphorus [36], suggesting that Ti₃C₂Tx have the potential in thermal related applications. The strong Ti-O bonds in Ti₃C₂Tₓ/PVA composite film can improve the thermal stability of Ti₃C₂Tₓ by decreasing the thermal coefficient of Eg mode [34]. The tensile strength of the Ti₃C₂Tₓ/PVA composite film (91 ± 10 MPa) is much improved than that of pure Ti₃C₂Tₓ (22 ± 2 MPa) [37]. These promising properties feature MXene as an ideal candidate material to be applied to high-performance 2D nanomaterials/polymer composite-based devices.

Herein, by taking advantage of the excellent photothermal effect of MXene Ti₃C₂Tₓ, an all-optical modulator with microsecond-level response time is demonstrated. The MXene/PVA composite film exhibits excellent thermal stability for over 1 h. A sandwich structure consisting of an MXene/PVA composite film and two fiber connectors achieves a great power-dependent temperature slope of 1.02 °C mW⁻¹. By detecting the state of polarization (SOP) variation of signal light, a broadband all-optical intensity modulator with a great modulation depth of 15 dB is realized. The binary codes carried by pump light is successfully copied onto the signal light, achieving all-optical information loading. The MXene-based all-optical modulator opens a door for the 2D materials-based all-optical devices in the next-generation communication network.

2. Fabrication and characterization of MXene nanosheets

The MXene Ti₃C₂Tₓ is prepared by the acid etching method. The raw material Ti₃AlC₂ with the di-ionic solution (>0.05 g mL⁻¹) was mixed with 40 w.t.% hydrofluoric acid. A magnetic stirrer was utilized to facilitate the delaminating process which sustained 13.5 h at 48 °C. The obtained deposition was washed via DI water until pH > 6 and dried in a vacuum furnace at 80 °C for 6 h. MXene powder was then dispersed into N-Methyl-2-pyrrolidone. MXene solution is prepared via centrifugation with a speed of 4000 rpm for 20 min at 10 °C. Its surface morphology is measured by a scanning electron microscope (SEM), illustrating that MXene nanomaterials with the layered structure are successfully realized, as shown in figure 1(a). The elements distribution of prepared MXene Ti₃C₂Tₓ is detected by the energy dispersive spectrometer (EDS) in figure 1(b), showing that the Al element of the MAX phase is almost completely etched away and the Ti, C, and F elements are uniformly distributed. The microstructure of MXene nanosheets is observed by high-resolution transmission electron microscopy (HRTEM), as shown in figure 1(c). To fabricate Ti₃C₂Tₓ-doped polymer film, polyvinyl alcohol (PVA) is chosen as the host polymer owing to its low optical absorption from visible to near-infrared, good thermal properties, low cost, and lightfastness. The Ti₃C₂Tₓ nanosheets dispersion is mixed with PVA solution (mass fraction of 3%) at a volume ratio of 1 to 3, followed by stirring for 2 h to obtain a homogenous composite dispersion. The dispersion is dripped on the glass slide and dried in a vacuum oven for 3 h to get a uniform MXene Ti₃C₂Tₓ/PVA thin film, as shown in figure 1(d). From the absorption spectra in figure 1(e), it is confirmed that the broadband absorption property of MXene/PVA film in the visible and near-infrared region comes from MXene Ti₃C₂Tₓ rather than PVA. The composite film is inserted into the middle of two fiber connectors to constitute a sandwich structure, as shown in figure 1(f).

Upon guiding a 980 nm continuous-wave laser into the sandwich structure, the MXene/PVA film absorbs the injected light to generate heat, leading to the thermal-induced birefringence. In the process of energy conversion, the temperature change of the composite film at different light power is continuously monitored by an infrared thermal camera. The temperature at the power of 0 mW and 10 mW are 21.1 °C and 31.5 °C, respectively, as shown in figures 2(a) and (b). The temperature increases linearly with the light power with a slope of 1.02 °C mW⁻¹ in figure 2(c). The power-dependent temperature slope in this sandwich structure improves approximately 50 times higher than that in 2D materials-deposited microfiber structures (0.02 °C mW⁻¹ [13] and 0.026 °C mW⁻¹ [20]), suggesting that the optimized structure improves the photothermal conversion of materials. The temperature variations of MXene/PVA film and pure PVA film within 1 h at the power of 10 mW are shown in figure 2(d). It can be easily concluded that the MXene/PVA film possesses excellent thermal stability and the generated heat indeed results from the photothermal conversion of MXene nanosheets rather than PVA.
Figure 1. SEM image (a) and EDS image (b) of etched Ti$_3$AlC$_2$. (c) HRTEM image of exfoliated MXene Ti$_3$C$_2$Tx nanosheets. (d) Optical microscope image of MXene/PVA film. (e) The absorption spectra of MXene, PVA film, and MXene/PVA film. (f) Image of MXene/PVA film installed on the end surface of the fiber connector.

Figure 2. The infrared thermograms at the light power of 0 mW (a) and 10 mW (b). (c) The temperatures of MXene/PVA film under different light powers. (d) Temperature stability of PVA film and MXene/PVA film within 1 h.

3. Experimental setup

A fiber-type all-optical modulator is established, as shown in figure 3. The signal light at 1550 nm emitted from amplified spontaneous emission (ASE) source is transformed into linearly polarized light by a polarizer. The signal light and the pump light are combined by a wavelength division multiplexer and injected into the
Figure 3. Experimental setup of the all-optical modulator.

Figure 4. Polarization state of the signal light at the pump power of 0 mW (a) 5 mW (b) and 15 mW (c). (d) Polarization evolution of signal light on the Poincaré sphere. (e) Azimuth and ellipticity as a function of the pump power.

proposed sandwich structure. Upon injecting pump power, the composite film will absorb the pump light to change the refractive index of the MXene/PVA film, which will modulate the SOP of the signal light. The pump light is ejected by WDM2 to prevent it from emerging at the output port. The signal light propagates through another polarizer with an extinction ratio of \( \sim 30 \text{ dB} \), and its intensity could be changed with the SOP evolution. Polarization controllers (PC1 and PC2) are employed to regulate the SOP of the signal light. The fiber type in the all-optical modulator is common single-mode fiber, which is fixed by adhesive tapes to avoid the fiber fluctuation. The SOP is detected by a polarimeter (PAX1000IR2). The output spectrum is measured by an optical spectrum analyzer (OSA) (Yokogawa AQ6370D). The intensity of the output signal light is recorded by a power meter and detected by an oscilloscope (Keysight DSOS104A, 1 GHz).

4. Experimental results and discussions

To investigate the influence of pump power on the birefringence of the MXene/PVA film, the SOP of the signal light after WDM2 is measured. When the pump power increased from 0 mW to 15 mW, the SOP of signal light approximately transforms from linearly polarized light, elliptically polarized light, to linearly polarized light, as shown in figures 4(a)–(c). The corresponding conversion efficiency of the phase shift is \( 0.06 \pi \text{ mW}^{-1} \). The polarization evolution of the signal light is shown in figure 4(d) with the increase of pump power. The ellipticity of signal light experiences firstly up and then down. The ellipticity of signal light is 0.5° and –1.3° at the pump power of 0 mW and 15 mW, respectively, as shown in figure 4(e). The azimuth is –46° and 37° at the pump power of 0 mW and 15 mW, respectively, indicating the two linearly polarized light is approximately orthogonal each other. Therefore, the SOP of the signal light is successfully modulated by the pump light.

In our experiment, the ASE light propagates through the proposed all-optical modulator, and the output spectrum of signal light can be observed on the OSA. By rotating the polarization controller (PC1 and PC2) to adjust the SOP of signal light, the maximum intensity modulation is obtained. The corresponding output spectrum is shown in the green line of figure 5(a). The output spectrum of the signal light at the pump
Figure 5. (a) Output spectra of the signal light at the pump power of 0 mW and 15 mW. (b) Output intensity of signal light versus pump power of pump light.

A pump light with a modulation frequency of 200 Hz and a pump power of 5 mW is introduced into the proposed all-optical modulator, as shown in figure 6(a). The frequency and duty cycle of the signal light is matched with that of the pump light. In figure 6(b), the falling edge and rising edge of signal light correspond to the presence and removal of pump light, respectively. It is clear that the rising edge and falling edge of signal light are smooth to some extent in comparison to the square wave of pump light, which can be contributed to slow heat generation and dissipation. If adjusting the polarization controllers (PC1 and PC2), a positive logic modulation can be realized (figure 6(c)). A magnifying drawing of all-optical intensity modulation is shown in figure 6(d). The two smooth edges are fitted by the exponential functions of $(1 - \exp(-t/t_r))$ and $\exp(-t/t_f)$, obtaining that the time constants of the rising edge and falling edge are estimated to be 108 µs and 150 µs, respectively, corresponding to a rising time of 238 µs and a falling time of 330 µs. In the field of fiber-type all-optical modulator based on thermo-optic effect, the response time of all-optical modulator based on polarization rotation is approximately ten times than that of MZI/MI-assisted all-optical modulators [12, 13, 18, 20], as shown in table 1, illustrating that the polarization rotation is an effective method to improve the modulation speed of all-optical modulator. As further improving the modulation frequency to 1 kHz and 2 kHz at the pump power of 5 mW, the signal waveform gradually transformed from edge-smoothed square wave to triangle wave (figures 6(e) and (f)), which mainly results from that the slow thermal response has no enough time to catch up with the rapid change of pump light. What we should note here is that the triangle wave still can be acted as a medium to transmit the information, because the deformed signal waveforms can be detected by the edge-detection method.

An all-optical information loading by using the proposed all-optical modulator is also realized. The information loading process only occurs in the photonics domain without optoelectronic conversion. In the experiment, the binary codes of pump light ('00101001') with a bit rate of 400 bit per second is introduced into the all-optical modulator. The experiment results of information loading are shown in figure 7. It is observed that the signal light efficiently copies the information of pump light with the same bit rate, and an all-optical information loading with two different logic states (positive and negative logic state) can be obtained. In table 1, we summarize the performance of fiber-typed all-optical modulators based on the thermo-optic effect. It is obvious that our work achieves all-optical modulation with fast response time in comparison with others, which provides a significant reference to develop high-performance all-optical devices based on 2D nanomaterials. It is believed that continuous optimization of optical structures and 2D materials is expected to achieve high-speed, low-loss, and integrated all-optical modulation.
Table 1. Fiber-typed all-optical modulators based on thermo-optic effect.

| Materials/structure | MD (dB) | Conversion efficiency of phase shift | Rising time | Ref. |
|---------------------|---------|--------------------------------------|-------------|------|
| Graphene/MZI        | 20      | 0.09π mW⁻¹                          | 3.2 ms      | [11] |
| WS₂/MZI             | 15      | 0.017π mW⁻¹                         | 7.3 ms      | [12] |
| BP/MZI              | 17      | 0.029π mW⁻¹                         | 5.5 ms      | [13] |
| Boron/MZI           | 11      | 0.013π mW⁻¹                         | 0.5 ms      | [14] |
| MXene/MZI           | 18      | 0.061π mW⁻¹                         | 9 ms        | [18] |
| MXene/MI            | 27      | 0.043π mW⁻¹                         | 5.3 ms      | [21] |
| Antimonene/MI       | 25      | 0.049π mW⁻¹                         | 7 ms        | [19] |
| MoS₂-PVA/PI         | -       | —                                    | 714 μs      | [15] |
| Graphene/MR         | 13      | —                                    | 295 μs      | [14] |
| Graphene/PR         | 20.8    | —                                    | 39 μs       | [16] |
| MXene-PVA/PR        | 15      | 0.06π mW⁻¹                          | 238 μs      | This work |

MD: modulation depth; BP: black phosphorus; MI: michelson interferometer; MZI: Mach–Zehnder interferometer; MR: microfiber resonator; PR: polarization rotation; PI: polarization interference.

Figure 6. Experiment results of the all-optical modulator. (a) The waveform of pump light. (b) Negative logic modulation and (c) Positive logic modulation of the signal light. (d) A switching profile of signal light and a corresponding fitting curve. The waveform of the signal light at the modulation frequency of 1000 Hz (e) and 2000 Hz (f).

Figure 7. Experimental results of all-optical information loader.

The signal light at 1550 nm is transformed into linearly polarized light by a polarizer and then transmits through the MXene/PVA thin film. Upon injecting pump power, the composite film will absorb the pump light to change the refractive index of the MXene/PVA film, which will modulate SOP of the signal light.
Figure 8. Principle diagram of the MXene/PVA thin film in the all-optical modulator.

(figure 8). When the polarization direction of linearly polarized light is parallel to that of the polarizer, the signal light possesses high output intensity. If the polarization direction of linearly polarized light is perpendicular to that of the polarizer, the output intensity will be extremely weak. Therefore, with the variation of the SOP of the signal light, the output intensity of the signal light will be modulated.

5. Conclusion

In conclusion, by utilizing polarization rotation of light and excellent photothermal effect of MXene, an all-optical modulator and an all-optical information loading operation are demonstrated with superior performance. The MXene/PVA composite film is facilely fabricated by a dispersion blending method. The sandwich structure achieves high power-dependent temperature slope (1.02 °C mW⁻¹), which extremely reduces the power loss of all-optical modulator. The all-optical modulator effectively achieves broadband modulation and improvement of response time. For an all-optical intensity modulator, the fast modulation speed is obtained. The rising time of 238 µs and the falling time of 330 µs has fast response time among fiber-typed all-optical modulators based on the thermo-optic effect. The all-optical modulator has been successfully applied to the all-optical information processing with a bit rate of 400 bit s⁻¹, suggesting that the MXene based all-optical modulator has a bright prospect in an all-optical signal process.

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Conflicts of interest

The authors declare no conflicts of interest.

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