Thermo-optic all-optical devices based on two-dimensional materials

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In this paper, we review our recent work on thermo-optic all-optical devices based on two-dimensional (2D) materials. The unique properties of 2D materials enable fast and highly efficient thermo-optic control of light. A few all-optical devices are demonstrated based on various thermo-optic mechanisms. Both fiber and integrated devices will be shown. © 2018 Chinese Laser Press

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1. INTRODUCTION

Two-dimensional (2D) materials have attracted wide attention for their unique photonic and optoelectronic properties. Since the discovery of first 2D material graphene [1–13], many 2D materials have been found, including graphene oxide [14], topological insulators (TIs) [15–18], transition metal dichalcogenides (TMDs) [19–26], black phosphorus (BP) [27–32], and MXene [33–36]. These novel 2D materials have exhibited various interesting photonic properties. For example, the controllable optical absorption by tunable Fermi level has enabled applications in compact optical modulators [4]. The wideband absorption has been utilized in photodetectors [6,29,30]. The intensity-dependent optical transmission has been widely used as saturable absorbers for ultrashort pulse generation [2,3,15,16,18,21–28,35]. The optical nonlinearity has been explored for all-optical devices, such as graphene modulators based on saturable absorption and the Kerr effect [5,10], thermo-optic switches with graphene [7,11] and phosphorene [31], and bistability devices with graphene [8]. By using graphene as a micro-heater, electrically tuned thermo-optic switches are also reported with high efficiency [12,13]. Some all-optical devices in other novel perovskite materials have also been demonstrated [37,38]. Among these demonstrations based on 2D materials, only a few thermo-optic all-optical devices have been reported, and not enough attention has been paid to this area of research.

In this paper, we summarize our recent work on all-optical devices based on 2D materials by exploring various thermo-optic mechanisms. For fiber devices, two TMD-based devices have been shown by using the wavelength-dependent absorption and thermo-optic effects of TMDs [39,40]. For integrated devices, a graphene-on-silicon nitride (Si₃N₄) switch has been achieved by using the high thermal conductivity of graphene [41]. A comparison among current reported works of thermo-optic all-optical devices based on 2D materials is also provided.

2. FIBER DEVICES

Optical fiber provides flexibility of device fabrication and characterization. Previously, two works with graphene have been reported, and very low pump power has been achieved [7,11]. A phosphorene all-optical fiber phase modulator has also been demonstrated recently [31]. In this section, we review our work on an all-optical fiber phase shifter and switches based on 2D materials.

Different from graphene, many 2D materials exhibit wavelength-dependent absorption due to the existence of a band gap. By choosing the proper pump wavelength (control light) in a high-absorption region and signal wavelength in a low-absorption region, a thermo-optic device can be constructed [39]. Figure 1(a) shows a typical all-optical phase shifter and switch made by using tungsten disulfide (WS₂). The band gap of WS₂ near 1.3 eV (954 nm) [25] allows good absorption of the pump (control light) at 980 nm and weak absorption of the signal light near 1550 nm. The all-optical phase shifter is shown in the red frame in Fig. 1(a). It consists of two 980/1550 wavelength division multiplexers (WDMs) and a WS₂-deposited tapered fiber. When the control light at 980 nm is applied, it is absorbed by WS₂ and generates heat. The WS₂ changes temperature and the tapered fibers change their refractive indices due to the thermo-optic effect. As a result, the 1550 nm signal light propagating through the WS₂-deposited tapered fiber is phase-shifted accordingly.

The WS₂ nanosheets are prepared by the standard liquid-phase exfoliation (LPE) method. Briefly, bulk WS₂ powder
is exfoliated into thin nanosheets by intense ultrasonic processing in a certain organic or inorganic solvent and then purified by centrifugation to remove the un-exfoliated flakes and obtain good-quality dispersions with WS₂ nanosheets. The WS₂ nanosheets’ dispersions are then deposited onto a tapered fiber by using optically driven deposition. That is, when a tapered fiber is immersed in the WS₂ dispersions and proper optical power is injected to the fiber, the WS₂ nanosheets will be attracted by the optical force and attach to the tapered fiber. Figure 1(b) shows a transmission electron microscopy (TEM) image of the WS₂ nanosheets, and Fig. 1(c) shows a microscopic image of the WS₂-deposited tapered fiber. The diameter of the tapered fiber is ~10 μm. The length of the deposition region is ~500 μm. The Raman spectrum of the WS₂-deposited tapered fiber is measured, and the successful deposition of WS₂ nanosheets is confirmed, as shown in Fig. 1(d).

To measure the actual phase shift of the phase-shifter device, the device is incorporated into a fiber Mach–Zehnder interferometer (MZI). The lengths of the two arms of the MZI are unequal so that a comb-like transmission spectrum can be obtained, as shown in Fig. 2(a). When there is a phase shift induced by the control light in one arm, the spectrum would shift, and the corresponding phase shift can be calculated based on this spectral shift. Figure 2(a) shows two spectra at 0 phase shift without control light (blue) and 5π phase shift with control light (red). Figure 2(b) shows the relation between the phase shift and the control light power. The slope efficiency is ~0.0174 π mW⁻¹. Normalizing to the length of the interaction region (i.e., 500 μm), a control efficiency of 0.0353 π mW⁻¹·mm⁻¹ is obtained. The phase shifter has an insertion loss of 3.5 dB at 1550 nm and 5 dB at 980 nm. This insertion loss is affected by the scattering loss of the randomly deposited WS₂ materials. Using chemical vapor deposition (CVD)-grown WS₂ thin film can help to reduce the scattering loss.

It is well known that an MZI converts the phase difference to the power variation and the two output ports of the MZI have complementary output. Therefore, the MZI with WS₂-deposited tapered fiber in one arm can function as an all-optical switch. An experimental result is shown in Fig. 2(c). The control light is a square wave (yellow), and the 1550 nm output signal at the output 1 port is a bright pulse train (blue). The complementary output signal at the output 2 port is a dark pulse train shown in Fig. 2(d). An exponential fit of the bright pulse in Fig. 2(c) indicates that the switch has a rise time constant of 7.3 ms and a fall time constant of 3.5 ms.

The thermo-optic coefficient of WS₂ can be estimated. By simulation, the optical power percentage is ~96% in the tapered fiber and ~3% in WS₂ both for 980 and 1550 nm. By increasing the pump power until the WS₂ material reaches its decomposition temperature in the air (with other samples),
we roughly estimate that the local temperature change of WS$_2$ tapered fiber is 40–60 K for a $\pi$ phase shift. The corresponding index change is $\Delta n = 1.08 \times 10^{-3}$ for an interaction length of 500 $\mu$m. The contribution of index change can be roughly estimated by $\Delta n = \Delta n_{WS2} \cdot 3\% + \Delta n_{TF} \cdot 96\%$ (see Appendix A). Noting that the thermo-optic coefficient for silica fiber is $\sim 1.1 \times 10^{-5}$ $\text{K}^{-1}$, and the index change of tapered fiber $\Delta n_{TF}$ is $(4.4 - 6.6) \times 10^{-4}$ for a temperature change of 40–60 K. The index change of WS$_2$ $\Delta n_{WS2}$ is then found to be $(1.49 - 2.19) \times 10^{-2}$, corresponding to a thermo-optic coefficient of $(2.48 - 5.48) \times 10^{-4}$ $\text{K}^{-1}$. To further confirm the estimation of the thermo-optic coefficient of WS$_2$, an ellipsometric measurement has been performed. The WS$_2$ sample is prepared by the CVD method on a sapphire substrate, and the sample is grown to a very large thickness of $\sim 200$ nm to meet the requirement of the ellipsometer. Figure 3(a) shows the refractive index calculated from the measurement data, and Fig. 3(b) is the index change at different temperatures. A thermo-optic coefficient of $3.36 \times 10^{-4}$ $\text{K}^{-1}$ is obtained. Although WS$_2$ with different thicknesses may have different thermo-optic properties, the analyses based on power distribution and based on ellipsometric measurement both indicate a coefficient of $\sim 10^{-4}$ $\text{K}^{-1}$, which is believed to represent the actual thermo-optic property of WS$_2$. This result indicates that WS$_2$ and tapered fiber have nearly equal contributions to the index change.

The device performance can be further improved by applying a new design [40], as shown in Fig. 4(a). The MZI structure in Fig. 1(a) is sensitive to environmental perturbation because the perturbations on the two arms are usually different and can affect the output. Another problem is that the deposition region is still long, which makes the response speed of the device low. To improve this, Fig. 4(a) shows a new all-optical switch with a polarization interferometer and a TMD-polymer thin film. Here, a MoS$_2$-polyvinyl alcohol (PVA) thin film is fabricated. MoS$_2$ nanosheet dispersions are prepared by the LPE method described above. Then the dispersions are mixed with a PVA solution and dried on a clean plastic dish under 50°C for 3 days to form the thin film. Figure 4(b) shows the fabricated MoS$_2$-PVA thin film and the film when it is cut and transferred to a fiber end.

The device principle can be explained by Fig. 4(c). The x–y plane is perpendicular to the direction of light propagation. By adjusting the polarization controllers (PCs), the polarization of the signal light is set to the $E_1$ direction with equal projection $E_x$ and $E_y$ on the x and y axes. The direction of the polarizer is perpendicular to $E_1$ so that the original polarizer output is very low. When the control light at 980 nm is applied, the MoS$_2$-PVA thin film absorbs the light power and generates heat. However, due to the naturally non-uniform distribution of the MoS$_2$ nanosheets in the PVA film, the heat distribution is not uniform either. This non-uniform heat distribution leads to a non-uniform index change and thus unequal phase shift to $E_x$ and $E_y$. In Fig. 4(c), the relative phase of $E_x$ is shifted by $\pi$,
and $E_x$ becomes $E'_x$ with the control light applied. The new polarization direction of the signal then becomes $E_2$, which has high transmission in the polarizer.

Figure 5(a) shows the output signal pulses (blue) from the polarizer when square-wave control pulses are applied (yellow). Figure 5(b) is a zoomed view of a single output pulse. By exponential fitting, the pulse has a rise time constant of 324.5 μs and a fall time constant of 286.7 μs. Compared with the WS$_2$-deposited tapered fiber in the MZI design, the MoS$_2$-PVA thin film allows much faster heat distribution and thus results in a more than 10 times faster switching time. Moreover, two interference beams share the same fiber in the polarization interferometer, and the output pulses are less sensitive to the environmental perturbation. Figure 5(c) shows a long-term output pulse train, and very stable output can be observed.

It should also be noted that the contribution of the saturable absorption of the TMD materials is negligible in the experiment because the modulation depth of TMD materials is only a few percent (the on–off power ratio is 10–15 dB in our work), and the saturation intensity is relatively high (20 MW/cm$^2$ intensity means 16 W power in fiber).

3. INTEGRATED DEVICE

To further improve the response time and shrink the footprint of the device, a graphene-on-silicon nitride (Si$_3$N$_4$) all-optical switch is proposed and demonstrated [41]. Previously, a similar device has been reported for thermo-optic bistability [8], and graphene has also been used as an efficient thermal heater in silicon devices [12,13]. In this work, the high thermal conductivity of graphene is utilized to efficiently transfer the heat to the waveguide and improve the switching speed.

Figures 6(a) and 6(b) show the experimental setup and the schematic diagram of the switch, which consists of a Si$_3$N$_4$ micro-ring resonator (MRR) with a graphene sheet on top. The diameter for the MRR is 60 μm. A straight waveguide is side-coupled to the MRR with a gap distance set to 100 nm. The dimensions for the straight waveguide and the waveguide forming the MRR are 1.2 μm × 400 nm. To enhance the coupling between the waveguides and tapered lenses, inverse tapers are integrated at the input and output terminals of the waveguides.

The device fabrication starts on a commercial wafer with a 400-nm-thick top Si$_3$N$_4$ layer and a 5-μm-thick buried oxide layer. Standard electron beam lithography (EBL) and inductive coupling plasma (ICP) etching processes are used to define the device structure. Then CVD-grown graphene on a copper foil is wet-transferred on top of it. The graphene layer is then patterned by EBL and an oxygen plasma etching process. As shown in the scanning electron microscope (SEM) image of the device in Fig. 6(c), only part of the MRR is covered by the graphene sheet, whose length is ∼43.4 μm. Thus, the loss from the graphene layer can be reduced.

Figure 6(d) shows the principle of the thermo-optic tuning switch. The solid line is the initial transmission spectrum.

![Fig. 5.](image1.png)  
(a) Pulsed control light (yellow) and output signal (blue). (b) A zoomed view of a single off–on transition of the output pulse (blue) and exponential fit (red). (c) A long-term stable output pulse train. Adapted with permission from Ref. [40].

![Fig. 6.](image2.png)  
(a) All-optical switch based on a graphene-on-Si$_3$N$_4$ device. (b) Device structure. (c) SEM image of device. (d) Device principle. Adapted with permission from Ref. [41].
When the optical control light at 1555 nm is injected into the device, part of the optical power is absorbed by the graphene layer and converted into heat. Then the heat can be directly transferred to the Si$_3$N$_4$ waveguide under the graphene. Thus, the transmission spectrum is red-shifted to the dash line due to the thermo-optic effect of Si$_3$N$_4$. Denoted as point A in Fig. 6(d), the transmission of the signal light at 1509 nm will be changed accordingly if its wavelength is set near the resonance wavelength.

The experimental setup is shown in Fig. 6(a). The control light at 1555 nm is first injected into an erbium-doped fiber amplifier (EDFA) for amplification. The wavelength of the control light matches the resonance wavelength of the MRR to maximize the power absorption. The signal light at 1509 nm matches another resonance wavelength to minimize the output power, as described in Fig. 6(d). The control and signal light beams are combined through a WDM and coupled to the chip through a lensed fiber. Here, both the control light and signal light are controlled by the polarization controllers to be quasi-transverse-electric (TE) polarized. The output light is coupled out by a second lensed fiber, and a WDM follows to separate the control light and the signal light. The signal light is then characterized by an oscilloscope to observe the all-optical switching process.

Figure 7 shows the dynamic switching property of the device when a pulsed control light is applied. The control light has a repetition rate of 200 kHz, a pulse width of 1 μs, an injected average power of 40 mW, and a peak power of 200 mW. By exponential fitting, the output signal pulse has a rise time constant of 253.0 ns and a fall time constant of 888.3 ns. The switching time is much faster than that of the conventional integrated thermo-optical switch [typically a few microseconds (μs)] due to ultra-high thermal conductivity of graphene. To further understand the physical mechanism of the device, a simulation based on coupled-mode theory (CMT) has been performed. The simulation result is shown in red in Fig. 7. It can be seen that the experimental result can be well reproduced by the simulation. The temperature change of the device is also shown in the inset of Fig. 7. The maximum temperature change and wavelength shift are calculated to be ~26 K and ~0.45 nm, respectively. The corresponding slope is 0.0173 nm/K, which is comparable to the reported experimental results in Refs. [42, 43]. More details of the CMT-based simulation can be found in Ref. [41].

It is worth mentioning that we have tried all three of our fiber and integrated devices with a fast pump pulse train of 10 GHz, and no modulation has been observed on the output probe signal. All the probe signal outputs show a rise time of the millisecond level for fiber systems and microsecond level for an integrated system, which are consistent with those of previously reported thermo-optic devices. This result indicates that the thermo-optic effect is the dominant mechanism in our devices rather than other fast nonlinear processes, such as the Kerr effect.

4. DISCUSSION

To summarize the progress of thermo-optic all-optical devices based on 2D materials, a comparison between our works and other reported works is provided in Table 1. In the table, a parameter called control efficiency is defined as the phase shift [with the unit of π radians (rad)] normalized to the pump power [with the unit of milliwatts (mW)] and the light-material interaction length [with the unit of millimeters (mm)].

For the fiber-based devices, three works use fiber MZIs with different 2D materials. The work with an MZI and graphene shows a very low pump power of 5.2 mW for π phase shift due to the long interaction length of CVD graphene [7]. Our work with an MZI and WS$_2$ has a low insertion loss of 3.5 dB at a signal wavelength of 1550 nm, benefitting from the low absorption of WS$_2$ at long wavelengths while maintaining a relatively good control efficiency of 0.035π mW$^{-1}$ · mm$^{-1}$ [39]. The work with an MZI and fluorinated phosphorene utilizes the good pump absorption of phosphorene and obtains a

![Image](image_url)

**Table 1. Comparison of Reported Works of Thermo-Optic All-Optical Switches Based on 2D Materials**

<table>
<thead>
<tr>
<th>Device</th>
<th>Integration</th>
<th>Switching Time Const.</th>
<th>Extinction Ratio</th>
<th>Control Efficiency</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fiber MZI w/graphene</td>
<td>No</td>
<td>3.2 ms</td>
<td>20 dB</td>
<td>0.018π mW$^{-1}$ · mm$^{-1}$</td>
<td>[7]</td>
</tr>
<tr>
<td>Fiber MZI w/WS$_2$</td>
<td>No</td>
<td>7.3 ms</td>
<td>15 dB</td>
<td>0.035π mW$^{-1}$ · mm$^{-1}$</td>
<td>Our work [39]</td>
</tr>
<tr>
<td>Fiber MZI w/phosphorene</td>
<td>No</td>
<td>2.5 ms</td>
<td>17 dB</td>
<td>NA$^*$</td>
<td>[31]</td>
</tr>
<tr>
<td>Fiber PI w/MoS$_2$</td>
<td>No</td>
<td>324.5 μs</td>
<td>10 dB</td>
<td>0.063π mW$^{-1}$ · mm$^{-1}$</td>
<td>Our work [40]</td>
</tr>
<tr>
<td>Fiber micro-ring w/graphene</td>
<td>No</td>
<td>~134 μs$^*$</td>
<td>13 dB</td>
<td>0.31π mW$^{-1}$ · mm$^{-1}$</td>
<td>[11]</td>
</tr>
<tr>
<td>Si$_3$N$_4$ micro-ring w/graphene</td>
<td>Yes</td>
<td>253 ns</td>
<td>10 dB</td>
<td>0.065π mW$^{-1}$ · mm$^{-1}$</td>
<td>Our work [41]</td>
</tr>
</tbody>
</table>

$^*$The deposition length of the phosphorene material was not mentioned in Ref. [31].

$^*$Estimated from Fig. 5(b) in Ref. [11].
relatively fast rise time constant of 2.5 ms and low pump power of 34.5 mW [31]. For our work with a polarization interferometer and MoS$_2$, the device exhibits a fast rise time constant of 324.5 μs due to the short interaction length [40]. For the work with a fiber micro-ring resonator and graphene, the small footprint and ring enhancement allow an even faster rise time constant of ~134 μs and a very high control efficiency of 0.31 π/mW/mm [41].

For the integrated devices, to the best of our knowledge, our graphene-on-Si$_3$N$_4$ device seems to be the only reported thermo-optic all-optical switch based on 2D materials. The device has a very fast rise time constant of 253 ns and a good control efficiency of 0.065 π mW$^{-1}$·mm$^{-1}$ enabled by the small footprint and high thermal conductivity of graphene [41].

5. CONCLUSIONS

In conclusion, we review our recent works on all-optical devices based on 2D materials by exploring various thermo-optic mechanisms. The unique properties of 2D materials allow fast and highly efficient all-optical control of light. A comparison among current reported works on thermo-optic all-optical devices based on 2D materials is also provided. The compactness of 2D materials makes them ideal candidates as light interaction media for both fiber and integrated platforms.

APPENDIX A

In the appendix, we briefly explain the equation $\Delta n = \Delta n_{\text{WS}_2} \cdot 3\% + \Delta n_{\text{TE}} \cdot 96\%$ for the estimation of index change. This equation is based on some empirical relations on conventional fiber. Here we use a standard single-mode fiber to explain this equation. We do not use the WS$_2$-deposited tapered fiber in the following simulation because the dimensional difference is too large between WS$_2$ and fiber, and the simulation result is less accurate. Figure 8(a) is the mode distribution (power) of a standard single-mode fiber by 2D simulation, and the power percentage in the core area is ~70%. The core index is 1.44, and the cladding index is 1.434816. The calculated effective index is 1.4370. Figure 8(b) is the change of the effective index with the change of the core index by simulation. The straight line has a slope of 0.7. Figure 8(c) is the change of the effective index with the change of the cladding index. The straight line has a slope of 0.3. Therefore, the relation of $\Delta n = \Delta n_{\text{core}} \cdot 70\% + \Delta n_{\text{clad}} \cdot 30\%$ can be used to estimate the contribution to index change in a standard single-mode fiber. Similarly, we use $\Delta n = \Delta n_{\text{WS}_2} \cdot 3\% + \Delta n_{\text{TE}} \cdot 96\%$ to estimate the index change induced by WS$_2$ and a tapered fiber.

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**Fig. 8.** (a) Mode distribution in a single-mode fiber. Relation between (b) index change and core index change, and (c) index change and cladding index change.


