Ultrafast All-Optical Polarization Switching Based on Composite Metasurfaces with Gratings and an Epsilon-Near-Zero Film

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Active control of light polarization is necessary for the development of laser physics, optical communications, and also the molecular dynamics. Conventional polarization control methods suffer from low speed, and the existing all-optical switching based on nonlinear materials solely has a lot of conducting restrictions,impeding its practical utilizations. Herein, an all-optical polarization switching in the near-IR range, composed of the plasmonic gratings deposited on an indium tin oxide (ITO) film with epsilon-near-zero response, is realized. A sub-1 ps response speed and the 19° rotation of the polarization ellipse are determined with the excitation power of 1.37 GW cm⁻². The device is demonstrated to output the varied signal with tuning the incident polarization and intensity in a spectral range from 1200 to 1500 nm. The ultrafast processing speed, broadband, and diverse tunability ensure the designed device a promising and necessary element for practically integrating into the next-generation signal-processing systems with high performance and large capacity.

1. Introduction

Polarization state is one of the key attributes of electromagnetic wave, and it can be worked as the information carriers for developing the optical signal recording and processing systems. Conventionally, the polarizations of the transmitted light are controlled by the polarizers fabricated by the birefringent crystals or dichromatic organic films, where the polarization state is fixed in a steady direction. For the on-demand active modulation of the polarizations, the electro-optical (Pockels cells), acousto-optical (Raman–Nath diffractometers), and magneto-optical effects (Faraday rotators) are explored.[4–6] However, all of these extensively applied devices are macroscopic and not suitable for the highly integrated applications. In addition, the involvement of electrons inside these devices determines the upper limits of switching speed to be several tens of gigahertz, which cannot satisfy the high-speed demands for next-generation signal-processing systems with large capacity.

Considering the integration requirements, artificial nano-/micro-structure arrays with ultrathin sub-wavelength thickness, i.e., metamaterials or metasurfaces, have been developed in recent years for shaping the phase of the transmitted wave to passively control the polarization, including nanoantennas, split-ring resonators, helical nanoarrays, and hyperbolic metamaterials.[7–11] While considering the active-modulation of the polarizations, a variety of physical mechanisms have been involved, containing mechanical deformation, thermo-induced materials’ dielectric–metal phase change, electrical p-i-n-diode, and so on.[12–14] Similar to the aforementioned macroscopic polarization-controlling devices, these metamaterial-based devices also suffer from the relatively-low tuning speed, attributed to the adopted modulation methods. Considering the ultrafast modulation, the all-optical polarization control is a necessary, where the polarization state of the transmitted signal is dynamically controlled with another injected pump and this scene relies on the optical nonlinearity of the involved materials.[15] Unfortunately, the commonly used materials for the formation of metamaterials, including noble metals and silicon-based dielectrics, provide small optical nonlinearity and are hardly to be tuned. The epsilon-near-zero (ENZ) materials, where the material’s real part of permittivity vanishes at a given wavelength, provide the upper limits of switching speed to be several tens of gigahertz, which cannot satisfy the high-speed demands for next-generation signal-processing systems with large capacity.

The ENZ response exists widely and can be reached in either natural plasmonic materials at its plasma
frequency or artificial metamaterials guided by permittivity homogenization. In the natural ENZ materials such as the indium tin oxide (ITO), aluminum zinc oxide, and indium-doped cadmium oxide, forced by the continuity of the displacement field at the boundary, the transverse magnetic polarized incident light undergoes a huge enhancement inside the ENZ materials with tilt incidence, whereas the transverse electric polarized light cannot provide such localization. Therefore, a bare film at its ENZ wavelength can directly perform the all-optical polarization switching. Also the film’s femtosecond-scale of the free carriers’ relaxation ensures these devices to be operated with an ultrafast processing speed. However, for utilizing the ENZ materials such as the ITO film solely, a rigorous tilt incident angle is necessary for efficient polarization switching. In addition, as the bare film can only provide the ENZ response at a single wavelength, the field localization and subsequent nonlinearity enhancement can only occur in a narrow band within less than 100 nm range around its ENZ wavelength, and therefore, the optical switching devices formed by a bare ENZ film are with relatively narrow operation bandwidth. For the artificial ENZ medium, the switching of the polarization state is originated from the anisotropy of the effective permittivity tensor. Although similar to the device with a bare natural ENZ film, it also suffers from the narrow operation bandwidth around the ENZ point and a rigorous tilt incident angle. These limitations impede the all-optical polarization control to replace the traditional control technique in a variety of photonic and material characterization applications.

In this work, we develop a femtosecond all-optical polarization switching based on the composite material system containing plasmonic nanogratings placed on an ITO film with ENZ response. The momentum compensation and the mode coupling to the ENZ film beneath, provided by the involved plasmonic grating, ensure the efficient localization of the incident light in a wide range of incident angle and broadband wavelength range. Combining such near-field localization with the large optical nonlinearity of the ITO film, we realize a red-shift of the resonance spectra for the two orthogonal polarizations simultaneously. Also this results in a distinguished change in the transmission coefficients in these two orthogonal polarizations in the broadband range, dynamically rotating the synthesized polarization angle of the output. At the same time, the ultrafast carriers’ relaxation time and low-quality factors of the plasmonic inclusions make the device to be performed within an ultrafast speed. Therefore, our designed system can be performed as an ultrafast polarization-switching device by eliminating the constraints for incident angle and operation band, and we show experimentally the transient rotation of polarization over with a pump intensity of 1.37 GW cm$^{-2}$, accompanied by a sub-ps response time, indicating a processing speed exceeding 1 Tbit s$^{-1}$. Moreover, the involved plasmonic gratings can be considered for large-scale fabrication with low cost by utilizing the existing nanofabrication technology. Our demonstrated devices open up the extensive application potentials and speed up the development of all-optical switching in the field of large-scale signal-processing system, ultrafast imaging, transient spectroscopy, and molecular dynamics.

### 2. Results and Discussion

#### 2.1. Sample Configurations

The designed device configuration in the study is schematically shown in Figure 1a, which is composed of the gold plasmonic nanogratings placed on an ITO film. The axis directions of the Cartesian coordinate are also shown in the figure. The light is at normal incidence with arbitrary polarizations and the polarization state of the transmitted wave is detected at the other side for static and transient situations, simultaneously. The period of the gold grating is 800 nm with a duty ratio of 0.5. Also the height of the gold grating is 30 nm, which is placed on a 100-nm-thick ITO film. The ITO layer was physically deposited by reactive magnetron sputtering method, and its optical permittivity was characterized by ellipsometry. The measured data was fitted through the Drude–Lorentz model, which could be described as

$$\varepsilon = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\omega_p^2}{\omega^2 - \omega_0^2 - i\Gamma_0 \omega}$$

(1)

where the ENZ response is primarily dominated by the Drude part. The parameters were fitted to be $\omega_p = 15 780.44$ cm$^{-1}$ and $\varepsilon_\infty = 3.615$. The $\omega_0$ is the plasma frequency which can be expressed as $\omega_0 = \sqrt{\frac{\omega_p^2}{m_{eff} \varepsilon_0}}$, where the $n$ and $m_{eff}$ are the free carriers’ concentrations and the effective mass, respectively. Also the permittivity of the film is plotted in Figure 1b, in which the ENZ wavelength locates at the wavelength around 1250 nm. At the ENZ wavelength, the deposited film shows an imaginary part of the permittivity around 0.39, which is determined by the damping rates $\Gamma_d$. The upper gold gratings were subsequently patterned and deposited by nanofabrication process. The gratings were fabricated in a 200 \times 200 \mu m$^2$ area. Figure 1c shows the magnified top-view scanning electron microscopy (SEM) image of the fabricated gratings. As evident from the figure, the samples are in large-scale uniformity and the contained gold nanowires in the sample are in good quality. It is worth to mention that, considering its simple configuration, this device can also be easily fabricated through the standard UV lithography technology, ensuring the potential of large-scale manufacturing with low cost, which is vital for practical utilizations.

#### 2.2. Statistic Transmission Properties and Operation Principle

The statistic transmission properties, i.e., the transmission spectra for the x/y-polarized directions, were measured through a confocal microscope setup coupled with an infrared spectrometer. A halogen lamp was utilized as the light source, and a commercial grid polarizer was placed before spectrograph for polarization-dependence measurement. To avoid the influences of the high-order diffractions, a set of spatial filters was inserted in the measurement setup. The measurement results were normalized to the substrate and plotted in Figure 2a for the two polarizations, respectively. We also made the numerical calculation of the transmission by utilizing the finite element method, where the measured ITO permittivity was used, as shown in Figure 2b. The experimental results are in excellent agreement with the numerical calculations.
agreement with the calculated model. For the \(x\)-polarized transmission, two dips appear at approximately 1150 nm and larger than 1600 nm. The formation of the two dips can be contributed to the spectrally antishift, which is formed by the mode coupling between the plasmonic resonances and the ENZ Berreman mode.\(^{[27,28]}\) For the \(y\)-polarized transmission, no distinct dip is observed, and the transmission is gradually decreased with the increase in wavelength. This illustrates that no resonant and subsequent field localization exists within this polarized incidence, and the decreased transmission is caused by the increased absorption loss of the ITO film. Considering the isotropic properties of the deposited ITO film, the measured strong polarization-dependence transmissions of the device are generated from the structural design of the nanogratings rather than from any material’s birefringence. The operation principle of the designed polarization switching relies on such artificial anisotropic and is schematically illustrated in Figure 2c. The input signal with arbitrary polarization state can be made an orthogonal decomposition along the \(x\)- and \(y\)- directions. Due to the anisotropic nature of the device, the transmission coefficients for these two directions are not the same, and thus the polarization state of the output signal is rotated compared with the input light at the static state. The dynamic all-optical operation relies on the nonlinearities of the ITO film. Under pump excitations, the free carriers in the conduction band are excited. As the utilized light is IR laser and the photon energy is lower than the bandgap, these free carriers experience the intraband transitions while the concentrations are maintained. The effective mass of the carriers undergoes an increase within the nonequilibrium distribution, and therefore, the plasma frequency shows a decrease in the nonlinear state with pumped pulse.\(^{[18]}\) The change in the plasma frequency induces the change in the ITO’s permittivity, indicating the optical nonlinearity of the ITO film. Therefore, the transmission spectra are shifted for both two orthogonal polarizations, which leads to the change in the two transmission coefficients, simultaneously. For a given wavelength, owing to the different transmission spectra for \(x\)-/\(y\)-polarizations, the change of the coefficients’ amplitudes for these two orthogonal polarizations are not the same. This can result in the polarization rotation of the transient output light compared with the static output signal. On the other hand, the device provides the on-/off-resonance properties for the two polarizations, respectively. According to the effective permittivity homogenization method for metamaterials, in the resonance region, the effective refractive index and the phase shift can be changed dramatically with abnormal dispersion relations.\(^{[29]}\) Therefore, the simultaneous anisotropic change in the amplitude and phase of the transmission coefficient for the two orthogonal polarization lead to the polarization rotation of the transient output signal, ongoing the active switching polarization properties of the device.

2.3. Calculations of the Field Localization

The involvement of the nanogratings in our design can not only help to build up the artificial anisotropy for eliminating the

![Figure 1. Device configuration and fabrication. a) Schematic of the composite coupled system with gold gratings deposited on the ITO film. The coordinate system composed of three orthometric axis is also shown. b) Measured real (\(\epsilon_1\), blue line) and imaginary (\(\epsilon_2\), red line) permittivity of the deposited ITO film. The \(\epsilon_1\) vanishes at the wavelength of 1250 nm. c) Top view of the SEM image of the fabricated sample. The sample is in uniform.](image-url)
requirements of the anisotropic nonlinear materials, but the provided plasmonic resonances can also loosen the restrictions for the angle of incidence, broaden the operation bandwidth, and lower the energy consumptions, simultaneously. **Figure 3a** plots the mode profiles of the calculated field intensity at the wavelength of 1150 nm under normal illumination. The dipole resonance of each gold nanowire can localize the incident electric field into near field, which ensures the energy to be effectively permeated into the beneath ITO layer. Thus, compared to the device with ENZ film solely, such permeation effect helps to remove the angle restrict and allows for efficient energy coupling even at normal incidence. The calculated electric-field enhancement factors, i.e., the ratios between the localized field intensity inside the ITO layer in our sample to the incident intensity, are shown in Figure 3b. As evidenced in the figure, the efficient energy coupling into the nonlinear ITO film exists in a wide range from normal incidence to a sharp tilt incidence, which provides the feasibility for practical utilizations with robust. The energy consumptions can also be lowered as the distinct energy localization in the near-IR range. The gratings and the supported plasmonic resonances also play key roles in broadening the operation bandwidth of the device. Due to the mode coupling between the ENZ mode and the resonant mode of nanogratings, the resultant spectral antishift can enlarge the region with resonance in our device, which allows for the broadband energy localization. **Figure 3c** shows the wavelength dependence of the field localizations and enhancement inside the ITO layer with x-polarized incidence, compared with the situation with a bare ITO film. The curves indicated that, within the range from 1100 to 1600 nm, the field-intensity enhancement is enlarged, exceeding 30-fold with respect to the bare ITO film in this 500 nm bandwidth. Although for the ITO films, the nonlinearity enhancement is within narrowband around its ENZ point, but the overall optical nonlinear performance of the device is also in positive relation with the field intensity inside the nonlinear materials. Therefore, the operation pump requirement of the device can be lowered in this broadband range, ensuring the broadband operation properties of our devices compared with the device with a nonlinear ENZ film solely.

### 2.4. Ultrafast Polarization Rotations

To quantificationally characterize the switching dynamics of the device, we conducted degenerated time-resolved pump–probe...
measurements, where the schematic of the experimental setup is shown in Figure 4a. The utilized source is a Ti:Sapphire laser with an output of 800 nm femtosecond pulses at 1 kHz repetition rate, combined with an optical parametric amplifier (See more details in the Experimental Section). The time-resolved pump-probe measurement is a well-known tool used to clarify microscopic electron dynamics, which is responsible for a macroscopic temporal response in optical materials. Through this method, the transient properties of the material are characterized by varying the time delay between the pump and probe pulses. The wavelength accordance for pump and probe makes the device more suitable for realistic applications. For efficiently coupling and localizing the injected light, we fixed the pump to be x-polarized, and it is also slightly obliq to be focused onto the device. This aims to isolate the transmitted probe signal for coupling into the detectors solely (See more details in the Experimental Section).

As indicated in Figure 3a, the slight tilt for pump can still maintain the large field-enhancement effect inside the ITO layer. Considering the existence of the plasmonic-ENZ mode coupling, the response speed of our switching is not only determined by the free carriers’ relaxation dynamics inside the host material of ITO, but the resonance might also influence this speed. To characterize the overall processing speed, the measurements were performed at the wavelength of 1250 nm, which is the ENZ wavelength of the ITO film. The polarizer placed before the detectors was rotated to ensure the signal intensity at static state was lower enough for improving the signal-to-noise ratio of the measurements. The result is displayed in Figure 4b, where the signal shows a temporary rise on injection of the pump, connecting with a subsequent recovery. The total response times are within the sub-1 ps range, which determines that the designed device can realize the polarization-switching function with a speed exceeding 1 THz, which cannot be reached by the existing devices where the electrons are involved. This speed is benefited from the ultrafast optical nonlinearity of the involved ITO film under pump injection, which is originated from the relaxation of the nonequilibrium free carriers in conduction band. Also at the same time, the low-Q nature of the plasmonic nano-resonators do not distinctly influence such Tbit s⁻¹ processing speed.

The polarization ellipse was experimentally characterized within a variety of incident situations. For each incident scene, we rotated the polarizer placed ahead of the detectors to different angles for intensity measurement respectively, and the signal’s Jones’ vector could therefore be fully determined based on the numerical fitting, followed by the method proposed by Yang et al. With a fixed incidence, both the static and transient state were measured respectively to retrieve the dynamic switching process. Figure 4c shows the polarization rotations at the wavelength of 1150 nm, where the values of the transmission coefficients are equal for the two orthogonal polarizations. Under pump injection with the intensity of 1.37 GW cm⁻², clear

Figure 3. Broadband field enhancements. a) Calculated near-field intensity at the wavelength of 1150 nm. The region of the ITO layer is marked by the two white dashed lines. b) Calculated intensity enhancement factors inside the ITO layer as functions of wavelength and incident angle. The energy can be effectively localized within a wide range of incident angle. c) The intensity enhancement factors of our device compared with the situation with ITO film solely.
modulations of the polarization components were observed. The polarization ellipse is relatively rotated around $12^\circ$ between the static and transient output, within the $45^\circ$ linearly polarized incidence. Also for the $30^\circ$ linearly polarized incidence, the rotation value is turned to $19^\circ$. Meanwhile, the rotation directions are also opposite for the two scenes. At the given wavelength, apart from the incident polarization, the dynamic polarization rotation angle can be also controlled by altering the power of the pump pulse. Figure 4d shows the power dependence at the wavelength of 1150 nm, where the incident polarization is fixed at $45^\circ$. The rotation angle of the polarization state of the output signal is increased by adding the pump power from 0.13 to 1.37 GW/cm$^2$, the rotation angle is monotonically increased, and no saturated phenomenon is observed. This is due to the larger optical nonlinearity of the ITO inducing the larger spectral shift of the device and the stronger anisotropy of the two transmission coefficients, obtained for increased control intensities. The diversity of the tunability is also

Figure 4. Dynamic characterizations. a) Schematic of the pump–probe measurement setup. The polarization state of the pump and probe can be tuned solely via linear polarizers. b) Temporal change in transmittance of the signal by degenerate pump–probe measurement. The overall processing time consumption of the device is less than 1 ps. c) The static and transient polarization ellipses for two linearly polarized incidence at the same wavelength of 1150 nm. d) Pump dependence of the rotation angle. e) Wavelength dependence of the rotation angle.
indicated through its operation bandwidth. Figure 4e shows the wavelength dependence of the rotation angle. In each wavelength, the incident polarization angle is fixed at 45° with the same setting of the pump power. Due to the existence of the involved plasmonic resonance, the direction of the rotation can also be tuned by the wavelength. On the other hand, in the whole measured range from 1200 to 1500 nm, the device can perform valid dynamic rotation functions, ensuring the device can be considered to be integrated into the broadband optical systems.

2.5. Discussion

Our polarization-switching device is designed considering the potential for future utilizations. From the perspective of overall performances, the ITO film with ENZ response is chosen as the nonlinear material in the device. Compared with other traditionally used nonlinear optical materials such as silicon and organic polymers, the nonlinearity of the ITO film originates from the excitation and relaxation of the free carriers inside the conduction band, which owns the fs-scale ultrafast dynamics. Therefore, the device base on this material owns the Tbit s⁻¹ switching speed, which can satisfy the requirements of the signal-processing system with high capacity. In addition, the energy consumptions and operation bandwidth are also concerned. The introduction of the gold gratings and the relatively broadband plasmonic resonances help the device for efficient near-field localization in this region, which can distinctly lower the optical power requirement and simultaneously broaden the bandwidth for device operation. Meanwhile, the fast damping rate and the relative broad resonant spectra ensures the utilization of the plasmonic resonator can simultaneously maintaining the ultrafast speed and broadening the operation band of the device. As a result, the combination of the ENZ materials and the plasmonic resonators of our design ensure the device to be operated in broadband range with ultrafast speed and low energy requirement. Also the broadband operation property ensures our device can be directly integrated with the optical networks working at telecom O band. From the perspective of diverse manipulations, the output of the device relies on a variety of degrees of freedom of the input signal, including the wavelength, polarization angle, and also the intensity. Therefore, our device can be used to encode the output signal with more diversity, which is vital to strengthen the computing capacity in the all-optical networks.

From the perspective of feasibility of large-scale fabrication, the simplicity of our device configuration, i.e., the uniform film and grating structures with large linewidth, eliminates the high requirement of the nanofabrication technology in other metamaterial-based optical devices. Our device can realize the large-scale fabrication via the widely used film deposition and UV lithography. Moreover, the gold materials can also be replaced by aluminum or other metals, such that the device can become complementary metal-oxide semiconductor (CMOS)-compatible and to be copackaged with other existing functional devices.

The device performance is also promising to be improved further. The low damage threshold of metals restricts the upper limit of the pump intensity at several GW cm⁻². As the rotation angle is in positive relation to the pump intensity, the oxide or nitride materials such as the SrVO₃ and TiN, which also support the plasmonic resonance in the infrared range, can be used for gratings with higher damage toleration. Therefore, the rotation angle can be enlarged further. It is noted that in practical utilizations, the thermal control should be taken into consideration, as the thermo-optic effect of the material has a negative influence on the performance of the device. In addition, due to the large third-order optical nonlinearity, some novel 2D optical materials including graphene, phosphorene, MXene, and antimonene have been explored effectively in the fiber-based all-optical switching. In our device, these materials can also be considered to be applied as the nonlinear components. The ultrathin thickness might enlarge the coupling strength and the excellent optical nonlinearity might lower the energy consumptions and promote the processing speed of the device further. On the other hand, as the operation bandwidth of the device is in close relation to the resonant properties of the nanostructures placed on the nonlinear materials, the homogenization gratings can be optimized to chirped configurations, which might enlarge the operation bandwidth.

3. Conclusion

In conclusion, we have proposed and demonstrated an ultrafast and broadband all-optical polarization switching based on a composite coupled system of ITO film with ENZ response and a gold plasmonic grating array. We characterized that the device can be optically manipulated with a response speed exceeding 1 THz, benefiting from the ultrafast carrier dynamics of the ITO film. The switching is experimentally operated with diverse control, including polarization angle, signal wavelength and pump intensity, within a broadband measured range from 1200 to 1500 nm. Benefiting from the near-field localization ability, we realized a 19° transient polarization rotation at the wavelength of 1150 nm with a relatively low pump power of 1.37 GW cm⁻². The results of this work pave the device as key elements for practically applying in next-generation signal-processing systems with large capacity.

4. Experimental Section

Sample Fabrication: The ITO film was deposited on silica substrates via magnetron sputtering (LAB18, Kurt J. Lesker Corp., USA). During the deposition, a mixture containing Ar/O₂ with ratios of 100:3 was used as the reactant gas. A subsequent 30 min vacuum annealing was conducted for tuning the free carrier concentrations through lattices’ defects repairs. The permittivity of the deposited film was characterized via ellipsometry (SE 850 DUV, Sentech GmbH, Germany) combined with an infrared Fourier-transform infrared spectrometer. The pattern of the designed grating arrays was written on the photosresist by electric-beam lithography and a subsequent develop and rinse process. Then, the pattern was transferred to gold using electronic beam evaporation, where the beam current was 32 mA with a deposition speed of 0.5 Å s⁻¹ and subsequent lift-off process.

Dynamic Switching Process Measurement: A home-made degenerated polarization resolved pump-probe setup was utilized to characterize the dynamic performance of the switching. The light source was a tunable infrared optical parametric amplifier (OPA, TOPAS, Light Conversion, USA) pumped by a Ti:sapphire amplifier system (Legend Elite, Coherent, USA) operating at a 1 kHz repetition rate. The power of the split pump and probe path was separately controlled by the adjustable neutral density.
filters to quantify the pump intensity and avoid the self-induced nonlinear response of the probe. The polarization was linearly controlled by the two polarizers. Meanwhile, for rotation of the polar angle of the signal, a Soleil–Babinet compensator was inserted. For time-response characterization, the piezo stage was moved continuously and the signals output from detectors was amplified by the lock-in amplifier. For determination of polarization, the piezo stage was moved to two positions where the pump and probe pulse were overlapped and separated in time domain, respectively. At each position, the polarizer placed ahead of the detectors was rotated to arbitrary four angles to measure the related intensity for characterize the polar state.

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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.

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