Reconfigurable metasurfaces that enable light polarization control by light

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Plasmonic metasurfaces have recently attracted much attention because of their novel characteristics with respect to light polarization and wavefront control on deep-subwavelength scales. A plethora of progress has already been achieved in the polarization and local wavefront of light at deep-subwavelength scales1,2. A plethora of progress has already been achieved in the development of metasurfaces, such as interfaces showing anomalous refraction or reflection3–5, the generation of vortex beams6,7, ultrathin metasurfaces8–10, novel quarter-wave plates11,12, the optical spin-Hall effect13,14, continuous harmonic nonlinearity phase control15 and high-resolution holograms16–19. However, most of the reported metasurfaces are passive, that is, the functionalities are predefined after the fabrication process and cannot be reconfigured dynamically. Substantial efforts are now dedicated to exploring metasurfaces with tunable responses; such metasurfaces would enable unprecedented applications, such as high-capacity communications, dynamic beam shaping and real-time holograms. To date, a number of techniques for such tunable components have emerged based on thermal20, mechanical21,22, optical23 and electrical mechanisms24–26. Nevertheless, above tunable metasurfaces are conventionally operated in the mid-infrared and THz spectral range and are based on light intensity modulation under weak continuous wave (CW) switching laser excitation of just a few milliwatts. Such components open the gateway toward the creation of various photonic functions, including dynamic spatial light modulation, pulse shaping, subwavelength imaging or sensing, novel quantum optics devices and real-time holograms.

INTRODUCTION

Metasurfaces, as two-dimensional equivalents of metamaterials, are ultraslim optical components consisting of artificially designed arrays of meta-atoms. Metasurfaces can impart control over the phase, polarization and local wavefront of light at deep-subwavelength scales2,3. A plethora of progress has already been achieved in the development of metasurfaces, such as interfaces showing anomalous refraction or reflection3–5, the generation of vortex beams6,7, ultrathin metasurfaces8–10, novel quarter-wave plates11,12, the optical spin-Hall effect13,14, continuous harmonic nonlinearity phase control15 and high-resolution holograms16–19. However, most of the reported metasurfaces are passive, that is, the functionalities are predefined after the fabrication process and cannot be reconfigured dynamically. Substantial efforts are now dedicated to exploring metasurfaces with tunable responses; such metasurfaces would enable unprecedented applications, such as high-capacity communications, dynamic beam shaping and real-time holograms. To date, a number of techniques for such tunable components have emerged based on thermal20, mechanical21,22, optical23 and electrical mechanisms24–26. Nevertheless, above tunable metasurfaces are conventionally operated in the mid-infrared and THz spectral range and are based on light intensity modulation under weak continuous wave (CW) switching laser excitation of just a few milliwatts. Such components open the gateway toward the creation of various photonic functions, including dynamic spatial light modulation, pulse shaping, subwavelength imaging or sensing, novel quantum optics devices and real-time holograms.

MATERIALS AND METHODS

The metasurface consists of a periodic array of L-shaped slits, each of which were cut via focused ion-beam milling through a 100-nm-thick gold film supported by a 500-μm-thick fused quartz substrate (as shown in Figure 1a). The period of the metasurface lattice is 300 nm, with an entire array footprint of 50 × 50 μm2. To combine the metasurface with the switching layer, ethyl-red powder (TCI, Tokyo, Japan) was first dissolved in ethanol to produce a solution with a concentration of ~18 wt-%. Next, the solution was mixed with polymethylmethacrylate (PMMA, Allresist GmbH, Strausberg, Germany) at a volume ratio (v/v) of 50%. The resulting polymer mixture was spin coated onto the metasurface (1500 rpm), forming an ~300-nm-thick layer. To investigate the polarization effects of the metasurface, a polarimeter consisting of a rotating super-achromatic quarter-wave plate and a Glan–Taylor calcite polarizer was used (see Supplementary Information)27,28. In our experiment, the light beam was normally incident and polarized along the x direction; in this case, ~30% of the light is transmitted through the sample (see Figure 1b, measured using a commercial microspectrophotometer (IdeaOptics Technologies, Shanghai, China)).
RESULTS AND DISCUSSION

The individual meta-atoms are chiral and anisotropic in geometry. Thus, a wave with initially linear polarization would become elliptically polarized, and its polarization azimuth rotates after passing through such a medium (as illustrated in Figure 1a)\textsuperscript{29,30}. The polarization modifications to the incident light are characterized in terms of polarization azimuth rotation $\phi$ and ellipticity angle $\chi$ in the wavelength range of 700–950 nm, as shown in Figure 1c. We achieved an azimuth angle rotation $\phi$ as high as 40° (red in Figure 1c) at 800 nm from the nanostructures, whose negative sign corresponds to the counter-clockwise rotation direction of the polarization azimuth as viewed by an observer looking into the beam. However, at the longer wavelengths, $\phi$ reverses to be positive and the azimuth rotates in the clockwise direction. In addition, the ellipticity angle $\chi$ (shown in blue) reaches $\sim 37^\circ$ at 820 nm. Moreover, its positive signs in the studied wavelength range indicate the right-handed polarized feature of the transmitted light (the end of the electric field vector rotates clockwise around the polarization ellipse, as observed against the propagation direction, see Supplementary Information for more information about polarization). Our measured results (circles in Figure 1c) could be well explained by the numerical simulations (solid curves in Figure 1c, and see Supplementary Information for more simulation details). Note that the metasurface exhibits a plasmonic absorption resonance at $\sim 800$ nm, where the azimuth rotation is also largest. This result implies the underlying relationship between the polarization effects and plasmonic resonances in our structure.

The tuning over the polarization effects relies on the modulation of the coupling situations between the plasmonic modes and the isomeric ethyl-red polymers. In our experiment, this tuning is achieved by irradiating the nanostructure by green light (532 nm, $y$-polarized, generated from a semiconductor pumped solid-state CW laser). It is well known that the changes in the dielectric properties of ambient media could efficiently modify the plasmonic response of the coupled metallic nanostructures\textsuperscript{31,32}. In our configurations, under external optical stimuli, the photoactive azo molecules structures effectively convert from the trans state to the cis state (Figure 2b). In addition, when the green light is blocked, the ethyl-red molecules return to its trans state through thermal relaxation in the dark. Such structural modifications result in changes of the molecular polarizability and finally change the refractive index of the polymer layer according to the Lorentz–Lorenz-condition\textsuperscript{33,34}. Hence, the plasmonic resonance and the resulting polarization effects of the hybrid metasurfaces are optically switched. In the experiment, the green light was 4 mW in power and was focused to a spot size of 9 $\mu$m in diameter, corresponding to an intensity of $\sim 6.3$ kW cm$^{-2}$, which is sufficient to observe large nonlinear modifications to the transmitted signal light polarization states without damaging the device (the damage threshold was measured to be $\sim 9.5$ kW cm$^{-2}$). As illustrated by Figure 2c, the most obvious effects of introducing control light are the dramatic blue shifts in the $\phi$ and $\chi$ curves corresponding to optically induced decreases in refractive index for cis-ethyl-red\textsuperscript{33} (see Supplementary Information for simulation details). Because the refractive index change of the polymer layer is determined by the number of molecules that undergo structural modification, the polarization modulation magnitudes is adjustable by changing the power of the control light. The optically induced nonlinear changes in the polarization parameters ($\Delta \chi$ and $\Delta \phi$) for various green light power levels are shown in Figure 2d. The $\Delta \chi$ curves were found to exhibit pronounced peaks between 760 and 820 nm, implying an increase in transmitted ellipticity angle for increased control light intensity. However, the $\Delta \phi$ curves exhibit peaks between 790 and 840 nm, leading to the smaller total azimuth rotation as the control light intensity increases. The $\Delta \chi$ and $\Delta \phi$ at 790 and 820 nm, respectively, as a function of green light power are given in Figure 2e. $\Delta \chi$ and $\Delta \phi$ achieve values of $\sim 16.7^\circ$ and 23.2°, respectively, for 4 mW of green light excitation. These numbers are more than one order-of-magnitude larger than the previously reported nonlinear extrinsic optical activity effects from split-ring metamaterials (only $\sim 1^\circ$ nonlinear polarization changes under $\sim 1$ GW cm$^{-2}$ excitation)\textsuperscript{35}. More importantly, rather than the self-nonlinear polarization action of one single beam\textsuperscript{35}, the device shown here demonstrates inter-beam polarization control by one beam over another, which is technically essential for realizing all-optical modulator or switching. Furthermore, in this case, the control
light could be from a low-power CW laser rather than an ultrashort laser pulse. These features would make the device much more convenient to implement in real applications.

To characterize the temporal properties of the tunable polarization effects, the green light (4 mW) was mechanically chopped, thereby causing dynamic refractive index changes in the polymer layer and resulting in dynamic modulations over the signal light. The signal light was chosen at 820 nm, where the nonlinear change of azimuth resulting in dynamic modulations over the signal light. The signal light initially. Thus, any modulations in polarization states were converted into the modulations of light intensity that leaked from the metasurface. By shifting the spectral range to shorter wavelengths for materials or chalcogenide phase-change media.

It may be expected that the operating wavelength range of our hybrid plasmonic polarization modulation frameworks could be flexibly tuned by simple adjustment of geometric dimensions of the metasurface. By shifting the spectral range to shorter wavelengths for RGB primary colors, the metasurface will be very useful in novel all-optical display devices. Moreover, by tuning the signal light wavelength to the telecommunications wavelength band, such devices could function as data encoders for interconnecting visible light communication systems and fiber telecommunication systems without any optoelectronic conversions. Note that for thin ethyl-red polymer films,
their optical performances could be degraded by phase separation and micro-crystallization inside the film\textsuperscript{11} or surface contamination caused by dust in air. An extra protection layer or covalent bonding of the azo moieties to the host polymer matrix\textsuperscript{12} could extend the lifetime of the whole device; a longer lifetime is useful in real applications.

CONCLUSION
In conclusion, we demonstrated a novel platform for creating dynamically reconfigurable optical devices that can achieve all-optical polarization control under weak optical excitation at optical frequencies. Our results could lead to breakthroughs in applications such as highly compact polarization (or intensity) modulating elements for optical computing/communications, novel optical display devices and encoders for quantum information processing by eliminating optoelectronic conversions and markedly reducing the size of the polarization modulation architectures. Furthermore, such modulation principles and schemes could be extended and applied to tune other metasurface functionalities, thereby creating various photonic functions such as dynamic spatial light modulation, pulse shaping, subwavelength imaging or sensing, novel quantum optics and real-time holograms.

CONFLICT OF INTEREST
The authors declare no conflict of interest.

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References:

1. Meinzer N, Barnes WL, Hooper IR. Plasmonic meta-atoms and metasurfaces. Nat Photonics 2014; 8: 889-898.
2. Yu NF, Capasso F. Flat optics with designer metasurfaces. Nat Mater 2014; 13: 139–150.
3. Ni XJ, Emani NK, Kildishev AV, Boltasseva A, Shalaev VM. Broadband light bending with plasmonic nanoantennas. Science 2012; 335: 427.
4. Huang LL, Chen ZX, Müllenbernd H, Li GX, Bai BF et al. Dispersionless phase discontinuities for controlling light propagation. Nano Lett 2012; 12: 5750–5755.
5. Sun SL, Wang KY, Wang CM, Juan TK, Chen WT et al. High-efficiency broadband anomalous reflection by gradient meta-surfaces. Nano Lett 2012; 12: 6223–6229.
6. Yu NF, Genevet P, Kats MA, Aeta F, Tétienne JP et al. Light propagation with phase discontinuities: generalized laws of reflection and refraction. Science 2011; 334: 333–337.
7. Yang YM, Wang WY, Moitra P, Kravchenko II, Briggs DG et al. Dielectric meta-reflacketarray for broadband linear polarization conversion and optical vortex generation. Nano Lett 2014; 14: 1394–1399.
8. Aeta F, Genevet P, Kats MA, Yu NF, Blanchard R et al. Aberration-free ultrathin flat lenses and axicons at telecom wavelengths based on plasmonic metasurfaces. Nano Lett 2012; 12: 4932–4936.
9. Pors A, Nielsen MG, Erikson RL, Bozhilovnyi SI. Broadband focusing flat mirrors based on plasmonic gradient metasurfaces. Nano Lett 2013; 13: 829–834.
10. Aeta F, Genevet P, Kats MA, Yu NF, Blancard R et al. Aberration-free ultrathin flat lenses and axicons at telecom wavelengths based on plasmonic metasurfaces. Nano Lett 2012; 12: 6328–6333.
11. Zhao Y, Ato A. Tailoring the dispersion of plasmonic nanorods to realize broadband optical meta-waves. Nano Lett 2013; 13: 1086–1091.
12. Li GX, Kang M, Chen SM, Zhang S, Pun EYB et al. Spin-enabled plasmonic metasurfaces for manipulating orbital angular momentum of light. Nano Lett 2013; 13: 4148–4151.
13. Yin XB, Ye ZL, Rho J, Wang Y, Zhang X. Photonic spin hall effect at metasurfaces. Science 2013; 339: 1405–1407.
14. Ni XJ, Kildishev AV, Shalaev VM. Broadband light bending with plasmonic nanoantennas. Nano Lett 2012; 12: 6328–6333.
15. Zhao Y, Ato A. Tailoring the dispersion of plasmonic nanorods to realize broadband optical meta-waves. Nano Lett 2013; 13: 1086–1091.
16. Li GX, Kang M, Chen SM, Zhang S, Pun EYB et al. Spin-enabled plasmonic metasurfaces for manipulating orbital angular momentum of light. Nano Lett 2013; 13: 4148–4151.
17. Ni XJ, Kildishev AV, Shalaev VM. Metasurface holograms for visible light. Nano Lett 2014; 14: 225–230.
18. Zheng GX, Müllenbernd H, Kenney M, Li GX, Zentgraf T et al. Metasurface holograms reaching 80% efficiency. Nat Nanotechnol 2015; 10: 308–312.
19. Rensberg J, Zhang SY, Zhou Y, McLeod AS, Schwarz C et al. Active optical metasurfaces based on defect-engineered phase-transition materials. Nano Lett 2016; 16: 1050–1055.
20. Gutruf P, Zou CJ, Withayachumnankul W, Bhaskaran M, Sriram S et al. Mechanically tunable dielectric resonator metasurfaces at visible frequencies. ACS Nano 2016; 10: 133–141.
21. Ee HS, Agnan R. Tunable metasurface and flat optical zoom lens on a stretchable substrate. Nano Lett 2016; 16: 2818–2823.
22. Wang Q, Rogers ET, Ghoshpuri B, Wang CM, Yuan GH et al. Optically reconfigurable metasurfaces and photonic devices based on phase change materials. Nat Photonics 2016; 10: 60–65.
23. Yao Y, Kats MA, Shankar R, Song Y, Kong J et al. Wide wavelength tuning of optical antennas on graphene with nanosecond response time. Nano Lett 2014; 14: 214–219.
24. Yao Y, Shankar R, Kats MA, Song Y, Kong J et al. Electrically tunable metasurface perfect absorbers for ultrathin mid-infrared optical modulators. Nano Lett 2014; 14: 6526–6532.
25. Miao QZ, Wu Q, Li X, He Q, Ding K et al. Widely tunable terahertz phase modulation with gate-controlled graphene metamaterials. Phys Rev X 2015; 5: 041027.
26. Basz M, DeCusati C, Enock J, Lakshminarayanan V, Li GF et al. Handbook of Optics, Third Edition Volume I: Geometrical and Physical Optics, Polarized Light, Components and Instrumentation(set), 3rd edn. New York: McGraw-Hill, Inc., 2010.
27. Ren MX, Chen M, Wu W, Zhang LH, Liu JK et al. Linearly polarized light emission from quantum dots with plasmonic nanoantennas. Nano Lett 2015; 15: 2951–2957.
28. Cai X, Huang LL, Chen XZ, Mühlenbernd H, Li GX et al. Remarkable polarization sensitivity of gold nanoparticle arrays. Appl Phys Lett 2005; 86: 183109.
29. Plum E, Fedotov VA, Zheludev NI. Asymmetric transmission: a generic property of two-dimensional periodic patterns. J Opt 2010; 13: 024006.
30. Homola J. Surface plasmon resonance sensors for detection of chemical and biological species. Chem Rev 2008; 108: 462–493.
31. Ren MX, Pan CP, Li QQ, Cai W, Zhang XZ et al. Isotropic spiral plasmonic metamaterial for sensing large refractive index change. Opt Lett 2013; 38: 3133–3136.
32. Lütkenmeyer T, Frankie H. Nonlinear and bistable properties of doped pinnma lightguides. Appl Phys A 1992; 55: 41–48.
33. Kittle C. Introduction to Solid State Physics, 8th edn. New York: Wiley, 2005.
34. Ren MX, Plum E, Xu JJ, Zheludev NI. Giant nonlinear optical activity in a plasmonic metamaterial. Nat Commun 2012; 3: 833.
35. Such G, Evans RA, Yee LH, Davis TP. Factors influencing photochromism of spiropyrans within polymeric matrices. J Macromol Sci C 2003; 43: 547–579.
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