Review

Yuanhang Che, Xiaoting Wang, Qinghai Song, Yabei Zhu and Shumin Xiao*

Tunable optical metasurfaces enabled by multiple modulation mechanisms

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Abstract: With their ultrathin characteristics as well as the powerful and flexible capabilities of wavefront modulation, optical metasurfaces have brought a new understanding of the interaction between light and matter and provided a powerful way to constrain and manage light. However, the unmodifiable structures and the immutable materials used in the construction lead to the unsatisfactory applications in most functional devices. The emergence of tunable optical metasurfaces breaks the aforementioned limitations and enables us to achieve dynamic control of the optical response. The work in recent years has focused on achieving tunability of optical metasurfaces through material property transition and structural reconfiguration. In this review, some tunable optical metasurfaces in recent years are introduced and summarized, as well as the advantages and limitations of various materials and mechanisms used for this purpose. The corresponding applications in functional devices based on tunability are also discussed. The review is terminated with a short section on the possible future developments and perspectives for future applications.

Yuanhang Che and Xiaoting Wang contributed equally to this work.

*Corresponding author: Shumin Xiao, Ministry of Industry and Information Technology, Key Lab of Micro-Nano Optoelectronic Information System, Harbin Institute of Technology (Shenzhen), Shenzhen, 518055, P. R. China; and Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, 030006, Shanxi, P. R. China, E-mail: shumin.xiao@hit.edu.cn

Yuanhang Che, Xiaoting Wang and Yabei Zhu, Ministry of Industry and Information Technology, Key Lab of Micro-Nano Optoelectronic Information System, Harbin Institute of Technology (Shenzhen), Shenzhen, 518055, P. R. China

Qinghai Song, Ministry of Industry and Information Technology, Key Lab of Micro-Nano Optoelectronic Information System, Harbin Institute of Technology (Shenzhen), Shenzhen, 518055, P. R. China; and Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, 030006, Shanxi, P. R. China, E-mail: qinghai.song@hit.edu.cn

1 Introduction

Optical metasurfaces are subwavelength layers with patterns that interact strongly with light, thus dramatically altering the light properties over subwavelength thickness [1]. In contrast to 3D metamaterials, metasurfaces offer a fundamentally new method of light manipulation based on scattering from small metallic and dielectric nanostructures and automatically have the features of relatively simple fabrication due to the planar geometries. As a result, optical metasurfaces have emerged as promising candidates to solve many bottlenecks of bulky optical elements [2].

Metasurfaces provide powerful ability of information modulating and unique ability of local field enhancement [3, 4]. Existing research has promoted the development of functional optical and electromagnetic devices, such as data storage and encryption [5, 6], polarization conversion [7], holographic imaging [8, 9], metalenses [10, 11], nonlinear photonic elements [12, 13]. Although they have indeed revolutionized our methods of management and restraint of light, most of the metasurfaces are static and focus on the specific optical response produced by the fixed structures and the immutability of the materials used in their construction. That is also the main reason that restricts their further extensive applications in various types of information control. Based on the problems mentioned previously, tunability and reconfigurability are significant factors for functional devices using active metasurfaces [14].

One way is to switch the active materials into different states through various modulation mechanisms, which often means a changeable refractive index (dielectric constant). In this way, metasurfaces with active material can be modulated under external stimuli including thermal, electrical, magnetic, optical, chemical, or electrochemical methods [15]. The active material can be regarded

as part of the metasurface structures or ambient medium around the structure [16, 17]. Strong and tunable light-matter interactions are well known to bring the several valuable effects to integrated with active materials for enhancing tunable optical response such as surface plasmon resonance (SPR) [18], Mie resonance [19], and Fano resonance [20]. When talking about the underlying modulation mechanisms of tunable metasurfaces, phase-change materials (PCMs) have shown great value in dynamic control of tunable metasurfaces through external stimuli. Another modulating method is to tune the carrier density of conductive materials (such as graphene, semiconductors, and transparent conducting oxides) through electrostatic gating or optical excitation [21, 22]. Furthermore, chemical reactions and thermo/magneto/electro-optic effects also provide approaches to alter the optical properties of active materials because of the variation from refractive indices under proper stimuli.

In contrast with the tuning method based on switching material properties, some researches attempt to tune the optical response of metasurfaces by structural reconfiguration. That is mainly because changes in the arrangement, shape and orientation of the micro/nano structures can bring about corresponding changes in the local field states and optical response of the entire system [23, 24]. In the visible, near-, middle- and far-infrared, and terahertz regions, the practical possibilities of tunable metasurfaces have been demonstrated. They can be used in many fields, for instance, beam steering [25, 26], dynamic color display [27, 28], dynamic holography imaging [29], tunable metalens [30], and ultrafast optical switches [31, 32]. Similar to the metasurfaces based on active materials, modulation speed, modulation depth (effect), cycle stability, reversibility, and process complexity are all essential factors.

In this review, we systematically introduce the recent progress in tunable optical metasurfaces. The organization of review is divided into two parts by different modulation mechanisms, including changing the properties of materials and tuning the geometry reconfiguration. We begin in Section 2 with a classification of tunable metasurfaces regarding changing the properties of constituents. Compared with previous reports, we focus on and discuss the modulation mechanisms of active materials, as well as the correlated innovative actuation and fabrication methods that determine the regulative ability. Section 3 presents some significant methods to tune the structures of metasurface and realize reconfiguration. In this section, we also introduced the combined modulation approach (metasystem) in detail. To conclude, we envision the future development of the tunable optical metasurface technology and applications.

2 Tuning by material property transition

2.1 Phase transitions

It is well known that PCMs can be applied in constructing tunable metasurfaces because these active materials will change among different states with various optical properties under proper external stimuli. Their transformation process is reversible, rapid, and nonvolatile [33]. For example, vanadium dioxide (VO_2) is one of the representative PCMs, because the lattice structure of VO_2 can change from monoclinic (insulating state) to tetragonal (metal state) to exhibit an insulator-to-metal transition under a characteristic temperature. Hence, most tuning methods of the metasurfaces based on VO_2 in existing research consist of thermal ways (global temperature), electrical heating, and optical stimulation (laser heating). Their modulation speed may also be limited to a certain extent by the modulation themselves [41].

In the field of at THz and infrared frequencies, thermal driven VO_2 metasurfaces have attracted great interest in the modulation of light amplitude [34, 35]. For example, an Ag/VO_2 hybrid split-ring resonators (SRRs) structure prepared on the Al_2O_3 substrate has shown a tuning range of resonant peak position only about 110 nm [36]. Only 4–13% tuning scope is covered in the NIR region by the Ag/VO_2 hybrid SRR [33]. When it comes to mid-infrared (MIR), the VO_2 hybrid metasurfaces can obtain modulation depth up to 80% due to their more pronounced change in the optical properties of VO_2 [35, 37]. A type of Y-shape antennas prepared on a VO_2 film (thickness = 180 nm) has been designed. The resonance of this nanostructure can be tuned from 28 to 85 °C owing to the VO_2 phase transition. The obtained tuning range of the resonance wavelength (λ) can reach about 10% (more than 1 μm when λ approaching 10 μm) [38]. When the temperature exceeds the critical point of about $T_c = 69$ °C (phase transition temperature of VO_2), the optical response (resonance peak on the reflection spectrum) of the overall structure will change abruptly due to the change in the conductivity of VO_2 . Moreover, dynamic structural color display is also known to be one of the representative works in the amplitude modulation of tunable metasurfaces. A VO_2 -Ag- SiO_2 hybrid metasurface has been fabricated where different colors could be obtained by thermally tuning [39]. However, the resonance peak of the reflection spectrum obtained by this metasurface structure is not sharp enough, so that the color saturation is relatively low, and the color change adjusted by the VO_2 transition is not apparent. Given this, a group of

researchers used FDTD software to simulate a new type of VO₂-based metamaterial absorber to improve the problems as mentioned previously, but more experiments need to be performed [40].

The phase modulation at optical frequencies of metasurface applying VO₂ phase transition has also been made good progress recently. Figure 1A shows a dielectric gap reflectarray metasurface integrated with the VO₂ unit, where great enhance of magnetic field magnitude can be figured out. The effective permittivity variation induced by electrically control Joule heating in VO₂ thin film is capable of leading great changes in the magnetic dipole (MD) resonance mode among 1520 and 1620 nm. By adjusting the dielectric region in a VO₂ film and the volume fraction of the coexisting metal, a phase shift of up to 250° can be achieved, and the modulation speed reaches a millisecond level [41]. When the insulating state of VO₂ is transformed to the metal state, the free carrier concentration will increase (showing a hysteresis loop with first-order phase transition characteristics). So that the transmittance of the VO₂ film decreases with increasing temperature, then the optical properties of the films will be changed. Similar characteristics of hysteresis curves will appear on the graph of the relationship between resonance frequency and temperature or phase shift and reflection resonance wavelength [41, 42].

Unlike thermal or electrothermal modulation methods, optical stimulation offers fine local heating of the VO₂ metasurface. The existence of this hysteresis effect also brings them to be used to construct rewritable memory cell devices [44, 45]. The programming diagram was written with a low-power laser, and the material was heated below 90 °C, then the designed phase array can be erased by decreasing global temperature. Figure 1B shows the process mentioned earlier, applying the temperature-dependent resistance of a VO₂ film. Some researches indicate that there is a close relationship between the stability of electronic hybridization and the insulator–metal transition temperature. When the VO₂ film is irradiated with an ion beam irradiation, the insulator-metal transition temperature will be changed, which is called selective defect engineering [43]. Combined with lithography, VO₂ film can be fabricated into artificially induced phase coexistence metasurfaces, such as tunable absorbers and tunable polarizers. Meta-holography and beam steering devices applying VO₂ metasurfaces also provide a good view of the application in this field [46, 47].

Based on similar principles, the reason why phase-change chalcogenides (PCCs) can be widely used for tunable metasurfaces is that these materials can achieve rapid transformation between amorphous and crystalline

phases under external stimuli (optical, electrical, or thermal). The bonding modes of these two states are various, which finally reflects a vast difference in optical properties [48, 49]. GeSbTe alloys (GST) are essential chalcogenide glasses which are widely used in this field. Comparing with VO₂, GST possesses a higher phase transition temperature ($T_c = 160$ °C), which results in a higher temperature resistance of the metasurface based on GST. In addition, the imaginary part (k) of the refractive index of the GST material is almost negligible at the MIR region. It means that the loss of the device prepared in this way will be very low so that the working efficiency will be significantly improved [50]. GST integrated metasurfaces are qualified for efficient and stable modulation of transmission or reflection amplitude in the NIR, MIR, even high-energy visible region, and recent research works continue to confirm it [51–53]. Perfect absorbers and thermal emitters based on PCCs are widely constructed for various specific applications [54, 55]. For instance, Giessen et al. reported a kind of Al–(GST-326)–Al hybrid nanoresonator performing a massive resonant absorption (>90%) in the MIR region with a spectral shift up to 25% [54]. A resonance shift about 500 nm has also been experimentally obtained in another metasurface consisting of the Au–(GST-225)–SiO₂ structure [56]. Similar to VO₂, structural color display can also be achieved when local amplitude modulation is performed on GST-based metasurfaces with higher resolution and more vivid pixels [57, 58].

The active Fano resonant metasurface with an asymmetric SRR array was reported. The researchers modulated the Q factor (8.5–31.4) and resonant wavelength (1.8–2.1 μm) through controlling the phase change process in the film made of GST because of its stability in any intermediate [59]. Figure 1C shows a conceptual description of an anapole excitation and electric dipole (ED) in a sphere of GST. Through bringing in an intermediate phase transition in the GST, a shifting of broadband mode (about 15%) between the two states mentioned previously could be achieved. There is also a work using multipolar Mie resonances modulated by the GST phase transition to achieve a multispectral optical switch [60]. As for the modulation speed, several works have been reported using GST metasurfaces to reach a nanosecond even faster level [35]. High-speed optical switching based on thin GST films also can be realized by the modulation of surface phonon-polaritons or phase transition process using laser pulses, which is regarded as one of the representative examples of rewritable devices [61, 62]. It is also an essential research direction to integrate a specific phase gradient with PCCs to achieve tunable metasurfaces based on their phase modulation. The recent research results also show their

Figure 1: Tunable metasurfaces based on phase transitions.

(A) Schematic of an electrically tunable VO_2 metasurface with simulated magnetic field magnitude around the metasurface and the curve of phase shift as the function of the applied voltage [41]. (B) The writing and erasing process on the metacanvas applying the temperature-dependent resistance of a VO_2 film [45]. (C) Conceptual diagram of an anapole excitation an electric dipole resonance in the GST sphere [60]. (D) Schematic of a planar air/GST/Au multilayer Fabry-Perot cavity with corresponding spectra of THG at $\lambda = 1450 \text{ nm}$ [66]. (E) Schematic of the transmission-type LC-based TiO_2 dielectric metasurface device [73]. (F) The relationship between the measured/simulated reflection spectra and the applied field/input polarization in the modulation of LC-based structural color [75]. GST, GeSbTe alloys; THG, third-order harmonic generation; LC, liquid crystal.

applicative values in the fields of tunable beam steering [63] and tunable metalens [64]. A $\text{ZnS}/\text{SiO}_2\text{-GST-ZnS}/\text{SiO}_2\text{-Au}$ hybrid structure has been demonstrated with 400% tuning range and 10 MHz modulation speed [65]. Besides, from the Fabry-Perot (FP) cavity metasurface structure consisting of the $\text{Au}/\text{GST}/\text{air}$ multilayer, researchers have found that a-GST obtained by phase transition significantly enhances the third-order harmonic generation (THG) efficiency (Figure 1D). That also proved

the great potential in nonlinear optics using tunable metasurfaces based on the PCCs [66].

The liquid crystal (LC) has a significant birefringence property ($\Delta n \sim 0.2$). One of the reasons why it can be used for tunable metasurfaces is that its dielectric constant (refractive index) experience changes because of the phase transition processes when it is exposed to external stimuli (thermal or electrical) [67]. At the outset, metal components and LCs are combined to obtain a tunable metasurface that

works from the terahertz frequency to the visible one [68, 69]. To obtain a higher modulation efficiency, the researchers turn attention to the application of dielectric metasurfaces, even all-dielectric silicon metasurfaces, for better optical response [70, 71]. Most of these works are to realign the nematic LCs by applying an electric field to change the resonance form of the local environment around the metasurface, to achieve the modulation effect. To reduce loss and obtain a better optical response, TiO_2 is often used to integrate with LCs in the structure [72]. Figure 1E shows a schematic of the transmission-type LC-based TiO_2 dielectric metasurface device, through controlling the LC alignment orientation under electrical voltages (3–5 V), the modulation of transmission can be reached to 65% [73].

Moreover, combining the polarization selectivity of the metasurface and the twisted arrangement induced by the electric field is also a fundamental modulation method. The representative work, in this way, is reflected in the LC-based structural color modulation [74]. Both the applied electrical voltage and input polarization of incoming light are influential factors for the final color obtained, as shown in Figure 1F. However, for the LC-based tunable structural color, there are still problems that the experimentally obtained color saturation and reflectance intensity (< 40%) are not so high, and the RGB color gamut is not wide enough. Therefore, more work is needed [75]. In addition, LC-based tunable metasurfaces have also achieved promising results in controlling the spontaneous emission [76] and beam steering [77]. It should be pointed out that the larger size of the LC-based tunable metasurface and only ms-level modulation speed limit their further application to a certain extent.

2.2 Chemical and electrochemical tuning

The use of chemical and electrochemical reactions to change the material's optical properties (such as refractive indices) combining with specific structures to achieve dynamic control of optical response is considered to be an effective and reversible construction method for tunable metasurfaces. One of the representative works is to use the hydrogenation and dehydrogenation kinetics of magnesium (Mg) nanostructures to achieve the purpose discussed previously, which was reported by Liu et al. In a previous study, a Ti/Pd–Mg–Ti hybrid nanostructure was prepared on a SiO_2 (100 nm)/Si substrate, Pd was used here for its significant difference in optical properties from Mg [79, 80]. The dynamic plasmonic color (erasing and storage) is reversible in combination with a chemical reaction

between Mg and MgH_2 [78]. The process from color generation to disappearance usually needs nearly 600 s while the recovery process takes more than 2000 s. Furthermore, based on similar principles, dynamic metasurface holograms that are considered to be meaningful for optical information processing and encryption have been demonstrated. Here, the light helicity and the chemical reaction time are the primary influence factors [81]. The researchers replaced the lower Ti in the cell structure with HSQ pillars and fabricated Al mirrors as the substrate. Through converting Mg to MgH_2 , a series of FP resonators with different cavity heights were formed in the structures. The difference from the previous work is that the original working state was a blank state, and various colors were formed after adding H_2 (Figure 2A). Furthermore, electron exposure dose (D) and filling factor (F) could be used to adjust different FP cavity modes to form different color modes, not just through the array period and spacing. The time required for the coloring process was reduced to less than 100 s, whereas the time to return to the blank state takes only 35 s [82]. There was also a structure composed of cross-shaped Teflon tubes with an internal filling chemical system in an aqueous solution. The modulation of transmittance and polarization can be achieved by injecting mixed solutions of different volume ratios to change the complex dielectric constant [83].

Inducing chemical reactions through other ways also plays an essential role in the realization of tunable metasurfaces. TiO_2 is one of the candidates due to its relatively higher refractive index and smaller extinction coefficient (k). Therefore, it has almost no optical loss in the visible range. Meanwhile, the significant change in the k value with material doping or various annealing temperature provide the possibility for large scale modulation. The TiO_2 -integrated metasurfaces can be applied to absorption enhancement (in tunable nanostructures) or energy storage [84, 85]. Tunable metasurfaces based on the transformation between TiO_2 and black TiO_2 enabled by ion implantation (H^+/O^-) was demonstrated by Xiao et al. Figure 2B shows their erasable properties in structural color. Here the oxygen vacancies and Ti^{3+} species were produced when these structures were placed in an ICP etcher with H^+ implantation. Then oxygen vacancies were recovered by doping O^- to form a stable cycle. Moreover, this metasurface possessing an ultrawide absorption band accelerated solar-based photochemical processes at a speed of 18.7 times [86]. The idea of using polymeric dyes to realize chemically tunable metasurfaces has also been proposed. One of the outstanding features of this method is to achieve polarization modulation by chemical reaction. A gold nanostructure layer combined with a thin layer of

Figure 2: Structures and optical response of metasurfaces tuning by chemical and electrochemical modulation.

(A) Schematic of the dynamic color display using stepwise FP resonators [82]. (B) Color transformation of TiO₂ metasurfaces with different cell sizes enabled by ion implantation (H⁺/O⁻) [86]. (C) Schematic of a tunable optical metasurface capable of polarization controlling at optical frequencies [87]. (D) Schematic of a device with a metallic cross-point junction to achieve electro-optical switching based on the mechanism of electrochemically conductive metallic filament formation [92]. (E) Schematic illustration of a solid-state electrochromic device to achieve dynamic tuning of gap plasmon resonances [97]. (F) Schematic of an eNPoM, showing hot spot and corresponding light scattering (solid lines) and absorption spectrum (dashed lines) with the PANI shell in different redox states (red to green: PANI⁰ to PANI²⁺) [107]. (G) In situ transmission curve obtained when CV anode scanning (-0.8 to 0.1 V) is performed on the nanohole electrode array in the NaClO₄ solution with the pyrrole monomer [110]. eNPoM, electrochromic nanoparticle-on-mirror construct.

photoisomerizable azoethyl red was prepared on a quartz substrate. A linearly polarized wave was switched to elliptically polarized with a polarization azimuth rotation ϕ appearing after passing through the nanostructure due to the chirality and anisotropy in metasurface structure (Figure 2C). The coupling between the adjustable photoisomerizable reaction of ethyl red polymer and the resonant plasma mode resulted in polarization modulation exceeding 20° under laser pumping with weakly continuous wave switching [87].

To further accelerate the modulation speed, tunable metasurfaces based on electrochemistry was considered. That is because the electric field applied across the electrodes can increase the movement rate of the reactive substances or accelerate the reaction process. A dynamic plasma nanostructure based on Au/Ag nanodomes has been demonstrated with the realization of the bionic mechanical ‘chameleon’. Its operating principle is to control the Au/Ag core-shell structure (the thickness of the Ag shell) or the variation of redox states through electrodeposition/stripping to show broad color dynamics [88]. However, during the redox repetition process, small changes in the surface topography caused by slow ion diffusion limit its modulation speed to only seconds scale and its reusability, too [89]. Optical memristive switches with electrically triggered growth of nanoscale metals can change the absorption and scattering losses of the basic plasmon mode, resulting in modulation in the spectral response [90]. For instance, a photodetection concept based on light-induced atom relocation to form metal quantum point contact was proposed, in which the effective interaction of photons and atom transport leads to an optical memristor behavior [91]. Figure 2D indicates a schematic of a device with a metallic cross-point junction to achieve electro-optical switching operated in the visible spectral range based on the mechanism of electrochemically conductive metallic filament formation inside the nanoscale gap [92]. The modulation speed of this kind of device was believed to be up to the nanosecond scale.

Besides the above metallic materials, electrochromic materials are regarded as the most representative materials in tunable metasurfaces with electrochemical dynamic control. That is because their refractive indices change significantly under the stimulation of an external electric field, and this variation is highly reversible and stable to a certain extent. Among them, inorganic electrochromic materials perform better cycle and durability, such as WO_3 , NiO , and V_2O_5 [93, 94]. An electrochromic distributed Bragg reflector (ECDBR) based on WO_3 of different porosity (refractive index) was fabricated using a GLAD electron beam evaporation method. The Bragg wavelength of

ECDBR can be determined by the deposition angle and the thickness of each layer to realize excellent control. In addition, the Bragg wavelength, reflectance, and transmittance can be dynamically modulated by applying a voltage to adjust the state of WO_3 [95]. Similar electrochromic devices aimed to enhance optical response have also been revealed in opal photonic crystals [96]. Figure 2E shows a polarization-dependent gap plasmon ($\text{Al}/\text{Li}_x\text{WO}_3/\text{Al}$) resonator device based on WO_3 . In this all-solid-state electrochromic device, the insertion and removal of Li^+ made the refractive index of Li_xWO_3 change continuously from $n = 2.1$ to 1.9 . So that a resonance wavelength shift about 58 nm ($-1.4 \sim 1.0 \text{ V}$) in the visible spectrum reflection spectrum was produced with structural color variation [97]. The WO_3/W hybrid structure enhances the reflectivity of visible light by forming an unconventional asymmetric FP nanocavity. Then working electrodes of different colors were realized by preparing WO_3 layers of different thicknesses, and the electrochromic properties of WO_3 were used to achieve the subtler electrical modulation of structural colors [98]. From the works discussed previously, the use of inorganic electrochromic materials for structural color electromodulation has certain limitations. Because the saturation of colors on the CIE chromaticity diagram is insufficient, the adjustment range of electronically controlled color is relatively small, and that the material itself shows singleness in electrochromism. For example, WO_3 and V_2O_5 can only be changed by two and three colors under electrical control, respectively [99, 100].

Organic electrochromic materials have lots of advantages such as simple manufacturing processes with low cost, relatively low power consumption brought by bistable state (doped/de-doped state), compatibility with flexible electronic products, and color-tuning versatility comparing with inorganic ones [101]. As one of the representative materials of electrochromic conductive polymers applied in tunable metasurfaces, polyaniline (PANI) has several merits such as high environmental stability, multiple colors between different states (up to more than five states with a significant difference in refractive index), and simple synthesis [102]. An array of oblate gold particles on the ITO surface with the PANI film (thickness = 100 nm) over it is reported, which was fabricated through electron beam lithography [103]. When PANI was electrochemically switched between its reduced and oxidized states, an intense blue-red shift of extinction spectrum was reversibly figured out between 633 and 571 nm . This operation can repeat more than 1 h without degradation. However, the polymer layer on those lithographically fabricated Au

arrays was just simply adhered to the substrate and the Au nanoparticles. Therefore, the spatial contact between individual Au nanoparticles and polymers was not sufficient.

A plasmonic structure combining a metal core with a uniform polymer shell named the core-shell composite structure was designed to overcome the problem mentioned previously. Its purpose is to make full use of local surface plasmon resonance (LSPR) to achieve near-field coupling amplification so that the system can perform enhanced response near a specific frequency under the external excitation [104]. A seed-mediated growth method was used to synthesize three kinds of colloidal gold crystals (nanospheres, rods, and double cones). Then the chemical reaction process of PANI happens on the surface of nanocrystals to form a shell through surfactant-assisted chemical oxygen polymerization and electrostatic attraction. Cyclic voltammetry (CV, 0.1–0.5 V) with the three-electrode system was performed to change the properties of the surrounding environment of the plasmon structure (variation of refractive index n of the single-particle PANI shell), then the scattering peaks will be moved. The measured longitudinal scattering peaks showed that the nanorod@PANI peak shift was 100 nm with a switching time of 0.001 s, and the stability was still good after 200 redox cycles [105]. Overall, the refractive index sensitivities have been increased by these three types of Au nanoparticles. Their synthetic modulating ranges of plasmon resonance cover the visible and NIR regions [106]. An electrochromic nanoparticle-on-mirror construct (eNPoMs) based on the Au–PANI–Au form has also been demonstrated. The light can be strongly confined in the gap of Au-mirror, resulting in a robust local cavity resonance (called the hot spot). Meanwhile, PANI in the gap can be modulated continuously from PANI⁰ (fully reduced) to PANI²⁺ (fully oxidized) through electrochemical control with a tuning range of the resonance wavelength about 100 nm, which corresponds to the reversible transition between green and red (Figure 2F). Furthermore, the switching time required to change the scattering peak intensity by 47% is 32 ms for oxidation, whereas for reduction, it is about 143 ms. The color contrast can reach more than 50% with ultralow energy consumption (9 fJ per pixel) [107].

In addition to the core-shell structure, researchers had also added other substances to modify the PANI films. For instance, rod-shaped nanocellulose/polyaniline nanocomposite films were prepared by in situ polymerization or opal-structured SiO₂@PANI photonic crystals [108]. Although the looseness of the modified film microstructure has been improved and the absorption of visible light has been enhanced, there is still room for further improvement in the film uniformity and color saturation. Nanoscale

PANI films can also be directly polymerized by the CV method. For example, by placing the prepared periodic Au nanoslits in an acid electrolyte solution containing aniline monomers, the final obtained hybrid PANI/Au nanostructure can be applied in optical switches of tens of milliseconds [109]. It is worth noting that because of the high loss of SPR transmission on the Au surface in the short wavelength region, the color is limited to the near-infrared region. The research of multicolor conversion display and optoelectronic devices requires further experimental experience.

Similarly, there are other valuable polymers which can also be used for tunable metasurfaces to provide probably better answers for electrochemical modulation applications. For example, a kind of arrays of nanoholes integrated with conductive polypyrrole (PPy) to perform a red-shift in SPR-enhanced transmission spectra was reported. Figure 2G implies the in situ transmission curve obtained when CV anode scanning (–0.8–0.1 V) was exerted on the nanohole electrode array in the NaClO₄ solution with the pyrrole monomer [110]. PPy has also been electropolymerized on the surface of plasmonic nanohole arrays to realize the reflection tuning of structural color with higher optical contrast [111, 112]. As an electrochromic polymer material, poly(3,4-ethylene dioxythiophene) (PEDOT) is almost transparent in its oxidized state (conductive), and deep blue in its reduced state (insulating) when it is applied a voltage of 0.6 and –1 V, respectively. A tunable device based on the gold nanoparticle arrays enveloped by PEDOT was reported. The change of LSPR with the electronic state of the polymer can be obtained. It was proved that the large red-shift from 685 to 877 nm covers nearly half of the visible spectrum in recent works that were larger than those using PANI [113–115]. There were also other works applying polycyclic aromatic hydrocarbon molecules [116] and poly(2,2-dimethyl-3,4-propylenedioxythiophene) (PolyProDOT-Me2) [109] to realize the electrochemical modulation of tunable metasurfaces.

2.3 Thermo-optical

The introduction of thermosensitive materials realizes thermally controlled metasurfaces. As mentioned in the PCM section, the thermal-induced PCMs (e.g., VO₂, GST, Si) exhibit refractive indices variation with a specific temperature. For instance, Si is the preferred optical platform for photonic devices because of its low cost and easy integration with CMOS manufacturing [117]. Rahmani et al. demonstrated a silicon nanodisk arrays metasurface,

which can be dynamically controlled by temperature. The experimentally measured scattering of the device forward and backward was recorded as a function of temperature (20–300 °C). The resonance of scattering spectra red-shifted 30 nm as the temperature increases. The sample can be regressive once the temperature is cooling down [17].

Apart from the aforementioned inorganic material, the thermosensitive polymer (organic phase-change materials, OPCM) performs a phase transition between hydrophilic state (swelling) and a hydrophobic state (collapse) at a specific temperature. As a result, the change of refractive index and volume size of the polymer leads to the variation of the resonant wavelength. Compared with inorganic material, OPCM is more stable and reliable [118, 119]. Moreover, the phase transition temperature can be adjusted by mixing various OPCM as hydrogel [121]. A thermally induced plasmonic change of gold nanodots arrays covered by poly (N-isopropyl acrylamide) (PNIPAM) was reported by Gehan et al. [120]. The integrated device indeed presented a blue shift of about 30 nm in resonance wavelength for a minimal temperature change from 16 to

52 °C (Figure 3A). Afterward, based on Gehan's work, Dong et al. developed a thermally tunable hybrid bowtie nano-antenna arrays which possess specially formulated PNI-PAM hydrogel coatings over their top surface. The experiment results indicated that the TM resonance of the coated bowtie arrays red-shifted by 16.2 nm from 22 to 42 °C, as shown in Figure 3B. By contrast, the uncoated bowtie arrays can only play a 3 nm shift [121]. The response time of temperature changed in the metasurface was only 250 ms.

In the PCM section, we have discussed that the modulation speed of thermal PCM depends on the thermal stimulation (contact heating, current heating, and radiation heating). The stimulating method of the metasurfaces mentioned previously is contact heating on the whole devices, which requires a metallic heating element and is supported by a heat sink. Close contact with the heating source may improve the tunability efficiency of the metasurface. However, it will also produce an optical coupling, resulting in optical performance degradation. Area heating as a thermal stimulation method through parts of the metasurface structure provides fast speed and avoids the

Figure 3: Tunable metasurfaces controlled by thermal, optical and magnetic stimuli. (A) Extinction spectra of the hydrogel-coated nanodots arrays at different temperatures [120]. (B) Reflection spectra of the hydrogel-coated BNAs vs temperatures [121]. (C) Simulated terahertz transmission spectrum of an ultrafast all-optical metasurface based on the carrier density [126]. (D) Schematic of a GaAs/AlGaAs heterostructure metasurface at low pump fluences [128]. (E) Faraday rotation enhanced by the EIT effect with a high transmission value, and a maximum FOM given by the overlap caused by this high transmission and high rotation [134]. (F) Schematic and experiment results of magnetic tunability through a surface of magneto plasmonic trimer nanoantennas [135].

optical disturbance from the external heater. Pierre et al. proposed a thermo-optical method by heating the Au stripes integrated by a kind of refractive index-matching polymer [122]. The long-range (low-loss) surface plasmon-polariton (LRSP) of the structure was optically symmetrical close to room temperature. Heating the Au stripes by current herein increased the temperature of the polymer around the metal structure, and induces partial refractive index changed substantially. The optical symmetric of the structure was broken as a result, and the LRSP was cutoff. The response speed of 60 μ s (heating time) and 6 ms (cooling time) were measured.

2.4 Optical and magnetic control

Similarly, tunable metasurfaces based on optical control by PCMs have been involved in the previous section. In addition to phase change, photovoltaic effect and photochromism also enable photoinduced modulation of optoelectronic devices. The optical excitation induced by pump light is one of the effective modulation methods. Here, semiconductor materials are commonly used in controlling THz waves due to their photocarrier dynamics. For instance, Si-based metasurfaces, whose conductivity can be tuned through pumping pulse, are fabricated to support tunable resonance [123, 124]. All-optical silicon on sapphire (SOS)-based tunable metasurface has been demonstrated where ion implanting technology is applied to decrease the recombination time of free carriers in Si. In this way, a 20 ps turn-on time, as well as 300 ps recovery time, was obtained with 38% transmission modulation [125]. Here the photo-excited free carriers will significantly enhance the ED resonance mode, and the transmission response of the plasma metasurface nanostructure will be blue-shifted at a pump influence of 1 mJ/cm², as shown in Figure 3C. Besides, the modulation of the optical response (especially the transmission spectrum) of the metasurface from the enhanced pumping power is also seen in the sharp Fano resonance produced by similar SOS/Al structure [126].

Moreover, the tunability in III–V semiconductor metasurfaces is also noteworthy, as they perform faster modulation [127]. A GaAs/AlGaAs heterostructure metasurface has been demonstrated to support the ultrafast all-optical tuning of Mie-type resonances in the NIR region through low pump fluence (<400 μ J/cm²), as shown in Figure 3D. The experimental results revealed an MD resonance mode blue shift of 30 nm (reflectance modulation up to 35%) with a recovery time of only about 6 ps [128]. The all-optical polarization switching devices applying In-doped cadmium oxide (CdO:In) material have also been fabricated on

epitaxial-polished MgO substrates. This as-prepared tunable perfect absorber with a high *Q*-factor performed an absolute reflectance modulation from 1.0 to 86.3% at 2.08 μ m under sub-bandgap pumping. The reflective polarizer based on this perfect absorber showed faster recovery time within 800 fs comparing with the devices mentioned previously [129].

The photochromic polymer was also one of the attractive candidates for tunable metasurfaces because of its stability and capability of strong coupling and ultrafast switching speed. An experiment of a cavity system containing a polymethyl methacrylate (PMMA) matrix with spiropyran (SPI)-based photochromic polymer and silver mirrors has proved a strong coupling resonance which can be reversibly switched [130]. The SPI-based photochromic transforms into the merocyanine (MC) form when the material exposes to ultraviolet (UV) light and reverses in visible light. An up-to 700 meV Rabi splitting of the device was observed under UV radiation, and the split peaks merge under visible light. As an exciting following up to these experiments, an Al nanodisk array coated by a PMMA layer with photochromic molecules was studied for creating rewritable waveguide [131].

Although the interaction between magnetized substance and light, which is named magneto-optical (MO) effect, is relatively weak, this effect can provide a fast subnanosecond response. Moreover, the metasurfaces based on the magneto-optical effect can provide unique polarization controllers or irreversible devices. Specifically, when linearly polarized light is projected onto the surface of a substance and propagates along the direction of the magnetic field, or it is reflected into space placed in the magnetic field, the optical polarization plane rotates (Faraday effect/Kerr effect). Through constructing a metasurface on a magnetic substance to form a resonance to strengthen these effects, and apply different magnetic fields, then tunable metasurfaces based on the MO effect will be achieved [132, 133]. Nanodisk arrays of MO materials with a sharp Fano-resonant peak were designed to enhance Faraday rotation for a high figure of merit (FOM) MO response, which was achieved by overlapping magnetic and ED resonances [134]. Figure 3E indicates that the Faraday rotation enhanced by the EIT effect is accompanied by a high transmission value of 96%, and the overlap caused by this high transmission and high rotation gives a maximum FOM = 7.35. Besides, a chiral plasma metasurface device has also been manufactured. Its basic structure consists of two Au nanodisks, which induce chirality and Ni nanodisks that provide a Faraday magneto-optical effect. By adjusting the direction and an imposed magnetic field, its transmission spectrum modulation can be

achieved, and the chiral differential transmission can be generated between the right-handed circularly polarized light and the left-handed circularly polarized light (Figure 3F). It is worth mentioning that, in the visible and NIR spectral range, researchers believe that this device can reach the limit of magnetic induction switching speeds up to 10 GHz and maintain a modulation range of 100–150% [135].

2.5 Electrically driven carrier movement

To change the carrier density of conductive materials to achieve the refractive indices change of the materials, then the optical response of the entire micro/nano structures will be influenced. That is also a feasible and standard modulation method for tunable metasurfaces. Transparent conducting oxides such as indium-tin-oxide (ITO) occupy a crucial position in the modulation application among the near-medium infrared and even visible light regions. Some recent representative works made full use of the above principle, and construct devices for different frequency ranges and verify them experimentally. A device based on a gate-tunable metasurface, which can modulate the phase as well as the amplitude of reflected light at the NIR region, was reported [136]. The demonstrated unit element of this metasurface nanostructure can be regarded as a metal-oxide-semiconductor (MOS) configuration where the gold antenna is used as a grid. At the same time, ITO was served

as a field-effect channel. The epsilon-near-zero effect happening when the real part of permittivity in ITO changes from a positive value into the negative one here will bring strong electromagnetic confinement in the accumulation layer [137, 138]. As a result, the optical performance around this interface was enhanced. Moreover, it can be electrically controlled to achieve low dissipation and high modulation speed. A phase shift of π and about 30% change in the reflectance can be figured out at about 1610 nm wavelength by applying 2.5 V gate bias in Figure 4A. It is worth mentioning that the device can be electrically controlled to modulate and switch ± 1 st order diffracted beams at frequencies over 10 MHz. In the MIR region, a similar structure with metal-insulator-metal (MIM) gap-plasmon resonator antennas has been proposed, as shown in Figure 4B [139]. The ITO state between under and over coupling was converted by moving through the critical coupling. The phase change achieved is up to 180° . However, this work can only realize the reflection amplitude modulation ratio of 35%, and the modulation frequency was limited to several hundred kHz. The main reason why the modulation depth was limited is that ITO only has carrier accumulation or depletion layers in nanometer due to the screening effect [140]. This problem could be solved by using the dual or multiple gates structure model, and more than 300° phase tuning ability can be achieved [141, 142]. These devices usually have relatively complex structures consisting of Al or Al_2O_3 because Al possesses higher reflectivity in the

Figure 4: Tunable metasurfaces based on the electrically driven carrier movement in ITO, graphene, and thin transition metal dichalcogenides (TMDs). (A) The relationship between the measured/simulated phase shift with the applied bias from 0 to 2.5 V in the gate-tunable metasurface (upper part); the relative reflectance variation measured from the gate-tunable metasurface under different applied voltages (lower part) [136]. (B) Schematic illustration of the tunable MIM gap-plasmon resonator antennas working in MIR region [139]. (C) Schematic diagram of the graphene composite metasurface working in MIR region and its corresponding reflection spectra measured under different gate bias [152].

infrared frequencies, and Al_2O_3 performs a higher relative dielectric constant [143].

The unique electronic and crystalline structures of graphene have excellent optoelectronic properties, which can be modulated by external stimuli [144, 145]. The integrated coupling of graphene and metasurfaces can achieve tunable optoelectronic devices with higher speed, amplitude, and stability [146, 147]. One of the reasons why graphene can be used for tunable metasurfaces is that its unique two-dimensional structure can substantially confine electrons in an atomic layer. Its Fermi level (E_F) can be shifted significantly with the change in carrier density through electrical gating due to its low density of states close to the Dirac point [148, 149]. The integrated tunable metasurfaces often consist of metallic or dielectric material loaded with graphene and dielectric spacer, which can be regarded as field-effect structures like discussed about previously. A thick (about a few hundred nanometers) dielectric spacer is required here, and the excessive modulation voltage required in this way is limiting the application of related devices [150, 151]. To overcome this obstacle, researchers have demonstrated a hybrid graphene metasurface modulator working in the MIR region [152]. This main characteristic of this structure is the use of a-Si as a dielectric spacer. Because a-Si has a small optical loss in the MIR region and is capable of conducting work at a particular gate voltage, in this way, it is possible to achieve a reflection change of 46.7% at a small voltage (Figure 4C) without reducing the thickness of the dielectric spacer. The modulation depth reached about 90%, with a modulation speed exceeding 1 GHz. The LRSP effect formed by the graphene integrated metallic resonator arrays generates strong absorption at the resonance wavelength. The mixed effect of heat and electromagnetic can be modulated by applying various voltages or tuning the incident angle of light to the total reflection region [146, 153]. In addition to amplitude modulation, graphene-integrated tunable metasurfaces for phase modulation also have encouraging results in recent years [150, 154].

Thin transition metal dichalcogenides (TMDs) with 2D structure also have a place in the field of electrically modulated metasurfaces with tunable optical properties. A device integrating a van der Waals heterostructure composed of hBN/MoS₂/hBN with graphene contacts was described, for efficient confinement and control of charged carriers and excitons in TMDs through electrical methods [155]. Nevertheless, as the thickness of TMD is close to the atomic limit, ohmic contact becomes more difficult. Therefore, they may need additional nanostructures to enhance the optical response further. So that the applications in optoelectronic devices based on tunable

metasurfaces can be extended [156]. There are also several other methods of electrically tunable metasurfaces based on carrier movement [157, 158].

3 Tuning by structural reconfiguration

3.1 Flexible and stretchable substrate

As for tuning the optical response of metasurfaces, structural reconfiguration is a relatively simple method. The size sensitivity and changeability of a flexible and stretchable substrate to external stimuli have been studied in recent researches. The dynamic structural color display is also one of the representative works using flexible and stretchable substrates to achieve tunable metasurfaces in the visible frequencies [159]. Polydimethylsiloxane (PDMS) is widely applied in this field as a representative material due to its high elasticity, nontoxic, and low optical losses. The tunability of combining flexible and stretchable substrates with micro/nano structures is mainly reflected in the change of the structural array period. For instance, a flexible high-contrast metastructure was demonstrated to obtain a color variation from green to orange (39 nm change in wavelength) within a periodic change of 25 nm [160]. As shown in Figure 5A, tiny Al cylindrical arrays were also fabricated on PDMS substrate with different periods, which were caused by various degrees of stretching. Then the reflective spectra were changed with different phase-matching conditions of SPR [161]. Finally, different structural color from various SPR resonance wavelength was figured out in the CIE chromaticity diagram. However, blue does not appear in this structural reconfiguration. From the perspective of the entire PDMS substrate sample, the precise direction of the total deformation generated during the stretching process may not necessarily be along the direction of the tensile force. This polarization-dependent phenomenon may be the reason why the above structure cannot meet the blue phase-matching condition when the deformation limit is reached [162]. One way proposed by Halas et al. to solve it was to perform different degrees of match compensation in the horizontal and vertical directions of the relative structural arrays [163]. In this way, the broader color gamut dynamic control of red, green, and blue had been realized in similar cube Al nanostructures with different stretching ratios (Figure 5B). Besides, there is also a work reporting that the use of chitosan hydrogel, which can swell with the change of relative humidity, brings about the change of the structure thickness.

Combining that with the MIM structure, the researchers can realize the modulation of the transmission structural color [164].

In the NIR operating wavelength, there are also several works about amplitude modulation or Raman scattering tuning of tunable metasurfaces [165]. For example, the Au plasmonic lattice grating fabricated by Kagan's group was experimentally measured to show shifts of 48 nm per 1% strain variation [166]. In addition, a kind of Au SRRs on PDMS substrates was believed to perform a tuning range up

to 400 nm of reflection peak in the MIR region [167]. For different application scenarios, micro/nano units with various shapes can be prepared on the PDMS flexible substrate [166, 168]. Noble metals such as Au should be selected for the upper layer material to meet the phase-matching conditions of SPR for resonance. Then the electromagnetic performance and optical response of the structure will be improved. Asymmetric parabolic-shaped Au arrays shape the unit element of a stretchable THz metasurface device shown in Figure 5D. This device

Figure 5: Structures and applications of tunable metasurfaces created on flexible stretchable substrate.

(A) Measured reflective spectra of the Al cylindrical arrays fabricated on PDMS substrate with different stretched lengths. The insets show corresponding SEM images [161]. (B) Working principle of a full-spectrum stretchable plasmonic device with different degrees of pitch compensation in the horizontal and vertical directions, as well as the CIE chromaticity diagram of the stretchable plasmonic device covering the sRGB color gamut with different stretching ratios [163]. (C) Schematic of a metasurface hologram on a stretchable substrate with changing from a 'frowning face' to a 'smiling face' [172]. (D) Schematic illustrations of a stretchable THz metasurface device composed of parabolic-shaped Au arrays operated by applying a stretching force [169]. (E) Schematic diagram of a device composed of a metalens and a dielectric elastomer actuator with five addressable electrodes to allow electrical modulation of the strain field in the metasurface [176].

structure had a resonant adjusting range of 0.45 THz (Q factor = 50) when exerted a deformation (60 μm) in one direction [169]. In addition, it exhibits an ultranarrow bandwidth with higher polarization dependence in transverse magnetic (TM)/transverse electric (TE) modes, which can be applied in high-quality sensors or biomedical imaging [170, 171]. Tunable metasurface hologram realized by a stretchable substrate is another inspirational work. By changing the relative position of the structure generated by the isotropic stretching of the flexible substrate, the position-dependent phase discontinuity is changed, and the resulting light wave surface is reconstructed [172]. Figure 5C shows that a hologram ‘frowning face’ was changed to ‘smiling face’ through stretching the flexible substrate with 130%. However, the ‘smiling face’ obtained was fuzzy in detail comparing with the original one, possibly due to manufacturing defects on the substrate, uneven size deformation of the graphics unit, lower transmission efficiency, or mutual interference. Thus, there is still much work to be carried out in efficient, colorful, and more stable holograms [9, 173].

Tunable metasurfaces embedded with flexible and stretchable substrate also provide an effective method to flat optics such as varifocal lenses or metalenses. There have been several works in this field in recent years. For example, a 1.7x zoom lens consisting of ultrathin flat Au nanorod arrays with the focal length tuned from 150 to 250 μm is reported [174]. Moreover, another one with over 130% focal length tuning (600–1400 μm) whose focusing efficiency is more than 50% was fabricated with a-Si and Al_2O_3 [175]. However, their modulation speed was still limited, and external equipment was required to apply strain. There is also no theoretical basis for the interaction between force field and aberration. An integrated device reported by Capasso et al. is shown in Figure 5E, which consists of five stretchable electrodes produced by single-walled graphene carbon nanotubes, a converging metalens (focal length $f = 50$ mm and diameter $\phi = 6$ mm), and the dielectric elastomer actuators substrate made of transparent polyacrylate. This device can realize the electrical modulation of the strain field in the metasurface through different stretching combination methods and the relative position changes of the five addressable electrodes. Therefore, researchers can reconstruct the outgoing optical wavefront to achieve the purpose of electronically controlling the focal length, shift, and astigmatism [176]. The single-layer device made of the integrated structure exhibits focal length modulation up to 107%. In comparison, f of the double-layer one performs 30% (tuning range = 15 mm) at 3 kV with higher average efficiency of 62.5%. Furthermore, the measured response time reached

33 ± 3 ms in the stretchable electrodes. The modulation speed could be faster through exploiting better elastomer materials or optimization of the manufacturing process [177, 178].

3.2 Shape memory effect

In addition to the aforementioned stretchable substrates, which can be combined with metasurfaces for modulation due to structural reconfiguration, shape memory materials with similar principles have also shown value in this field. Shape memory polymers (SMPs) are highly deformable materials based on the shape memory effect (SME). They can be programmed to respond to various environmental and local stimuli (such as light and heat) from a temporary shape to a permanent memorized shape with reversible network transformation [179, 180]. The combination of SMPs and metasurfaces is a novel research direction for regulation. The researchers microconfigured the architecture conversion by femtosecond laser scanning on a prestretched SMP surface [181]. In this way, the encryption and decryption conversion process of the images in the optical frequencies can be realized (Figure 6A). Moreover, a Janus high aspect ratio magnetic response microplate array (HAR-MMA) was applied in light manipulation by magnetic fields. Figure 6B shows a kind of optical shutter in which the on/off state of light can be switched by modulating the bending state of HAR-MMA [184]. A similar modulation method was demonstrated to fabricate the groove structures on the stainless steel surface to obtain different structural colors, which can be observed from different viewing angles [185]. SMP-based micro-optical devices operated through compressing, cooling, and heating process have also been reported in other works, such as quasi-structural color display and diffractive optical images. The basic structural units used are generally gratings and micropores [186–188]. However, colors produced by these devices are with poor monochromaticity and low purity and that limited their further application. Several works were reported to apply “cold” programming caused by vapors, solvents, or microwave radiation to induce the temporary and disordered configuration in macroporous SMP photonic crystal micropatterns, so as to achieve instant shape recovery under ambient condition. Experimentally, a reflection intensity changing from 90 to 50% can be obtained at the resonance wavelength [182, 183].

In addition, the thermal response SMP grating structures processed by a thermal imprinting method have been reported. A smooth stamp flattened the SMP structure into a temporary shape [189]. As shown in Figure 6C, when the

Figure 6: Metasurfaces based on SMPs tuned by shape memory effects.

(A) Encryption and decryption conversion process of the images realized by femtosecond laser scanning on a prestretched SMP surface [181]. (B) An optical shutter switched by modulating the bending state of HAR-MMA [184]. (C) The conversion process of the diffractive optical element made of thermal response type SMP and the reversible change of its diffraction pattern [190]. (D) The working principle of the IR SMP sensor [191].

laser beam passed through the diffractive optical element, a far-field diffraction pattern was displayed on the screen. By adjusting the surface temperature ($40\sim 70^\circ\text{C}$) with a hot air gun, the structure and period of the grating will change ($9\sim 3\text{ }\mu\text{m}$) and eventually return to the original state to present a similar diffraction pattern. In this process, the obtained diffraction pattern was related to temperature and time, and the modulation speed of one recovery was also reduced to 240 s [190]. The modulation speed of tunable metasurfaces based on SMP was believed to be faster through exploiting novel SMP materials or optimizing the optical properties of the constituent substances. For example, a high-sensitivity resonant infrared sensor based on a thermally responsive SMP resonator with a high-Q silicon nitride film has been illustrated. Figure 6E summarizes its working principle [191]. This device performed the response time of $210 \pm 10\text{ ms}$ in atmospheric pressure and $376 \pm 8\text{ ms}$ in a vacuum, which can be decreased to 7 ms through decreasing the radius/thickness of the SiN_x membrane.

3.3 Metasystem

In this section, we review the modulation system of one metasurface combined with other metasurfaces, micro-electromechanical systems (MEMS), and nano-electromechanical systems (NEMS) devices, which is referred to as the metasystem. Metasurfaces, as the 2D metamaterial, are very suitable to be vertically integrated through monolithic processes; the interaction of multiple metasurfaces can expand the modulation range. The metasystem consists of using MEMS control multiple metasurfaces interacting, modulating of one single metasurface by MEMS technique, applying MEMS as both actuator and metasurface to regulate light. For example, Figure 7A shows a metasystem consisting of two metasurfaces. One is a stationary metalens fixed on a glass substrate. The other one is fabricated on a SiN_x membrane as the moving metalens. The position of the moving metalens can be driven by electricity to control the focus distance between two metalenses. The tuning effective focal length achieved in

the experiment realized over $60\ \mu\text{m}$ (from 565 to $629\ \mu\text{m}$). Furthermore, the absolute efficiency of the doublet met-lenses was around $40\sim 45\%$ for the applied voltage from 0 to $80\ \text{V}$, whereas the singlets were expected to be 75% efficient [192].

In addition to position, MEMS platform also provide angle conversion for metasurface devices. Researchers

have integrated the flat plasmonic lens and a two-dimensional scanning MEMS for dynamically controlling the focusing light [193]. The angle of the lens was controlled by a MEMS platform along the two orthogonal axes by $\pm 9^\circ$, so that the incident angle changed relatively. The dynamical tuning of the reflective beam can be realized, as shown in Figure 7B. MEMS platform with a

Figure 7: Tunable metasurfaces based on metasystem.

(A) Schematic of a MEMS tunable metalens doublet (left); the mechanical resonances of the membrane at different frequencies (right) [192]. (B) Schematic of metasurface lens structure and the modulation mechanism [193]. (C) Illustration of devices and methods for the fabrication of electromagnetic metasurface on a polymer scaffold and SEM images of the fabricated scaffold metasystem [199]. (D) Schematic diagram of the tunable cantilever metasurface array (top left); the photo of the integrated chip (top right); the deflection curve of cantilever curvature under different voltages (lower left) and the measured tip height (lower right) versus applied voltage [200]. (E) Schematic of a NEMS tunable metasurface (left) and retardation as a function of applied voltage [203].

flexible substrate can be designed as a dynamically tunable metasurface controlled by manipulating the geometry of the meta-atom [194–198]. A tunable metasystem at the upper right corner of Figure 7C was fabricated on a mechanical scaffold. The scaffold, which is soft and deformable, is capable of tuning gap space between the unit cells by a single direction force F_{applied} . When the stress was exerted, the unit was stretched, and then the gap space was reduced. Thus, this scaffold metasystem can be applied in a tunable reflection spectrum. The simulation results of the strained and unstrained reflectance spectra revealed a red-shift of a resonance peak in the long-wavelength range when the applied strain was reduced [199].

MEMS platform can also be used for manipulating the deformation of the shape memory component in the metasurface. A tunable terahertz metasurface of the quarter-wave plate was integrated with electromechanically actuated microcantilever arrays (Figure 7D) [200]. The 1 μm wide cantilevers were fabricated as both an optical meta-atom and electromechanical actuator. When a voltage was applied across the cantilevers and substrate, the induced electrostatic force will pull down the free end of the cantilever. Therefore, the polarization of the transmitted light was converted. The polarization can be tuned by voltage from circular to linear. The resonance frequency for x polarization (perpendicular to the cantilever) of the metasurface for different voltages shows a red-shift from 1.05 to 0.85 THz when applied voltages from 0 to 40 V. In consideration of the relationship between working wavelength and construction dimension, NEMS technology has been adopted to achieve tunable metasurface in IR [201, 202] and visible spectral [203]. A gold nanograting metasurface integrated with a thermal bimorph actuator was designed for retardation reconfiguration, as shown in Figure 7E. The slit width between grating beams was set to be 240 nm, and the retardation modulation of 180° can be achieved at the wavelength of 532 nm.

4 Conclusion and perspectives

In this article, we have summarized the recent research progress in the field of tunable optical metasurfaces, and focus on the main mechanisms and control principles for achieving their modulation. Meanwhile, the advantages and limitations of optoelectronic devices based on them corresponding to different application scenarios are also discussed. Tunable metasurfaces show the extraordinary abilities to control electromagnetic waves through artificial subwavelength units. They also exert their unique modulation capabilities of optical response and information. Thus, a

deep understanding of the light–matter interaction and the possibility of more valuable optoelectronic technologies will be provided. Modulation speed/depth (efficiency), cycle stability, and reversibility are significant evaluation standards in the dynamic control of these metasurfaces. These aspects also vary in different control principles and mechanisms. To more clearly show the overall understanding and mutual comparison between the multiple modulation methods of tunable optical metasurfaces involved in this article, a detailed summary based on key performance parameters (such as excitation mode, working wavelength, and modulation depth/speed) is shown in Table 1. A brief description of the advantages and limitations of these technologies is also included in this table [35, 201].

Specifically, from Table 1, we can find that although the chemical reactions limit the modulation speed, the requirements for external driving conditions are relatively mild with good reversibility. In addition, electrochemical methods for modulating metasurfaces have shown their essential role in accelerating chemical reactions. Therefore, those electrochemical methods are widely used in the design of functional devices based on tunable metasurface. For instance, the electrochemical modulation of the conductive organic materials can provide more valuable meaning for flexible optoelectronic devices. For LCs, the main problems are their relatively slow modulation speed (microsecond level), larger structural volume and that they can only work in a specific linear polarization. Some of the tunable metasurfaces based on PCMs (like VO_2 and chalcogenides), semiconductors and graphene are believed to perform faster modulation speed, even reaching GHz frequency and more extensive amplitude/phase tuning range. However, large energy consumption originating from the driving conditions required and the optical losses from materials or structures themselves are issues that need to be considered in the realization of technical applications. These anxieties also exist in tunable metasurfaces based on optical modulation using laser pumping, even if their modulation speed can reach picosecond or even femtosecond levels. It is significant to point out that the modulation abilities of metasurfaces and materials are different against different incident light states. Different response results can be obtained by designing the structure to light waves in a specific state, which is also considered as a way of light control. Specifically, when the wavelength, polarization angle [204–206], spin condition [207, 208], and coherent characteristics [209] of the incident light change, metasurfaces will provide different optical responses, which can be applied to holographic imaging, structural color display, and other fields. In addition to using external stimuli to achieve variation in the optical properties of

Table 1: Comparison of multiple modulation mechanisms to achieve tunable optical metasurfaces.

Modulation mechanism	Active component	External incentives	Operating spectrum	Modulation depth/speed	Advantages	Fundamental limitations	Refs	
Phase transitions	VO ₂	Thermal	Visible to THz	10~80%/sec level	Reversible	Low heating speed	[37]	
		Electrical heating		250°/ms level	Relatively high speed	Energy loss	[38] [41]	
	PCCs (GST)	Optical stimulation	100%/1s 25%/NA 400%/100 MHz NA/ns ~ fs level	Fine local heating	Cooling rate limited	[45]		
		Thermal, optical, electrical		Efficient, stable, nonvolatile	Energy loss, diffusion affects nanostructures during cycling	[54] [65]		
		LC		Thermal, electrical	65%/NA ms level	Widely used, large birefringence with facile stimuli	Slow response, larger structural volume, specific linear polarization	[35] [73] [140]
	Chemical and electrochemical tuning	Mg/MgH ₂	Hydrogenation	Visible to NIR	100%/35~100 s	Mild driving conditions with good reversibility, low cost	Modulation speed, operating wavelength, tuning range	[82]
		Au/Ag (shell)	Electrochemical		150 nm/s level		[88]	
Memristive switches			NA/ns level		[92]			
WO ₃ , V ₂ O ₅			58 nm/ms level		[97]			
PANI, PEDOT			100~192 nm/ms level		[107] [113]			
Thermo-optical	PCM	Thermal, optical, electrical	Visible to THz	75 nm/NA	Reversible, sensitive	Low reversible speed, tuning range	[17]	
	OPCM			3~30 nm/250 ms			Variable phase transition temperature	[120] [121]
Optical and magnetic control	Si	Photo-induced, optical pumping	Visible to THz	38%/20~300 ps	Very fast speed, nonvolatile	Power consumption, modulation depth	[125]	
	GaAs/AlGaAs			35%/6 ps			[128]	
	CdO:In			86%/800 fs			[129]	
	Photochromic polymer	32%/6 fs	[130]					
	Au/Ni	Magneto-optical effect	Visible to NIR	100~150%/10 GHz	High speed/depth	Weak interaction	[135]	
Electrically driven carrier movement	ITO	Electrical	Visible to MIR	30%/10 MHz	Fast speed, good stability	Power consumption, modulation depth, operating wavelength	[136]	
	Graphene			35%/hundreds kHz			[139]	
			NIR to THz	90%/1 GHz			[152]	
Structural reconfiguration	Stretchable substrate	Mechanical	Visible to THz	0.45 THz/NA	Simple operation, low cost, extensive tuning range	Modulation speed, stability, difficulty in synthesis	[169]	
	SMPs	Shape memory effect		107%/33 ms			[176]	
	MEMS, NEMS	Electromechanical		15%/ms level	High integration, movable, extensive tuning range	Complex process, high cost	[182] [191] [192]	
				60 diopters (4%)/0.1 ms			[203]	
				Retardation 7.6°/NA			[200]	
			0.2 THz/NA					

MEMS, microelectromechanical systems; NEMS, nanoelectromechanical systems; SMP, shape memory polymer.

active materials, the use of dimensional changes brought by structural reconfiguration is also an effective method to achieve tunable metasurfaces. Those methods for realizing variation of shape and size usually include mechanical deformation (e.g., piezoelectricity and magnetoelectricity), SMEs, and MEMS [210]. However, the difficulty in synthesis of proper SMPs, as well as the complex process and high cost in MEMS are the significant problems that need to be solved urgently.

In short, much work is needed to improve the modulation speed/depth and cycle stability of tunable metasurfaces, as well as reduce energy consumption and optical losses. It is worth noting that we need to discuss the actual results of those tunable metasurfaces in combination with different specific application scenarios. After all, if one scene fails to meet the requirement, it may be able to meet the primary bottom line of another one. Moreover, we believe that there are lots of practical issues that need to be addressed. They include the large-scale unified control of multiple active cells with tunable metasurface structures, and the subsequent issues of cross-cell crosstalk and additional energy consumption, as well as compatibility with current CMOS semiconductor fabrication processes. All these are vital steps in large-scale practical applications based on tunable metasurfaces. Multidisciplinary research involving material science, physics, chemistry, optics, computer science, as well as the design and implementation of flexible and high-performance optoelectronic devices, are the critical trends in this field.

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