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REVIEW

A historical overview of nano-optics: From near-field optics to plasmonics

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Nano-optics is an emergent research field in physics that appeared in the 1980s, which deals with light–matter optical interactions at the nanometer scale. In early studies of nano-optics, the main concern focus is to obtain higher optical resolution over the diffraction limit. The researches of near-field imaging and spectroscopy based on scanning near-field optical microscopy (SNOM) are developed. The exploration of improving SNOM probe for near-field detection leads to the emergence of surface plasmons. In the sense of resolution and wider application, there has been a significant transition from seeking higher resolution microscopy to plasmonic near-field modulations in the nano-optics community during the nano-optic development. Nowadays, studies of nano-optics prefer the investigation of plasmonics in different material systems. In this article, the history of the development of near-field optics is briefly reviewed. The difficulties of conventional SNOM to achieve higher resolution are discussed. As an alternative solution, surface plasmons have shown the advantages of higher resolution, wider application, and flexible nano-optical modulation for new devices. The typical studies in different periods are introduced and characteristics of nano-optics in each stage are analyzed. In this way, the evolution progress from near-field optics to plasmonics of nano-optics research is presented. The future development of nano-optics is discussed then.

Keywords: nano-optics, near-field optics, surface plasmon, plasmonic modulation

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1. Development of nano-optics

Nano-optics, a field of studying optical physics at the nanoscale, is an important component of mesoscopic physics. It started in the 1980s with the fast development of instrumentation of characterization and fabrication for nano-devices. Nowadays, nano-optics has expanded in diverse material systems from metal to dielectric, and hybrid materials. A huge change happens in the research focus of nano-optics. Nano-optics as an important component of nanotechnology, has infiltrated many crucial scientific areas, such as optical waveguiding, optical sources at nanometer scale, integrated circuits, chipmaking, fiber communication, and novel materials.[1–5] It is important to investigate the development path of nano-optics for tracing future development directions in this area.

A large amount of work and reviews have discussed the development progress of nano-optics.[6–9] However, discussions about the transversion of nano-optics study in general pictures are few. In this review, the development process of nano-optics is mainly focused on the transversion from near-field optics to plasmonics. The derivation of near-field optics is retrospected and the further development of surface plasmon is reviewed. Furthermore, the emergence of algorithms in recent years has influenced the field of nano-optics. The future development of nano-optics is further discussed.

2. Derivation of near-field optics and surface plasmon in nano-optics

2.1. Breakthrough of optical diffraction limits: the development of near-field optical microscopies

With the advancement of science and technology towards small-scale and low-dimension, the special scale of physics entered micro-nanometers in the 1980s. Mesoscopic physics as a new branch of physics was first proposed by N. G. Van Kampen in 1976.[10] The prefix meso- refers to the scale between the micro and macro levels. Mesoscopic optics also emerged during that period, and efforts had been paid to make traditional optics enter a higher resolution stage, i.e., in the range of smaller than the half of optical wavelength. The scale of optical resolution as high as tens of nanometers became the core objective in mesoscopic optics. As optical physics enters the scale that is approximate to the diffraction limit, nano-optics as a new direction in nanoscience and nanotechnology emerges according to the requirements of the new spatial scale. Compared with macroscopic and microscopic optics, nano-optics exhibits extremely different special optical properties because the structural dimensions usually possess the same order as light wavelength. Complex and rich physical phenomena in optics occur at the nanoscale.

The invention of the most important instrumentation of

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nano-optics can be traced back to the 1980s and is led by the invention of scanning near-field optical microscope (SNOM). It is an optical scanning probe microscopy technology developed based on the principle of near-field detection. In 1984, SNOM, which can detect and characterize near-field information with a higher resolution over optical diffraction limit, was first invented by Dieter Pohl.[11] The principle of SNOM referred to the suggestion proposed by Sir Edward Hutchinson Synge in 1928. The statement declared that a device with an aperture placed on an illuminated surface, near-field light could be detected with images beyond the diffraction limit. In the 1980s, Dieter Pohl was employed by International Business Machines Corporation (IBM). In 1982, the scanning tunneling microscope (STM), which was awarded the Nobel Physics Prize, was invented by his colleagues at IBM. This inspired Dieter Pohl who has a background in laser research to develop optical techniques. Through pressing a corner of a metal-coated quartz crystal against a glass plate to achieve the aperture in Synge's proposal, Pohl and his colleagues finally detected a faint light transmitted at the near field. A SNOM image at visible wavelengths was obtained for the first time with a resolution of 20 nm.[12]

It is clear that the breakthrough of optical diffraction limits was directly related to near-field optics. The SNOM image with a resolution over diffraction limits is actually detected at near-field. Therefore, the early development of nano-optics is mainly around near-field detection and imaging. Compared with far-field optics, near-field optics mainly studies optical phenomena within the range of tens to hundreds of nanometers from a light radiation source. Within this range, electromagnetic field distributions exhibit anisotropy and unique characteristics due to the specific lattice arrangement and nanostructures. Near-field optics includes radiation field information and non-radiative field information. Radiation field information is a field component that can be transmitted outward while non-radiative field information refers to the field component that is confined on the surface samples and rapidly decays at a distance. Therefore, near-field optics is different from far-field optics. However, near-field optics is not independent from far-field optics. On the contrary, near-field optics can effectively reflect and predict far-field responses. In other words, the near-field signal and the far-field response are inseparable in optics. The electromagnetic distributions in the near-field can successfully correspond to the far-field optical signals. Benefits from that, the near-field responses of light radiation modes are widely used in predicting and explaining spectral behaviors in the far-field.[13] In near-field optics, the distributions of electromagnetic fields in physical space are mainly concerned, including three major aspects: the local density of states (LDOS), the distribution of charge density, and the distribution of electromagnetic field intensity. These parameters are closely dependent on the radiation source and can achieve various optical responses and functions in the far field, which implies broad application prospects based on near-field optics.

The invention of SNOM marked the first breakthrough in the diffraction limit resolution of the optical microscope by humans. Since then, near-field optics, rooted in the birth of conventional optics, has emerged in the field of optics. Thereafter, different configurations appear. Photon scanning tunnelling microscope has been developed in 1986, on the basis of detecting signal through obtaining scattering of photons or fluorescence.[14] Subsequently, it has been further developed to probe the evanescent field for the near-field optical imaging.[15] In the early 1990s, single-mode optical fibers were applied as optical probes and promoted near-field microscopy as a new type of optical instrument for observing and studying the appearance, morphology, and intrinsic properties of sub-wavelength objects. Besides optical imaging, near-field microscopes were also extended to spectroscopic analysis. For example, near-field Raman spectroscopy was achieved based on a fiber optic probe with a subwavelength aperture.[16] However, although SNOM has obtained achievements in nanoscale resolution, its drawbacks, such as significant sample damage and limited observation of the surface, limit its applications for observing biological samples. Therefore, efforts have been made to develop super-resolution imaging technologies which are suitable for imaging biological samples. Around that time, studies of fluorescing molecules coincidentally provided new imaging tools for molecular biologists. Therefore, in the following few years, probing and observing based on near-field optics were widely applied in the fields of physics, chemistry, biology, medicine, and information at the nanoscale. Specifically, Stefan Hell at the Max Planck Institute for Biophysical Chemistry in Germany proposed a method to break through the diffraction limit based on fluorescence for deep imaging in 1994, and demonstrated fluorescence excitation and quenching microscopy. In this method, molecules can be excited to fluoresce by light at a specific wavelength and be suppressed by a different wavelength. A standard round beam and a doughnut-shaped beam are used as excitations, of which the central region can achieve over diffraction-limit dimensions. The stimulated emission depletion (STED) microscopy was further developed by Hell based on this method in 1999. The relevant studies further led to the Nobel Prize for Chemistry in 2014.[11] Besides Hell, Eric Betzig at the Howard Hughes Medical Institute and William Moerner at Stanford University were awarded the Nobel Prize together due to their important contributions to photo-activated localization microscopy (PALM) and stochastic optical reconstruction microscopy (STORM) in 2006.

2.2. Explorations of higher resolution: the derivation of surface plasmon

As discussed in Subsection 1.1, SNOM can detect and characterize near-field information under optical diffraction limits. Therefore, the core task of the SNOM probe is to capture the radiation from a selected dipole while discriminating the radiation from neighboring ones. Since SNOM was first invented, efforts have been made to improve the efficiency of SNOM probe further. In near-field optics, the optical information and energy can be concentrated on metallic interfaces or in small metallic nanostructures due to a kind of near-filed interaction mode called surface plasmon.[17] Owing to that, metal-coated probes that have an aperture at the tip have been developed, as shown in Fig. 1(a). Light can travel down to the SNOM tip and go out from the nanoscale aperture at the end then illuminate samples at near-field. Apertureless tips kind SNOMs with atomically sharp metal tips have been further developed, in which the field is enhanced at the end of the tip according to resonant oscillations of surface plasmons on the probe surface, as shown in Fig. 1(b).[11] The electromagnetic field can be concentrated and amplified in a highly localized area at the tip of SNOM. When this enhanced near-field scatters from the sample, the interactions of the sample signals and tip are intensified, which can produce a much stronger signal with clear images. Therefore, through a tip with a metal coating to scatter the incident light and forming nano-optical cavities, the local light field can be enhanced and controlled by changing the local electromagnetic environment under the tip owing to the near-field interactions induced by surface plasmon. The throughput of photons is improved in this way. The apertureless tips kind SNOMs further led to the development of other spectroscopy techniques. For example, surface plasmons were considered to work as Raman signal amplifiers.[18] The surface-enhanced Raman spectroscopy (SERS) and the tip-enhanced Raman spectroscopy (TERS) were both invented based on plasmonic resonances.

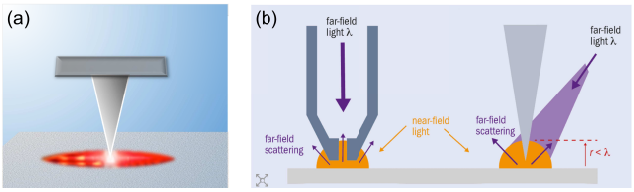


Fig. 1. The schematics of SNOM. (a) The schematics of SNOM with metal-coated probes. (b) The comparison of SNOM with (left) and without (right) an aperture.[11] Reproduced with permission from Ref. [11].

Detecting the evanescent waves of the sample, which exists as surface plasmons, has also become an efficient method to characterize subwavelength structures and the corresponding optical information by SNOM. Surface plasmon, rising along with near-field optics, mainly concerns the confinement of electromagnetic fields into subwavelength dimensions, and further becomes vital research aspects in nano-optics.[19] The condition for the existence of surface plasmon is that dielectric function ε has a negative real part. Resonances of surface plasmons are well pronounced when the losses are small enough.

In retrospect of the history of surface plasmon, the term plasmon was named by David Pines in 1956 to describe the phenomenon of plasma oscillations in gas discharges.[20] He found that fast electrons traveling through metals experienced energy loss, which can be attributed to the collective oscillations of free electrons in metal. In 1974, the term surface plasmon polariton (SPP) was first introduced by Stephen Cunningham to emphasize the coupling between the oscillating electrons of surface plasmons and electromagnetic fields.[21] Due to the near-filed coupling, SPPs are electromagnetic waves propagating along the interface of metal and dielectric media, they are confined evanescently in the direction perpendicular to the interface. In other words, an enhanced optical near-field of sub-wavelength dimension is formed as propagation modes on the interface, as shown in Fig. 2(a).[20] Such waves can be guided by metallic nanostructures with a size smaller than the optical diffraction limit and can be characterized by SNOM. SNOM can image surface plasmon excitations and the field states with high spatial resolution.

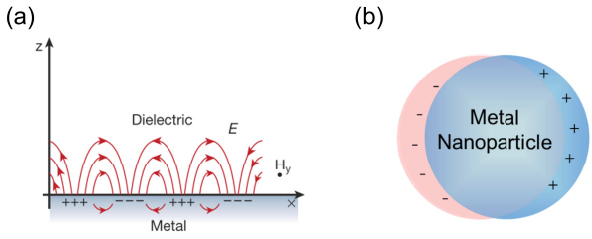


Fig. 2. The SPP modes and LSP modes in surface plasmons. (a) SPPs exist as propagating electromagnetic waves on the interface of the metal and dielectric media, which are confined evanescently in the direction perpendicular to the interface.[20] Reproduced with permission from Ref. [20]. (b) LSPs exist as localized modes confined into volumes smaller than the diffraction limit and exhibit strong near-field enhancement.

There are some important properties of SPP. SPPs only exist for TM polarization, SPPs possess non-linear dispersion effects in metallic nanostructures, and SPPs always decay with an energy attenuation length. Later on, Pohl *et al.* raised another important aspect of SPP, i.e., optical antenna in nanostructures. In 1998, Thomas Ebbesen published their new results based on principles of SPP, the extraordinary optical transmission through subwavelength metal apertures was observed, which was not possible earlier.[22] In 2000, Sir John Pendry predicted that a thin metallic film may serve as a perfect lens[23] and later on the transform optics[24] theoretically. It was shown that SPP, or interaction of light with metallic nanostructures may provide a vast new optical property. To emphasize the importance of surface plasmons, and their interplay with electrons, photons, and excitons, plasmonics was proposed as a new terminology by Brongersma and Atwater in 1999.[25]

Besides SPP, another existent mode of surface plasmons is localized surface plasmon (LSP). Different from the propagating plasmonic modes of SPP, LSP exists as localized plasmonic modes. LSP in metal nanoparticles, electromagnetic energy can be confined into volumes smaller than the diffraction limit $(\lambda_0/2n)^3$, where $n = \sqrt{\varepsilon}$ is the refractive index of the surrounding medium. This high confinement leads to a strong field enhancement, as shown in Fig. 2(b). Many applications can be developed based on the strong near-field enhancement and the localized confinement effect of LSP, such as optical sensing, energy conversion, etc.[26] SNOM is also an efficient tool for studying LSP in metal nanoparticles or metal nanostructures (meta-structures). In these experiments, the light coming from the sub-wavelength aperture of a fiber probe can be collected, and the local field distribution can also be imaged.

3. Attention attracted to near-field properties of surface plasmon

3.1. Further development of surface plasmons in SNOM exploration: nano-antennas

It is known that to improve the SNOM probe as effectively as possible, the tip of SNOM should satisfy two conditions. One is capturing the radiation from a selected dipole efficiently and the other is discriminating the radiation from neighboring ones distinctly. However, although the scattering tip probe can satisfy the first condition due to the efficient near-field enhancement, the performance on discrimination of the target detection source is relatively weak. On the other hand, the aperture near-field optical probe has the advantage of satisfying the second condition. In seeking a probe that satisfies both conditions, the concept of the optical antenna rooted in near-field optics nanoantenna, whose dimensions have to be reduced to sub-micrometric size, was proposed for its potential as SNOM probes and other applications of near-field optics. Furthermore, Dieter Pohl proposed that the near-field optics can be seen as an antenna problem.[27] He pointed out that there are similarities among the traditional telecommunication antennas with electromagnetic properties of fluorescing molecules and small scattering objects. Therefore, the antenna theory could be applied in near-field optics.

Nano-antennas are developed based on plasmonic emitter and receiver.[19,28] The objective of optical antenna design is to optimize the energy transfer between the source/receiver and radiation fields, as shown in Fig. 3(a). Recently, the term optical antenna has been defined as a transducer between free radiation and localized energy.[19] The aim of the probe of a near-field optical microscope is to capture the radiation from a selected dipole and discriminate the surrounding radiation with the highest efficiency. For a suitable optical antenna, its dimension should be reduced to a sub-micrometer scale. To adopt the antenna theory to optical frequencies, the metallic probe should retain metallic properties, i.e., their plasmon frequency should be distinctly above the operating frequency, such as Au and Ag in the visible regime and Al in the infrared regime of the spectrum.

In 1985, John Wessel first proposed that a gold particle could form an antenna.[29] The first demonstration in experiments was developed by Dieter Pohl and Ulrich Fischer in 1989. They studied plasmon modes in small protrusions on a flat gold film by SNOM and observed extremely narrow resonances in terms of scattering intensity versus distance. The antenna of the SNOM which they approved was this small, illuminated protrusion with a diameter of 50 nm to 150 nm on the flat gold film, as shown in Fig. 3(b). They further proposed that the tip-like probe antenna facilitates an approach to the sample surface should be an important consideration for practical applications.[30] In the following years, optical antennas were applied in near-field microscopy and spectroscopy.[31] Bow-tie antennas were further proposed as near-field optical probes in 1997[32] and performed with atomic force microscopes (AFM) by Dieter Pohl *et al.* in 2005, as presented in Fig. 3(c).[33] Resonant antennas for near-field optics had been discussed, which are used to improve the radiation efficiency of a molecule, i.e., half-wave dipole probe, monopole probe, full-wave dipole probe, as well as broadband antennas for near-field optics.[27]

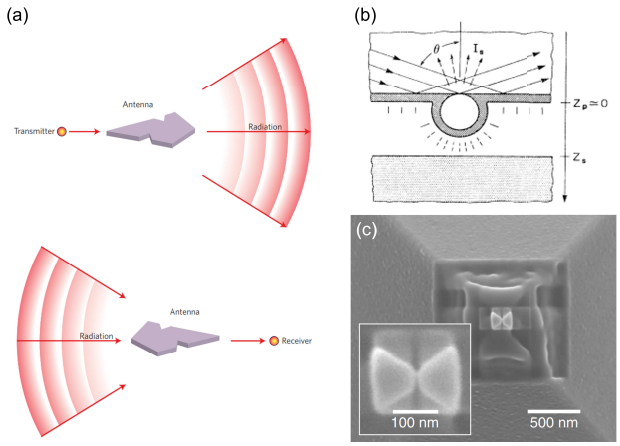


Fig. 3. Nano-antenna. (a) Transmitting antenna and receiving antenna. Arrows represent the directions of energy flow.[19] Reproduced with permission from Ref. [19]. (b) SNOM probe and the sample arrangement of the antenna.[30] Reproduced with permission from Ref. [30]. (c) Bow-tie nano-antenna applied at the apex of an AFM tip.[33] Reproduced with permission from Ref. [33].

So far, it has been emphasized that optical antennas can convert propagating optical radiation into localized energy, and vice versa.[19,28] A series of optical antenna geometries are proposed and studied, including bow-tie antennas, half-wave antennas, monopole antennas, particle antennas, and gap or slot antennas. The control and manipulation of optical fields at the nanoscale by nano-antennas possess great opportunity for enhancing the performance and efficiency of photodetection, light emission, and sensing. Relevant applications have

obtained significant progress, such as photodetection and photovoltaics, nonlinear signal conversion, and information processing. Furthermore, nano-antenna based on surface plasmons provides opportunities for super-resolution imaging in the near-field microscopy.[34]

3.2. Early applications proposed based on surface plasmons: optical transform

Since the extraordinary near-field properties of surface plasmon can overcome the deficiency of SNOM, surface plasmon has attracted attention in the research area and gradually occupied core positions in nano-optics. Early applications in nano-optics have been proposed based on surface plasmon. Optical transform systems, including perfect lens and optical cloak, are typical application aspects of surface plasmon independent from SNOM exploration.

The pioneer idea of superlens was proposed by Pendry in 2000.[23] When it comes to optical lenses, the operation has been well understood according to conventional optics that the light is focused by curved surfaces through virtue of the refractive index contrast, which possesses limitations of focusing light onto an area smaller than a square wavelength. The proposed superlenses based on SPP can overcome the limitations of traditional optical lenses, of which the sharpness is maintained regardless of the wavelength of light. In the conventional optical imaging system, the sharpness of the image is always limited by the wavelength of light, which causes chromatic distortion, this has been an unsolved obstacle for the optics engineers. Furthermore, when a light is propagating, the low spatial frequency components propagate into the far-field and can be collected by a conventional lens. In this case, the high spatial frequency components, which contain subwavelength structural information of the object, decay evanescently away from an object, which limits the optical resolution since the high-resolution spatial information is lost. However, when a slab of negative refractive index material is used, all Fourier components of a 2D image are focused, even those that do not propagate in a radiative manner. Figure 4(a) shows the principle of an optical perfect lens. A negative refractive index medium bends light to a negative angle with the surface normal. Light formerly diverging from a point source is set in reverse and converges back to a point. Released from the medium the light reaches a focus for a second time, which is not able to be realized in conventional optics according to Snell's law of refraction.

The key condition for this medium is that both the dielectric function, ε, and the magnetic permeability, μ, should be negative. Note that dielectric function $\varepsilon = -1$ is exactly the condition needed for a surface plasmon to exist: there is a link between focusing action and the existence of well-defined surface plasmons. This design can get perfect focus without the constraints of the wavelengths. This is achieved by using the negative refractive index material, inside which light restores not only the phase of propagating waves but also the amplitude of evanescent waves. This original idea stimulated many exotic optical designs. The possibility of focusing magnetic fields into sharp images in the GHz band was also predicted, which might find application to separate magnetic objects from nonmagnetic media. Pendry's initiative was also adopted in super-resolution imaging, where nanoparticles and molecules are used as the imaging media.

To further extend the application of surface plasmon, Pendry *et al.* developed an optical cloak, called transformation optics and subwavelength control of light in 2012. In principle, this technique could be used for optical cloaking, negative refraction, and energy harvesting.[24] Recent developments in optics and materials have seen interactions between light and matter move to the subwavelength regime. Figure 4(b) sketches the principle of transformation optics. The field lines of a uniform electric displacement field are compared with the lines in a distorted space. It can be seen that in such a distorted field, i.e., a bent rubber sheet, the field lines form a peculiar shape. Transformation optics provides a general platform for controlling and manipulating light, literally transforming the field of optics. By using materials design, the electric and magnetic components of the light can transfer their coordinate system so that new subwavelength optical phenomena arise. It is shown that transformation optics can be applied to many fields: such as the adaptation of computer programs to varying geometries, negative refraction devices, perfect lenses, optical cloaks, and subwavelength optical devices.

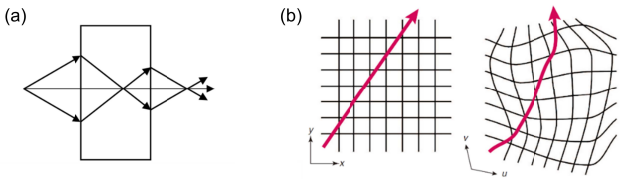


Fig. 4. Optical transform system based on SPPs. (a) The principle of superlenses. A negative refractive index medium can bend light to a negative angle with the surface normal. Light can be set in reverse and converge back to a point. Light released from the medium reaches a focus for a second time.[23] Reproduced with permission from Ref. [23]. (b) The comparison of field lines in a uniform electric displacement field and field lines bent in a deformed space after transformation.[24] Reproduced with permission from Ref. [24].

4. Further development of surface plasmon

4.1. The transition of research focuses on nano-optics: from pursuing higher optical resolution to near-field manipulation

The success achieved by the invention of SNOM marks the optical microscope resolution strides into extent over the optical diffraction limit. Broad applications have been devolved in research fields such as surface plasmon resonance, quantum device luminescence, and interface electron

tunneling.[35] As the scale of optical detection and characterization entered tens of nanometers, the research focuses on improving the optical resolution gradually develops to the bottleneck. Photonic devices, which possess three orders of magnitude faster than electronic devices and hold great potential in meeting the needs of high speed and small size, have entered the vision of scientific researchers. In this context, the major concerns in nano-optics have gradually turned to study near-field interactions and photonic applications.

When nano-optics developed to the 2010s, the research concentration of near-field characterization and enhancement in plasmonics further progressed to near-field manipulations. As discussed before, surface plasmons possess abilities to confine electromagnetic fields within tens of nanometers and enhance light significantly in the far-field under plasmonic resonance. They have given rise to abundant methods to manipulate light in the near-field. SPP possesses advantages as propagation and waveguides, while LSP offers a way to modulate light with the localized field. The extraordinary near-field enhancement and super high resolution of surface plasmon provide a beneficial platform to enhance and manipulate light at the nanoscale.[34,36–38] Besides, with the development of nano-processing techniques, artificial nanostructures with precise structures at the nanoscale supporting different near-field modes occur. As introduced in Section 2, near-field optical responses are closely related to the nanostructures. Surface plasmons can present as a special distribution of near-fields in artificially designed nanostructures to interact with light or materials.[39–41] Therefore, different plasmonic nanostructures can achieve different optical responses and functions to meet various physical scenarios and have led to a large number of applications on the basis of near-field manipulation, such as plasmonic waveguide,[17] nano-antennas,[19,28] strong coupling system,[42] optical tweezers,[43] bio-sensing technology,[44] photocatalysis,[45] and so on. The potential of surface plasmons for highly integrated photonic design occupies the center position in plasmonic research. The flexibility of plasmonic design becomes a powerful foundation for supporting diverse functions in nano-optics, and further expands the application prospect of plasmonics.

As the research focus of nano-optics transited from pursuing higher optical resolution to near-field manipulation, two major research areas have appeared in plasmonics. One research area focuses on new photonic phenomena and functions based on the superior plasmonic nanostructures. Metasurface is a typical representative of this area. The other pays attention to controlling other material systems by plasmonic modes, especially two-dimensional (2D) materials prospering after 2010. The manipulation of 2D materials properties based on near-field effects of surface plasmon has become quite popular in recent years. Furthermore, these two areas also have big overlaps. Works based on the manipulations of 2D materials by metasurfaces are massive. In this section, we discuss the recent development of plasmonics in two main aspects. One is the development of nano-processing techniques, the other is the modulation properties of surface plasmons. Metasurface and surface plasmons & 2D materials (especially 2D semiconductor materials) are selected to represent the nano-processing progress and the modulation methods of surface plasmons, respectively.

4.2. Metasurface with specific design

Metasurfaces are artificially manufactured periodic nanostructures, which belong to planar optical elements. Metasurfaces with different structures can support different plasmonic modes and achieve abundant optical responses. Benefits from near-field advantages of surface plasmons, plasmonic metasurfaces also possess strong LDOS and support significant electromagnetic field enhancement at near-field. What's more, owing to the specific design and precise nano-processing progress, metasurfaces possess abundant and customized optical responses according to the design targets. As discussed before, near-field information and far-field responses are inseparable. By manipulating the properties of light, such as phase, amplitude, and direction, through near-field interactions, the far-field responses of light will be influenced by metasurfaces manipulations and can achieve multiplex optical functions. Therefore, such planar elements afford facile integration with electronic, mechanical, magnetic, chemical, and bio-technologies in nano-optics. This remarkable capability has unique prospects for the design of highly integrated photonic signal-processing systems, optical imaging techniques, and sensors.

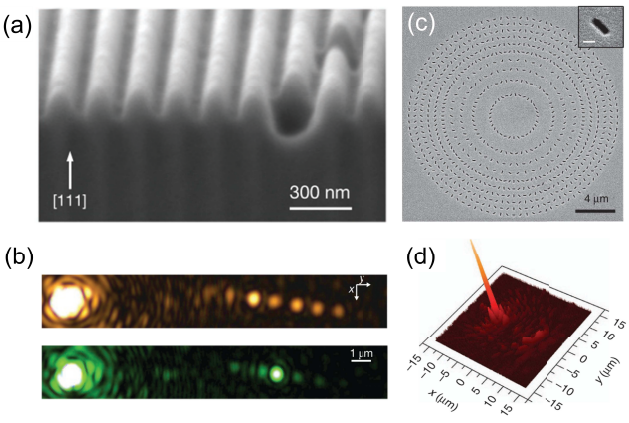


Fig. 5. Metasurface supporting SPPs and metalens supporting phase modulation. (a) The scanning electron microscope (SEM) image of a metasurface composed of nanometer-scale silver grating on a single-crystalline silver film. (b) SPP modes are observed. Optical images of SPP scattering at the out-coupling slots (right-hand side). The SPPs are excited with unpolarized light by a single in-coupling structure.[46] Reproduced with permission from Ref. [46]. (c) The developed metalens with different focal points achieved by conic shaping metasurfaces. (d) The measured field intensities at the real focal plane.[47] Reproduced with permission from Ref. [47].

Here, we introduce several examples of metasurfaces in detail to represent the development of plasmonic metasurfaces at the recent stage. The first study is a realization of a visible-frequency hyperbolic metasurface that supports SPP modes in 2015 published in *Nature*.[46] The metasurface with structures of a nanometer-scale silver grating on a single-crystalline silver film is manufactured by electron-beam lithography and plasma etching, as shown in Fig. 5(a). This developed preparation method produces smooth and high-quality silver nanostructures with superior aspect ratios, which is vital for supporting SPP modes in Fig. 5(b). The plasmonic spin Hall effect (PSHE) is further achieved. This work exhibits an advanced method of metasurface processing with superior precision. It provides the possibility of realizing low-dimensional transformation optics and further development in metamaterial-based devices, such as integrated metamaterial circuits. Besides, the developed metasurface possesses small mode volumes and increased plasmonic LDOS, thus it has the potential to be applied for enhancing interactions of SPPs with individual quantum emitters.

Phase modulation is also developed based on metasurfaces. Among them, metalens is a typical application of phase modulation by metasurfaces in photonic devices. Conic shaping metasurfaces with different focal points maintaining high imaging quality are developed, as shown in Fig. 5(c).[47] Owing to the specific design of the metasurface, incident lights can be constructively focused at the foci of the metalens with high quality, as shown in Fig. 5(d). This work exhibits a design of a multifocal lens possessing dual polarity under incident circularly polarization lights with improved imaging performance. Thereafter, the integration of different functionalities in one module is achieved in metasurfaces, which is in the interest of ultracompact photonic devices.

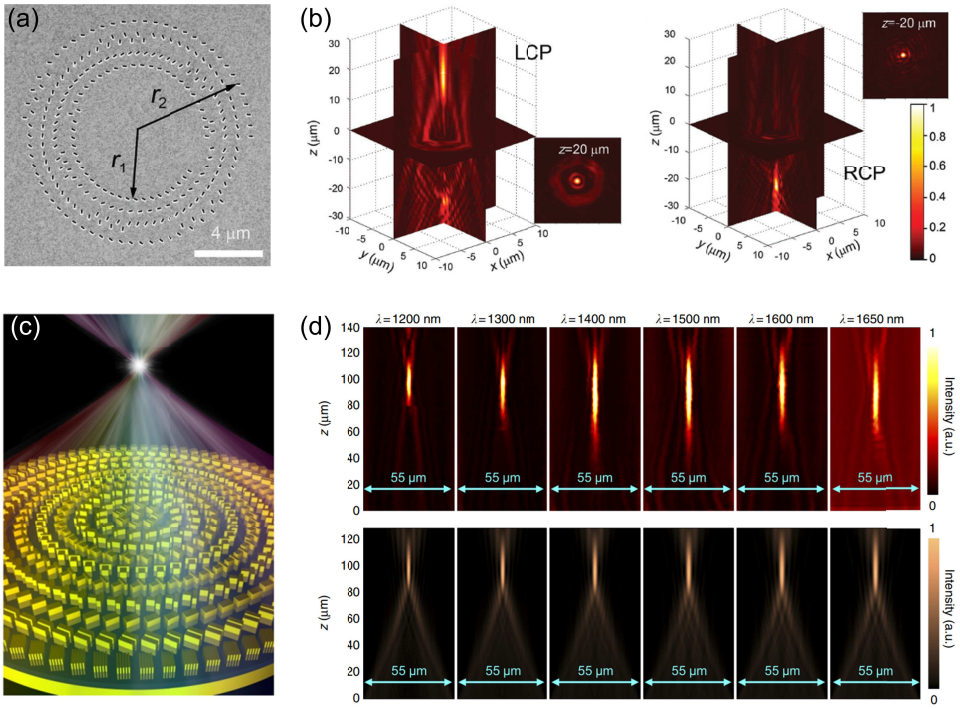


Fig. 6. Nanostructures of metasurfaces and the corresponding far-field performances. (a) The SEM image of the designed circular metasurface. The inner and outer radii are $r_1 = 3.5$ μm and $r_2 = 7$ μm. (b) The measured optical far-field intensity distributions under opposite circularly polarized incidences. The inset figures show the intensity distributions at the real and virtual focal (x–y) planes.[48] Reproduced with permission from Ref. [48]. (c) Schematic for the achromatic metalense. (d) Experimental (top) and numerical (bottom) intensity profiles of the achromatic metalens along axial planes at different incident wavelengths.[49] Reproduced with permission from Ref. [49].

The excellent ability of metasurfaces in manipulating light fields has been proposed for a variety of functional devices. An optical launcher based on a metasurface is developed, which successfully integrates the near-field and far-field functionalities.[48] The metasurface, which is presented in Fig. 6(a), can support the unidirectional launching of SPPs at the near-field while focusing on scattered light at the far-field simultaneously. The active controls of both near-field and far-field information are further materialized with the SPP propagating direction and the real/virtual focus can be manipulated by the spin state of the incident light, as shown in Fig. 6(b). This work represents the progress of photonic integration with different optical component manipulations in nano-optics and provides a method for designing multifunctional optical devices. In addition to the visible light range, metasurfaces also support near-infrared regime and mid-infrared. For example, achromatic metasurface devices with the chromatic aberration eliminated in a wide wavelength region are developed, as shown in Fig. 6(c).[49] The light intensity profiles along the cross-section plane in experiments and simu-

lations are presented in Fig. 6(d). It can be seen that a broadband achromatic converging property in near-infrared regime is achieved. By combining geometric phase with phase compensation through designed integrated-resonant elements, the demonstrated metasurfaces can efficiently suppress the chromatic aberration in a wide wavelength range with continuous bandwidth in the near-infrared regime. This work is an innovation of controlling phase dispersion and may promote the full color control in metasurface applications. In another work, gradient metasurface which can support high-efficiency anomalous reflections following the generalized Snell's law is also demonstrated in near-infrared regime.[50] Besides the near-infrared regime, considering the great application values of the mid-infrared band in infrared imaging and detection, metasurfaces have been studied to achieve mid-infrared imaging or manipulation. For example, recently, an achiral metasurface consisting of a gaped dual-germanium-dimer array has been developed to enhance the mid-infrared chiral sensing under both circularly polarized light and linearly polarized light excitations.[51] Manipulating light in near-infrared and mid-infrared regimes has greatly expanded the applicable scenarios of metasurfaces, which possesses extraordinary prospects in developing optical control devices in infrared regimes.

4.3. Plasmonic manipulation of 2D semiconductor materials

Excellent near-field responses of surface plasmons have been applied for enhancing, modulating, and manipulating light within the near-field range. It has been confirmed that the enhancement effect of surface plasmons can reach over 10 times under plasmon resonance modes with light–matter interaction efficiency significantly improved. On the basis of near-field enhancement achieved by surface plasmons, strong optical manipulation, and modulation capabilities are also applied in different material systems. Owing to the respective advantages of different plasmonic modes, the modulation of surface plasmons applied on other materials can be various. For example, the propagation characteristics of SPP can be used as a waveguide to transmit optical information. LSP can confine and modulate light through specific localized fields.[41,52] Specifically, with the increasing maturity of micro/nano processing technology, the design of LSP nanostructures has tremendous flexibility, which enables LSP to have customized electromagnetic field regulation. As a result, the flexible design characteristics at the near-field of surface plasmons have abilities to couple and modulate materials under different physical targets.

In recent years, the shadow of the unsustainable Moore's law has deeply enveloped the semiconductor industry. The birth of 2D semiconductor materials after the 2010s provides enormous potential for the continuation of Moore's law at nanoscales. 2D materials have atomic interactions within the layer while van der Waals interactions between layers. The thickness of the layer is much smaller than the wavelength of light. Among them, the rise of transition metal dichalcogenides (TMD) as a kind of 2D hexagonal pattern semiconductor material has driven the tremendous development of optoelectronic properties in nano-optics. TMD materials possess rich optical responses with unique physical properties, such as high carrier transport efficiency, exciton tunability, and fast photoelectric response, which have led to a large amount of work and applications.[53–55] Specifically, a TMD material, which possesses degenerate energy levels with antiparallel spin in K and K' valleys, will transform into direct bandgap semiconductors and exhibit strong light–matter interactions when it appears as a single layer.[56–59] However, the inherent deficiencies of TMDs limit the development of TMDs in practical applications. One is the photoluminescence (PL) generated by TMDs is weak due to the low light–matter interactions; the other is the PL of TMDs that is relatively passive due to the fixed band-gap and is hard to be modulated by external excitations. The significant near-field effects of surface plasmons have become an efficient method to overcome these two congenital defects of TMDs. Plasmonic nanostructures with specific designs can modulate the optoelectronic characteristics of TMDs with significant flexibility and achieve diversified optical functionalities, which has provided application prospects for new functional nanophotonic devices.[60,61]

One typical kind of study is the enhancement of PL emission from TMDs by surface plasmons. The PL of TMDs can be enhanced several times or even over ten times under plasmonic resonance. Furthermore, active modulation of TMDs PL by LSP produced by Au nano-particles is further achieved, which means the relatively fixed TMDs PL can be tailored or manipulated by external excitations. Different mechanisms have been established to realize active modulations of TMDs PL, such as hot-electrons transportation,[60] and electrode doping.[62–64] Specifically, doping plasmon hot electrons into the conduction band of monolayered MoS_2 by LSP from nanodots, as the schematics shown in Fig. 7(a), can achieve active control of the PL spectra of TMDs in Fig. 7(b).[60] Besides the light control system, electron-optical modulation is also achieved. Through near-field coupling between the silver disk and the monolayered MoS_2 in Fig. 7(c), a deep Fano resonance effect occurs and can be actively tuned through gate voltage, as shown in Fig. 7(d).[62]

In addition to improving the exciton radiation efficiency of TMD materials, there is another fundamental issue with exciton radiation, which is that the signal emitted by excitons to the far field is usually directionless, and the difficulty of signal collection limits the application of optical signal processing on chips. Surface plasmons can effectively guide and collect non-radiative photons through directional coupling by manually customizing special structures. Therefore, nano-waveguides

and metasurfaces based on surface plasmons have been developed to guide and collect light coupled with TMDs excitons appropriately. These structures can enhance the exciton radiation rate while utilizing the near-field conduction, localization, and confinement of plasmons to directionally transmit or radiate light. For example, transferring a monolayered WSe_2 to the top of a chemically synthesized silver nanowire can construct local defects of WSe_2 which further couple with SPP modes. The coupling information thus can propagate directionally along nanowires.[65] Especially, a crucial direction of this study area is the valleytronic modulation of TMDs by surface plasmons. As mentioned before, TMDs have degenerate energy levels with antiparallel spin in the K and K' valleys, resulting in different responses of two valleys to external excitations with opposite rotations.[52,59,66–70] The valley emission of TMDs in different valleys can be separated into opposite directions by plasmonic nanowires through near-field interactions. For example, Gong *et al.* established a system of plasmonic nanowires and monolayered TMDs, which was published on *Science* in 2018.[71] The valley emission of TMDs can be separated into opposite directions and led to determinant directions by SPP, as shown in Figs. 8(a) and 8(b).

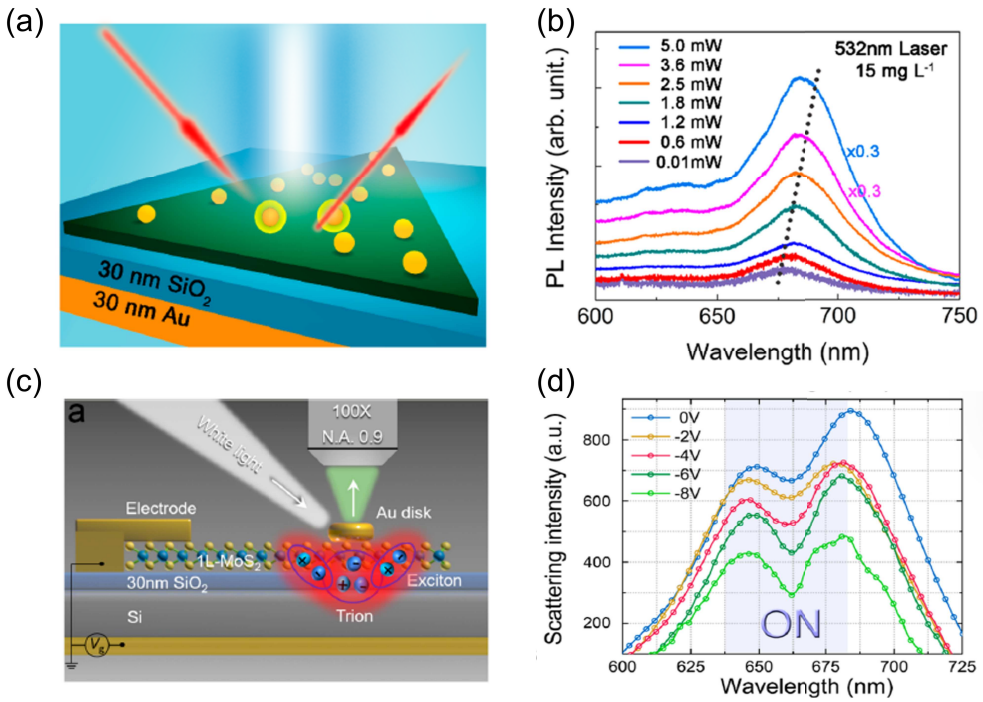


Fig. 7. PL emission from TMDs modulated actively by surface plasmons. (a) Schematics of hybrid structures combined with monolayered MoS_2 and Au nanodots. (b) Active control of the MoS_2 PL spectra is achieved, of which the PL peak exhibits red-shift with the intensity increased.[60] Reproduced with permission from Ref. [60]. (c) Schematics of Ag disk and monolayered MoS_2 under electro-modulation. (d) Deep Fano resonance effect occurs under electro-modulation with the intensity actively tuned by a gate voltage.[62] Reproduced with permission from Ref. [62].

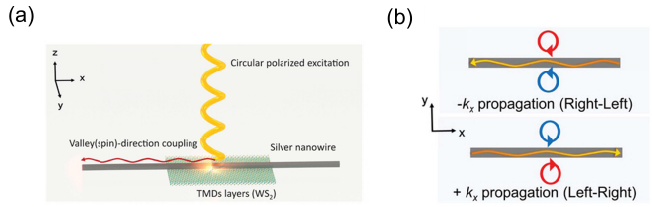


Fig. 8. Separation of valley emissions to opposite directions by SPPs. (a) Schematics of WS_2 valley-polarized excitons emitting to opposite directions of Ag nanowire due to PSHE induced by the interaction between valley excitons and SPP. (b) The propagation direction of valley emissions from WS_2 is determined by the excitation position and circular polarization.[71] Reproduced with permission from Ref. [71].

4.4. Hybrid system of metasurface and 2D semiconductor materials

According to the pattern classification of surface plasmons, metasurfaces can generate surface plasmons with different modes of SPP and LSP according to the design goals. While achieving different functions, they can also effectively combine with different materials. Plasmonic metasurfaces are an effective means to regulate the optical properties of materials in the near-field range, and can further regulate the far-field spectra of materials. Hybrid structures of metasurface and 2D materials have been further developed.

The enhancement of TMDs PL can be improved prominently by metasurfaces and the modulations of PL emission can become more plentiful. For example, the establishment of energy transfer between metasurface with $C3$ structural symmetry (Fig. 9(a)) and TMDs hybrid structures can achieve tens of nanometers blue-shift of PL spectra with the intensity enhanced over 10 times, as shown in Fig. 9(b).[39] The near-field energy transfer from metasurface radiation to TMD emission is studied. Due to the efficient near-field energy transfer, the PL wavelength and intensity of hybrid structures can be mod-

ulated by external excitation power, which is caused by the renormalization of energy levels in TMDs. The peak shift modulated by excitation power can reach tens of nanometers, as shown in Fig. 9(c). In another work, metasurfaces with helical structures are developed to amplify the exciton emission of TMDs by SPP focusing field induced by the interaction of TMDs exciton and metasurface.[41] The emission signals are enhanced and controlled by optical spin states and excitations. Besides, the PL intensity of hybrid structures can be determined by odd or even numbers of helical rings. The PL mapping patterns can be switched by circularly polarized excitations, as shown in Fig. 9(d).

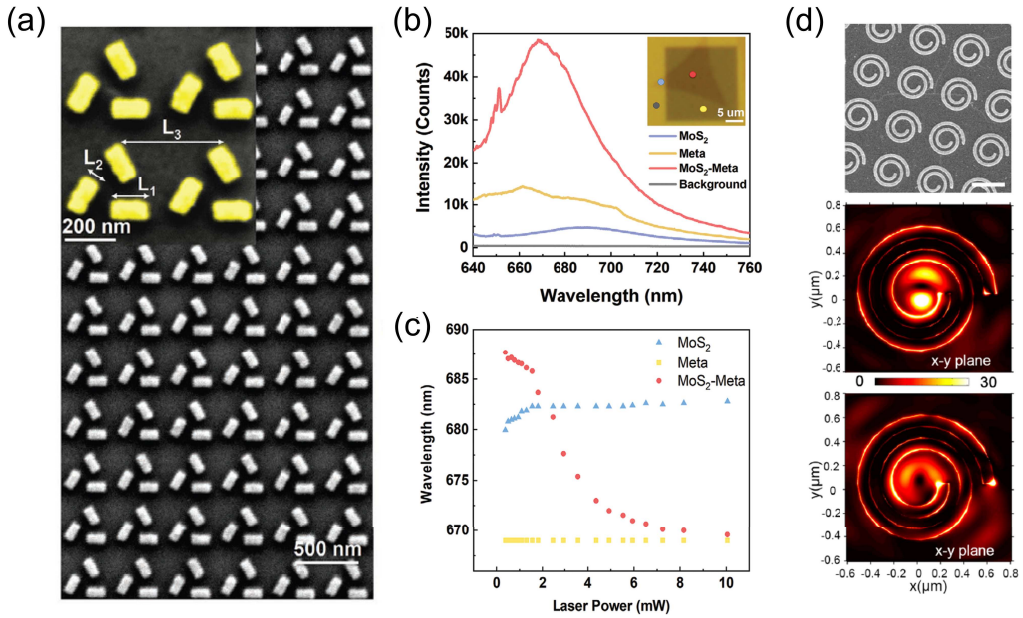


Fig. 9. Active modulation of TMDs by metasurface in hybrid systems. (a) The SEM image of the metasurface with *C*3 symmetry unit. (b) The PL spectra of MoS_2, metasurface and MoS_2- metasurface hybrid system. The PL of the hybrid system is enhanced over 10 times compared with bare MoS_2. (c) The relationship between the excitation power and PL peak position of MoS_2 metasurface and MoS_2- metasurface. The PL peak behaves an obvious blue shift up to 20 nm with the excitation power increased.[39] Reproduced with permission from Ref. [39]. (d) The developed metasurface with helical nanostructures to amplify the exciton emission of TMDs by SPP focusing field. Near-field intensity at *x*–*y* plane with/without focusing spot under opposite circularly polarized excitations.[41] Reproduced with permission from Ref. [41].

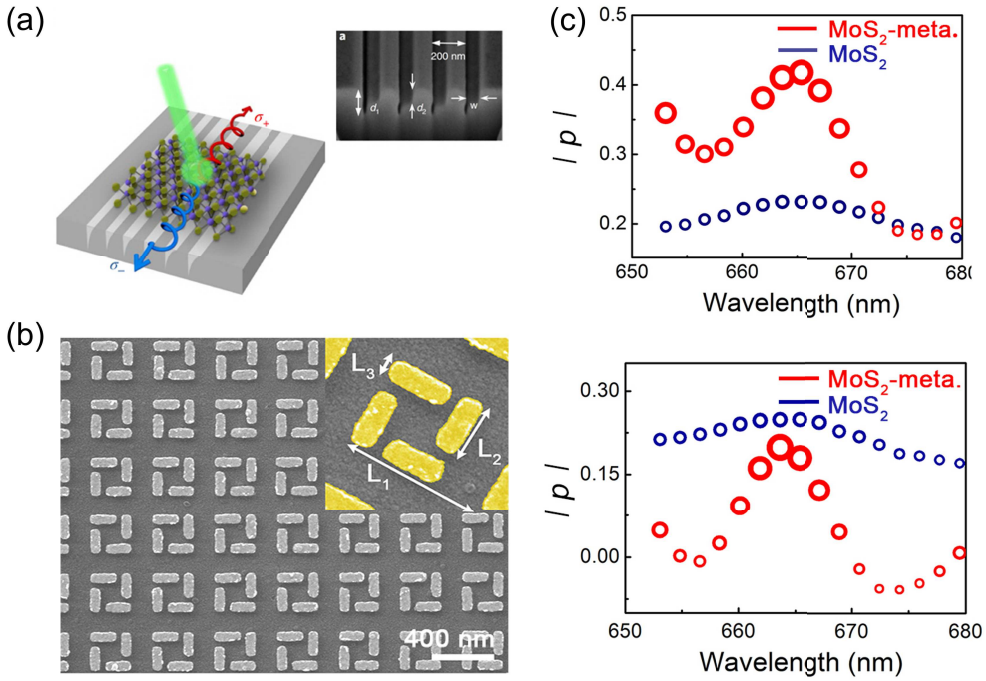


Fig. 10. Valley emission of TMDs modulated by metasurfaces. (a) Schematic of the hybrid system and the SEM image of the metasurface with asymmetric grooves. The valley emission from MoS_2 is separated in opposite directions due to the PSHE induced by the efficient coupling between monolayered MoS_2 and metasurface.[72] Reproduced with permission from Ref. [72]. (b) SEM image of metasurface with chirality. The metasurface is further combined with monolayered MoS_2 to manipulate the degree of valley polarization. (c) The degree of valley polarization of MoS_2 and MoS_2-metasurface hybrid systems under different circularly polarized excitations.[52] Reproduced with permission from Ref. [52].

Metasurfaces are also developed to modulate valley emission of TMDs. The modulation principle of valleytronic emission by plasmonic metasurfaces mainly contains three aspects.[40] The first is to enhance low intrinsic quantum yields of valley emissions through near-field interactions induced by surface plasmons. The second is to improve the relatively low exciton valley polarization and shorten the exciton recombination lifetime by specific plasmonic structures. The third is to lead and collect the directionless valleytronic emissions by near-field couplings. Here, we introduce two studies based on SPP and LSP metasurfaces respectively. Metasurface with asymmetric grooves is developed to support specific SPP, which can further induce asymmetric near-field coupling modes. The metasurface is combined with TMDs to effectively separate valleytronic luminescence and transport to different directions, as shown in Fig. 10(a).[72] On the other hand, metasurface supporting chiral LSP mode (Fig. 10(b)) can selectively enhance or suppress valley emission under different circular polarization excitations when it is combined with TMDs, and further modulate the degree of valley polarization actively, as shown in Fig. 10(c).[52]

In addition to the TMDs, metasurfaces have also been applied to manipulate other 2D materials. For example, black phosphorus (BP) possesses tunable direct bandgap from 0.3 eV to 2.0 eV and has the potential to develop photodetectors operating in the mid-infrared band. Plasmonic metasurfaces supporting LSP modes are designed. A BP-based mid-infrared band photodetector is fabricated on the plasmonic metasurface and achieves high peak responsivity and operation speed.[73]

5. Summary

In the competition to find the solution of higher optical resolution over diffraction limit, near-field optical microscopy plays an important role in demonstrating this effort, including the development process of the SNOM technique of aperture-SNOM and apertureless-SNOM. However, due to the limitation of optical throughput and difficulties in integrating this technique with optoelectronic devices, an alternative approach has gradually occupied the core position, i.e., using plasmonic principle and precise nano fabrications to reach the higher optical resolution, versatile nano-optical regulation, flexible control of the beam directions, and possible in-chip integration of optical source and detection. In this article, the original development of near-field optics microscopy from the 1980s has been reviewed. With the increasing demands of higher resolution, higher throughput, higher speed, and/or versatile optical regulation, the field of nano-optics has deviated to plasmonics-based research. More specifically, the impetus for earlier nano-optics development is pursuing higher optical resolution to hundreds or tens of nanometers. The invention of SNOM labels the first breakthrough in breaking diffraction limit resolution in optics. Near-field optics arises at the historic moment, which was further developed around surface plasmons. In explorations of improving the SNOM probe further, surface plasmon as a unique near-field enhancement platform has developed a lot. After the scale of optical characterization has entered tens of nanometers, the development of improving optical resolutions meets the bottleneck. The research area of nano-optics gradually turns to near-field manipulation focus and exploiting photonic applications, which provides a platform to develop new photonic devices for integrated circuits. Benefits from the excellent near-field exhibition of surface plasmons, a large number of studies and applications have been developed to enhance and manipulate light at the nanoscale by surface plasmons.

From the historical perspective, we can consider that the driving force for nano-optics development was dependent on the progress of techniques. At the beginning of nano-optics, the development of nano-optics was determined by the improvements in technology, i.e., SNOM detection. Thereafter, surface plasmon emerged as an efficient near-field enhancement method. The further development of nano-optics based on plasmonics was then propelled by the improvements in nano-processing technology, i.e., precision nano-fabrication and manipulations, so that surface plasmon can become an efficient near-field manipulation platform in nano-optics. With the development of nano-processing techniques, plasmonic nanostructures with specific designs can achieve different optical responses and functions to meet plentiful target scenarios. Design methods beyond conventional ways have been further developed. Nowadays, photonic design based on algorithms has gradually spread in the field of nano-optics. Among them, artificial intelligence algorithms, including end-to-end neural networks and unsupervised learning design methods, have brought vitalities for nano-optics. In the future, the broad design space hidden behind powerful algorithms has the opportunity to seek more powerful photonic devices and further expand the developing prospects of nano-optics.

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