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PERSPECTIVE

New frontiers in metamaterials research: Novel electronic materials and inhomogeneous metasurfaces

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In reviewing some recent work in metamaterials, we highlight two exciting new frontiers just emerging in this field – metamaterials made by new electronic materials (particularly graphene) and inhomogeneous metasurfaces to control light wave-fronts.

Keywords metamaterial, new electronic material, graphene, inhomogeneous metasurfaces, wave-front control

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1 Introduction

Metamaterials (MTMs), artificial materials made by subwavelength functional microstructures (usually called "meta-atoms") arranged in some specific "orders", have received extensive interests in the past two decades following the pioneering theoretical paper of Veselago [1]. Via engineering the "meta-atom" and/or choosing an appropriate "order", one can design an MTM with extraordinary electromagnetic (EM) properties to control light and EM waves in a desired manner [2–21]. Historically, this field was mainly driven by researches along two lines – "meta-atom" and "order". Earlier studies largely focused on the "meta-atom" part, but paid less attention to the "order" part by simply assuming the MTMs to possess periodic or homogeneous orders. For example, designing "meta-atoms" exhibiting double-negative EM responses, people discovered negative refraction [2, 3] and super lensing effects [4–6]; Designing anisotropic or chiral "meta-atoms" with desired responses to lights polarized along different directions, people realized functional polarization-control devices working in different configurations and frequency domains [7–10]; Reducing the sizes of meta-atoms further and further, people realized MTMs functioning at higher and higher frequencies [THz, Infra-red (IR) and visible] [11]. On the other hand, the development along the "order" line is a bit retarded. Since 2006, stimulated by the establishment of the transformation optics (TO) theory [12, 13] and its related phenomena [14], people gradually realized the importance of "order" in expanding the light-manipulation abilities of an MTM. With TO as a power tool, one can design gradient index materials to guide light flowing in pre-designed manners, leading to fascinating physical effects such as invisibility cloaking [12, 13], illusion optics [15, 16], and several functional devices [17–21].

Despite the great successes already achieved, the development of MTM meets grand challenges recently. For example, while typically noble metals are considered as good candidates to make meta-atoms, they suffer limitations such as poor tunability, weak EM field confinements at low frequencies and inevitable losses at high frequencies [22]. On the other hand, the TO-based materials are typically requested to be inhomogeneous, anisotropic and magnetic, which sometimes limits their practical re-

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alizations, while a wide range of inhomogeneous MTMs with different "orders" remain unexplored and thus their abilities to control light are yet to be discovered.

Challenges sometimes mean opportunities. In struggling with these challenges, people tried different approaches on different systems, and thus new research frontiers gradually emerged along with these efforts. On the one hand, researchers start to explore new materials with usual electronic band structures (EBS) to build "meta-atoms", with the hope to overcome the limitations faced by conventional metals [23]. On the other hand, people begin to work on inhomogeneous MTMs with more fascinating global "orders", particularly a class of planar meta-surfaces (ultra-thin MTM layers) with tailored EM responses which exhibit even stronger abilities to control light propagations [24, 25]. In what follows, we briefly introduce the two emerging topics to the readers based on our own perspectives. We emphasize that this paper is by no means a comprehensive overview on the historical development of the two topics. Rather, this represents our personal viewpoints on possible new directions in MTM research.

2 Making MTMs with novel electronic materials

We first present a unified framework based on which the optical (plasmonic) properties of different electronic materials can be easily compared. Consider a general electronic material with dimensionality D and EBS $\varepsilon_{s,\vec{k}}$, the dynamical conductivity function $\sigma(\vec{q},\omega)$can be obtained based on the linear response theory [22]. Taking only the intra-band contributions, expanding $\sigma(\vec{q},\omega)$ to series of $\vec{q}$ and keeping the leading term, it was shown in Ref. [26] that

$$\sigma(\vec{q},\omega) \approx \frac{\mathrm{i}e^2}{(\omega+\mathrm{i}\tau^{-1})D} G(E_f)\cdot(v_f)^2 \tag{1}$$

where $G(E_f)$ and $v_f = \hbar^{-1}\nabla_{\vec{k}}\varepsilon_{\vec{k},s}\big|_{\varepsilon=E_f}$ are the density of states (DOS) and velocity of electrons measured at the Fermi surface (FS). To understand the "*tunabilties*" of different electronic materials, we show in Fig. 1(a)–(c) how $\mathrm{Im}(\sigma(\omega))$ (and thus the plasmonic property) changes as we dope the same number of electrons into three typical electronic materials – a three-dimensional (3D) conventional metal, a two-dimensional (2D) semiconductor and a graphene. The first two examples exhibit usual parabolic EBS, while a graphene exhibits highly unusual linear (Dirc–Cone like) EBS. Insets to Fig. 1 show that such a doping can hardly change the FS in a conventional metal, but can dramatically lift the FS in a doped semiconductor and a graphene. Therefore, the plasmonic properties are essentially unchanged for a conventional metal [Fig. 1(a)], but are remarkably modified in the latter two systems [Fig. 1(b)–(c)], upon the same amount of electron doping.

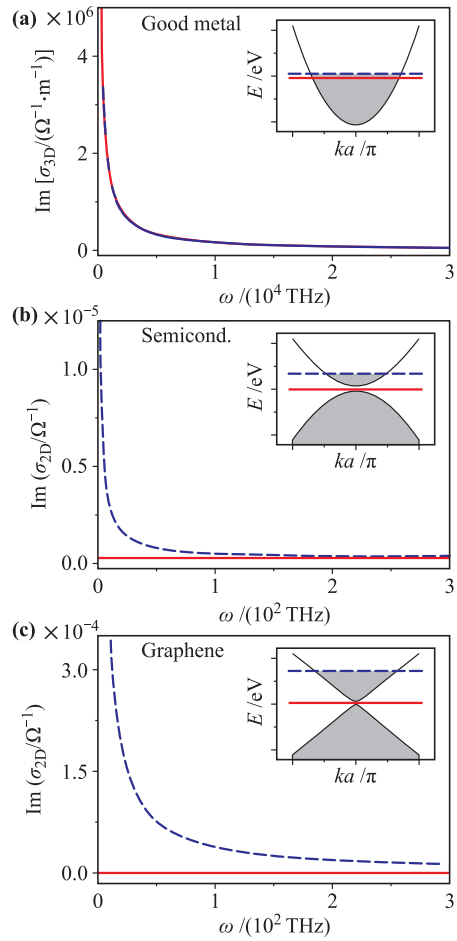

Fig. 1 $\mathrm{Im}(\sigma)$ versus frequency for **(a)** a 3D metal, **(b)** a 2D semiconductor and **(c)** a graphene, calculated with different electron densities. Solid lines represent un-doped cases in which (a) $n = 5.9\times10^{28}\mathrm{m}^{-3}$ (representing the case of Au) and (b) (c) $n' = 0\mathrm{m}^{-2}$, and dash lines represent the doped systems with additional electrons (a) $\Delta n = 3.33\times10^{24}\mathrm{m}^{-3}$, (b) and (c) $\Delta n' = 1\times10^{15}\mathrm{m}^{-2}$ added. Insets show the band structures of three different systems under different doping situations, with solid and dashed lines denoting the Fermi levels in un-doped and doped systems.

The ability of a particular electronic system to confine EM waves is another interesting topic to be discussed. Without using too many mathematics, we can easily understand this point by examining the dispersion of surface-plasmon-polariton (SPP) on an air/metal interface. Figure 2 shows that the SPP is very loosely confined on the metal surface ($k_{\mathrm{spp}} \approx k_0$ with $k_0 = \omega/c$) at low frequencies where the system is too metallic [$\mathrm{Im}(\sigma)$ is too large], but becomes tightly confined on the metal surface ($k_{\mathrm{spp}} \gg k_0$) only when $\mathrm{Im}(\sigma)$ is small enough. A better SPP confinement implies a more *compact* MTM unit cell size if we pattern the system to obtain an MTM,

which is highly desired in MTM designing. Therefore, at low frequencies (THz and far-IR), doped semiconductors and graphene are probably better candidates to fabricate MTMs with deep-subwavelength unit cells since they are much less conducting [with much smaller Im(σ) due to lower Fermi levels] than a conventional metal.

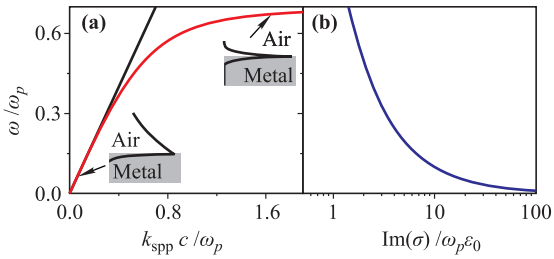

Fig. 2 **(a)** Dispersion relation of SPP on an air/metal interface with metal's permittivity given by $\varepsilon = \varepsilon_0(1-\omega_p^2/\omega^2)$. Insets show the field distributions calculated at different frequencies represented by the arrows. **(b)** Imaginary part of the conductivity function for the metal as a function of frequency.

Knowing the plasmonic properties of different electronic materials, we can easily understand those recent efforts in using new electronic materials to build MTMs. In 2011, Boltasseva and Atwater presented an interesting materials map to compare the plasmonic properties of available materials in different EM spectra [23]. In 2012, Hosang Yoon *et al.* fabricated an MTM based on patterning a 2D electron gas, and showed that the system exhibited negative refraction in microwave regime [27]. Recently, Engheta's group used indium-tin-oxide – a semi-conducting material with plasmonic responses in IR regime – to make functional components for MTMs [28].

Meanwhile, graphene has attracted more and more attention in connection with MTM and plasmonics research, and experimental breakthroughs appear in recent two years. Nearly at the same time in 2012, two groups independently characterized the SPP properties of un-patterned graphene, based on the near-field scanning microscopic technique [29, 30]. Since a huge momentum mismatch exists between a propagating wave and an SPP on the graphene, it is difficult to use conventional far-field-based techniques to measure the SPP dispersion relation in graphene. As shown in Fig. 3, Koppens' group and Basov's group put tips on top of graphene samples to launch SPPs, which propagate on the graphene surface and are then reflected back by the boundary. Interferences between excited and reflected SPP signals form standing waves on the graphene surface, making it possible to obtain the wave-vectors of the SPP by measuring the established near-field patterns. Measured SPP dispersion relations coincided well with theoretical ones, confirming the excellent confinement of SPP in graphene.

The outstanding SPP confinement of graphene must stimulate more research attempts in employing graphene to make MTMs. In fact, pioneering work has already been done by Wang's group in 2011 [31], where the authors patterned a piece of graphene into nano-ribbons [Fig. 4(a)]. By measuring the transmission/reflection

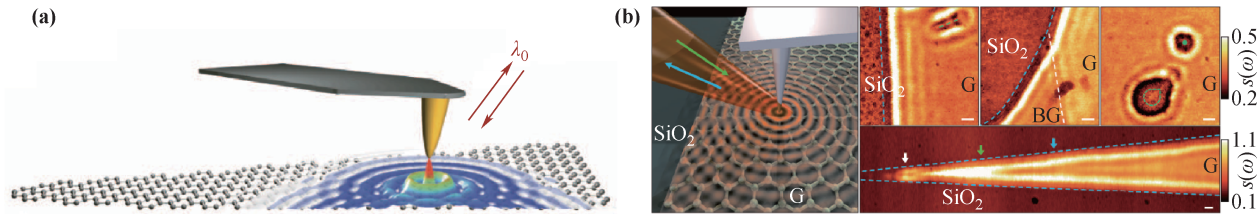

Fig. 3 Experimental setups to characterize the SPP dispersions in graphene, adopted by **(a)** Koppen's group [29] and Basov's group [30] **(b)**. Reproduced from Refs. [29, 30], Copyright © 2012 Nature Publishing Group.

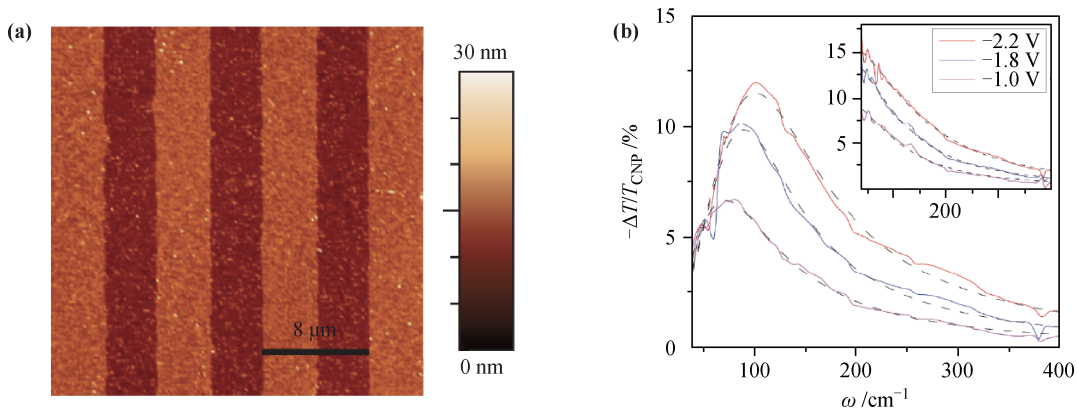

Fig. 4 **(a)** Scanning electron microscopic (SEM) image of a graphene nano-ribbon sample. **(b)** Relative transmission spectra for a graphene nano-ribbon under different gating voltages. Reproduced from Ref. [31], Copyright © 2011 Nature Publishing Group.

spectra of these graphene ribbons [Fig. 4(b)], Wang's group showed that such MTMs exhibited well-defined deep-subwavelength resonances, manifesting in the pronounced reflection peaks in the spectra. Moreover, such resonances can be easily tuned by applying external voltages [Fig. 4(b)], in consistency with theoretical conjecture.

The experimental work set up a solid basis for graphene-based plasmonics and MTMs, and must stimulate much follow-up work in this field. In fact, there are already quite a few theoretical proposals waiting for experimental verifications [32–34]. For example, it was predicted that the propagation of SPPs can be controlled by locally gating a graphene [32], and the performance of a nano-emitter near a graphene substrate can be significantly enhanced due to its couplings with SPP on the graphene [33]. However, grand challenges exist in realizing these proposals, such as how to fabricate large-area high-quality graphene samples and how to reduce the losses in SPP propagations. Even on theoretical side, lots of challenges exist. For instance, what are the consequences of the highly unusual behaviors of Dirac Fermions in graphene, especially in determining the nonlocal and nonlinear optical responses of such systems? Is a classical theory, which is good enough for noble-metal-based MTMs, sufficient for MTMs built with new electronic materials? Answering these questions must open new sub-branches in MTM research, and can stimulate future work in both science and technology, which may eventually lead to useful functional optical devices.

3 Inhomogeneous metasurfaces for wave-front control

One key motivation in MTM research is to manipulate EM waves at will. Recently, in addition to the "meta-atom" degree of freedom, people realized that the "order" of an MTM is another important degree of freedom to be explored. To reshape the wave-front of an incident light, conventional idea is to modulate the local phases/amplitudes of lights passing through an optical device at different positions, so that the re-interference of these modulated waves can lead to the desired wave-front. Since the permittivity of ordinary material is rather limited (say, for glass $\varepsilon_r \approx 2.1$), an optical device typically exhibits a curved shape and is much thicker than wavelength. Combining the TO concept [12, 13] with MTM, many new optical devices were realized which exhibit gradually varying EM properties in a pre-designed manner in order to guide the flow of light at will [12–21]. While these devices do not necessarily exhibit curved shapes and have much stronger light-manipulation abilities, they are still optically bulky and the scattering losses due to impedance mismatch (both at the surfaces and inside the devices) are hard to avoid.

Very recently, a class of inhomogeneous ultra-thin MTMs (usually called meta-surfaces), with planar "meta-atoms" possessing distinct EM responses arranged in a specific order, draws a great deal of attention [24, 25, 35–46]. Instead of modulating the transmission phase inside the bulk medium, such systems utilized the *abrupt* material discontinuities to control the local reflection/transmission phases for lights incident onto different lateral positions in the system. Compared to the cases of conventional and TO-based devices [12–21], such meta-surfaces are much thinner than wavelength and suffer less on the loss issues since light does not transport inside the inhomogeneous medium for a long time [24, 25]. In 2011, Capasso's group fabricated a series of meta-surfaces based on V-shaped optical antennas working at $\lambda = 8$ μm, and demonstrated several astonishing light-manipulation effects [24]. The first sample was designed such that the transmission/reflection phase (denoted by Φ) of each V-shape antenna varies *linearly* as a function of position on the surface [Fig. 5(a)]. The authors found that light reflection/refraction at such a planar surface follows a generalized Snell's law [24]:

$$k'_{//} = \xi + k_0 \sin\theta_i \tag{2}$$

in which the parallel wave-vector is not conserved when light is reflected or refracted by the meta-surface, but rather with an additional k vector contributed by the gradient ξ of the phase variation [$\xi = \nabla\Phi(x)$]. Here $k'_{//}$ denotes the parallel wave-vector of the reflected or refracted light, k_0 the free-space wave-vector and θ_i the incident angle. Note that ξ can take different values in cases of reflection or refraction [24]. The generalized Snell's law was experimentally verified by far-field measurements on the sample [see Fig. 5(b)]. The second meta-surface sample was designed such that Φ varies as a function of azimuthal angle [Fig. 5(c)], and the authors showed that such a meta-surface can generate an optical vortex [Fig. 5(d)] [24]. Shortly after this study, Shalaev's group pushed the idea to a shorter wavelength (∼2 μm) by down scaling the optical antenna's size, and demonstrated that the functionality of such a device is rather broadband [35].

Despite these achievements, one obvious drawback of these meta-surfaces is that the conversion efficiencies to the anomalous refracted or reflected beam are quite low [24], since these devices support not only anomalous refraction and reflection modes but also the normal refraction and reflection modes. Another drawback of such meta-surface is that the generated anomalous modes take different polarization with respect to the

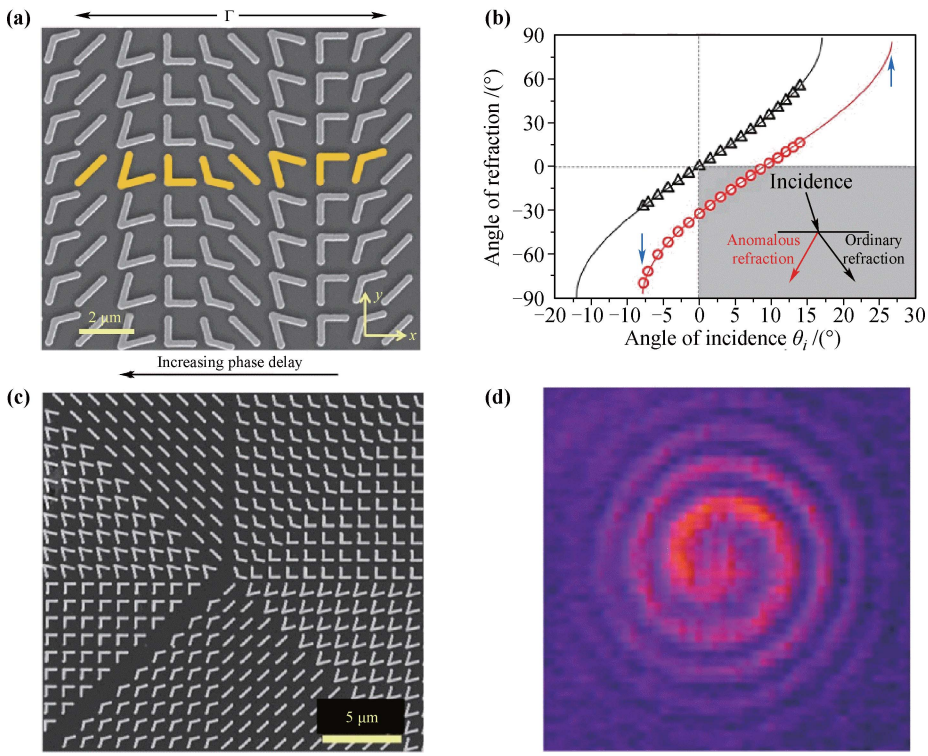

Fig. 5 **(a)** SEM image of a meta-surface for demonstrating the generalized laws of reflection and refraction. **(b)** Angle of refraction versus angle of incidence for the ordinary (black curve and triangles) and anomalous refraction (red curve and dots). **(c)** SEM image of a meta-surface that creates an optical vortex. **(d)** Measured far-field intensity distributions of an optical vortex with topological charge one. Reproduced from Ref. [24], Copyright © 2011 American Association for the Advancement of Science.

incident one, since such systems utilized the bi-anisotropic response of the V-shape antenna [24, 36, 37]. In 2012, Zhou's group proposed a new type of meta-surface, with building block as a sandwich structure with a continuous metal sheet on the back, and demonstrated that such meta-surfaces can overcome the shortcomings of previous designs [25]. These new meta-surfaces do not allow transmissions and kill almost completely the normal (specular) mode, so that the efficiency for the anomalous reflection is nearly 100% (Fig. 6). In addition to the apparent improvement, Zhou's group pushed the concept of generalized Snell's law significantly forward by showing that the incident propagating wave (PW) can be converted to surface waves (SW), when $\xi > k_0$ or the incident angle is larger than a critical value [see Fig. 6(h) and 7(a)] [25]. Note that such a PW-SW converter is operated on a non-resonant mechanism, which is conceptually different from conventional devices such as prism and grating couplers operating on a resonant coupling scheme, and thus has much higher conversion efficiency. The authors experimentally characterized such a PW-SW conversion based on near-field scanning technique and further demonstrated that such SW can be guided out of the meta-surface to flow as SPP on another system (Fig. 7). The initial idea was demonstrated in microwave regime [25], but was soon pushed to optical regime [38].

These studies again open up a new sub-field in the MTM research, in which many interesting effects can be explored. For example, modulating the $\Phi(x)$ profile as a parabolic shape, flat lenses were realized in different frequency domains and in different geometries [39–42]. Other interesting extensions include optical vortex plate [24, 43], out of plane light reflector [44], broad-band wave plates [45], aberration free axicons [42] and anomalous reflection for circularly polarized light [46]. In addition to these already realized applications, many more possibilities are waiting to be explored. For example, is it possible to realize the PW-SPP conversion in optical domain? Can we realize the inverse effect (i.e., SPP-PW conversion)? Do we have other physical laws to be generalized? It is our perspective that this is just the beginning of the sub-field, and answering these questions must lead to many new discoveries and new functional devices.

4 Conclusions

In summary, based on our own perspectives, we

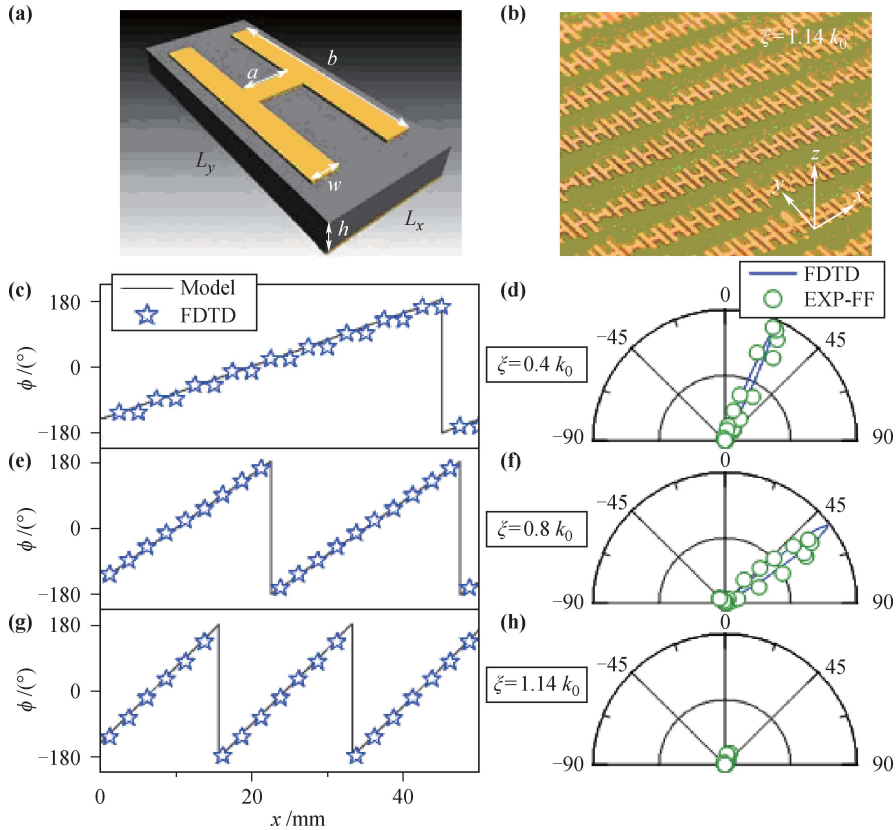

Fig. 6 **(a)** Geometry of a building block for gradient meta-surfaces: a metallic H and a metal plate separated by a dielectric spacer ($\varepsilon_r = 3.9$). **(b)** Picture of a representing sample. **(c)**, **(d)** and **(e)** are reflection phase distributions ($\Phi(x)$) and **(f)**, **(g)**, **(h)** are measured and simulated scattering patterns for three meta-surfaces with $\xi/k_0 = 0.4$, 0.8 , 1.14. The EM waves are reflected to non-specular channels in (d) and (f) following the generalized Snell's law and are converted to SWs bounded on the meta-surface in (h). Reproduced from Ref. [25], Copyright © 2012 Nature Publishing Group.

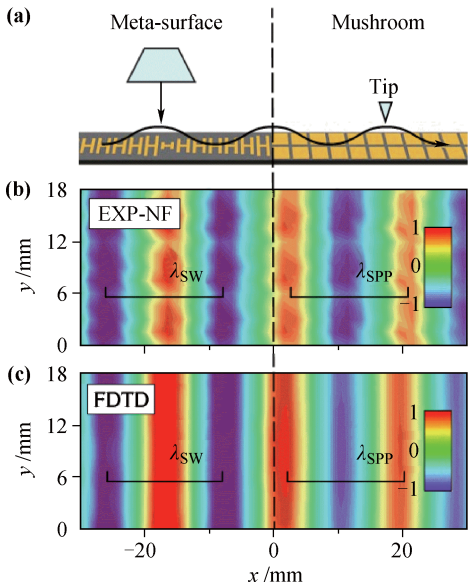

Fig. 7 **(a)** Schematics of the PW-SW-SPP conversion. Shining the meta-surface by a PW, SWs will be generated on the meta-surface (left) which then flow across the boundary to the mushroom structure (right) as a SPP. Field patterns obtained by **(b)** near field experiment and **(c)** FDTD simulations at 15 GHz. Reproduced from Ref. [25], Copyright © 2012 Nature Publishing Group.

highlighted two new frontiers in MTM research to the readers, with one being new material based MTMs (especially graphene MTMs) and another being inhomogeneous meta-surfaces to control light wave-front. Although some pioneering work has already been done during the last two years, there are still enough vacant spaces to be explored and we look forward to more exciting results in these two fields.

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