

Metasurface-assisted broadband optical absorption in ultrathin perovskite films

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Abstract: Ultrathin hybrid organic-inorganic perovskite (HOIP) films have significant potential for use in integrated high-performance photoelectric devices. However, the relatively low optical absorption capabilities of thinner films, particularly in the long-wavelength region, pose a significant challenge to the further improvement of photoelectrical conversion in ultrathin HOIP films. To address this problem, we propose a combining of ultrathin HOIP film with plasmonic metasurface to enhance the absorption of the film effectively. The metasurface excites localized surface plasmon resonances and deflects the reflected light within the HOIP film, resulting in an obvious enhancement of film absorption. Finite-difference time-domain simulation results reveal that the far-field intensities, deflection angles, and electric field distributions can be effectively varied by using metasurfaces with different arrangements. Examination of the reflection and absorption spectra reveals that embedding a specifically designed metasurface into the HOIP film produces an obvious enhancement in broadband optical absorption compared with pure HOIP films. We further demonstrate that this broadband absorption promotion mechanism can be effective at a wide range of HOIP film thicknesses. Comparison of the absorption spectra at various incidence angles of ultrathin HOIP films with and without underlying metasurfaces indicates that the addition of a metasurface can effectively promote absorption under wide-angle incident light illumination. Moreover, by extending the metasurface structure to a two-dimensional case, absorption enhancements insensitive to the incident polarization states have also been demonstrated. This proposed metasurface-assisted absorption enhancement method could be applied in designing novel high-performance thin-film solar cells and photodetectors.

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1. Introduction

Owing to properties such as tunable bandgaps [1,2], long carrier diffusion lengths and lifetimes [3–6], and low fabrication costs [7,8], hybrid organic-inorganic perovskites (HOIPs) have emerged as promising candidates for use as active materials in photoelectrical devices. In addition, their high absorption coefficients ($\sim 10^4 - 10^5$ cm⁻¹) improve the generation of photo-induced carriers under illumination, enabling photon harvesting in photovoltaic devices [9,10]. It has been reported that the optical penetration depths of perovskite films are comparable to those of many conventional materials used in photovoltaic devices, such as GaAs and silicon [11]. However, rapidly developed integrated photoelectrical devices have more stringent requirements in terms of photoactive material thickness [12,13], requiring a tradeoff between thickness reduction and absorption capabilities of ultrathin perovskite films. For example, light absorption can be effectively enhanced by guiding and retaining incident light through optical path enhancement or the spatial redistribution of light intensity by adding artificially constructed textured interfaces [14–17]. Light absorption can be significantly enhanced through the excitement of localized Mie

resonances in dielectric resonators (e.g., silicon or TiO₂ nanoparticles) to produce near-field enhancement and promote light trapping within the perovskite layer [18,19]. The absorption of nanostructured perovskite layers can also be enhanced by engineering the excitation of localized and guided modes within the perovskite film [20–22]. In addition to dielectric resonators, nanoscale metal resonators have also been reported to enhance the absorption of HOIP films. By exciting localized surface plasmon resonances (LSPRs) in metal nanostructures, incident light can be trapped at the subwavelength scale to achieve localized field enhancement, thereby enhancing the absorption of the perovskite film [23–25].

In this study, with the goal of creating another method for achieving highly efficient absorption enhancement in ultrathin HOIP films, we develop an approach for the effective improvement of ultrathin HOIP film absorption through the use of a plasmonic metasurface to manipulate the light wavefront, which is rarely proposed before. It has been reported that plasmonic metasurfaces can be used to control the amplitude and phase of electromagnetic waves to manipulate the wavefronts of output light effectively [26–31]. For example, broadband quarter-wave plates can be implemented using phased antenna arrays to produce scattered light waves with arbitrary polarization states [33]. By combining a metallic metamaterial with a dielectric interlayer, it is possible to cancel the intrinsic dispersion of metallic structures, thereby achieving the metastructures that can change the polarization states at broad band [34,35]. Besides, highefficiency holography can be achieved by taking advantage of the geometric nature of the phase profile of plasmonic metasurfaces to perform light control has rarely been applied to the enhancement of absorption in ultrathin perovskite films.

Here, we propose an application of plasmonic metasurfaces to enhance the optical absorption of ultrathin perovskite films. The metasurface can bring accurate phase modulation to the perovskite system and control the light deflection effect. In this case, the metasurface-induced LSPR excitation and extension of the optical path within the HOIP film synergize to produce a significant enhancement of the broadband optical absorption of the HOIP film. Simulation results obtained using finite-difference time-domain (FDTD) methods indicate that far-field intensities, deflection angles, and electric field distributions can be significantly altered by introducing metasurfaces with different arrangements. Reflection and absorption spectra reveal that the LSPRs and the deflection of reflected light induced by the metasurface obviously enhance the absorption of the film. Unlike pure HOIP films, HOIP films with metasurfaces have strong broadband absorption capabilities over a wide range of film thicknesses. Besides, we demonstrate that broadband absorption enhancement can also be effectively achieved under wide-angle incidence conditions. Furthermore, we extend the metasurface structure to a two-dimensional case and realize the absorption enhancements insensitive to incident polarization states. Our results can be applied to integrated perovskite photoelectrical devices, such as solar cells and photodetectors.

2. Metasurface design

Plasmonic metasurfaces support LSPRs and reflection light deflection through introducing phase gradients that allow incident light to be effectively trapped and absorbed within the HOIP film. Here, we designed a plasmonic metasurface located at the bottom of the perovskite absorbing layer, as shown in Fig. 1(a). The metasurface building block comprises four gold nanostripes with varying widths (w1, w2, w3, and w4) placed on top of a SiO₂-gold bilayer substrate, with a separation between adjacent stripes of p. The thicknesses of the stripes (g), gold substrate (h2), and SiO₂ spacer (h1) are fixed at 40, 200, and 40 nm, respectively. A CH₃NH₃PbI₃ (MAPbI₃) perovskite film, with a thickness of t and optical constants from Ref. [37], is then laid on top of the metasurface. At the condition without metasurface, as shown in Scenario 1 in Fig. 1(b), the gold substrate reflects normally incident light and causes it to propagate in opposite direction

following Snell's law. When we introduce the metasurface at the bottom of MAPbI₃ film, the metasurface can deflect the reflected light, thereby enlarging the optical path within the film and promoting absorption. Eventually, the deflected light exits the film at a deflection angle of θ (Scenario 2 shown in Fig. 1(b)). If an appropriate metasurface is selected to increase the deflection angle further, the reflected light can be completely converted into a wave propagating along the *x*-direction, thereby entirely trapping the reflected light within the film (Scenario 3 shown in Fig. 1(b)).



Fig. 1. (a) Schematic of metasurface-assisted perovskite film absorption enhancement. Lower images are building blocks of the metasurface. (b) Schematic of absorption enhancement mechanisms. (c) Reflection phases of gold gratings with various nanostripe widths under the illumination of normally incident light with $\lambda = 700$ nm. The grating periods are all 150 nm. Pentagrams indicate four chosen widths of 20, 60, 80, and 120 nm. (d) Jointed reflected phase distributions of eight gratings with four chosen widths from (c), and *p* is set as 200 nm. The dashed line defines the wave front. Here, the position of *z* = 0 is defined as the top surface of the perovskite film, *t* = 100 nm, *g* = 40 nm, *h*1 = 40 nm, and *h*2 = 200 nm.

The spatial distribution of the metasurface-induced phase discontinuities governs the deflection of reflected light within the film. In this case, the phase gradient, $\partial \varphi / \partial x$, of the reflected light induces a wave vector along the *x*-direction following the generalized Snell's law, thereby redirecting the reflected light [38,39]. An appropriate spatial phase distribution can be achieved by exciting various LSPRs on the optical resonators (i.e., metal nanostripes). In the proposed method, the resonance wavelength is set to 700 nm due to the optical absorption of the ultrathin MAPbI₃ film decreases sharply at the long-wavelength range, in particular at wavelengths of approximately 700 nm [40]. In numerical calculations using the FDTD method to determine the geometric parameters of the metasurface, a plane wave source with transverse magnetic polarization was placed 200 nm away from the top surface (*z* = 0) of the MAPbI₃ film at a normal incidence. Periodic boundary conditions and perfectly matched layer boundary conditions were applied in the *x*- and *z*-directions, respectively.

Considering that phase variation of the reflected light can be achieved using resonators with different widths, we first calculated the phase response of a reflected beam with the wavelength of 700 nm from different structures comprising periodic gratings of varying nanostripe widths with a period of 150 nm. The phase shift between the reflected and source beams at z = 800 nm as a function of nanostripe width is plotted in Fig. 1(c). It can be observed that the phase shift gradually increases with enlarged nanostripe width as a result of the change in the phase delay caused by the LSPRs in the nanostripes. Four characteristic widths were chosen with successive

phase shift differences of approximately $\pi/2$, corresponding to widths of 20, 60, 80, and 120 nm, respectively (indicated with pentagrams in Fig. 1(c)). By successively joining the phase distributions of light reflected from periodic gratings with these four characteristic widths, we could equivalently represent the phase distribution of light reflected by the entire inhomogeneous metasurface, as shown in Fig. 1(d). In this configuration, interference between waves reflected from different units forms a new wave front (white dashed line), indicating an obvious deflection from the normal direction. Because the phase difference within the metasurface is nearly constant ($\pi/2$), the parallel wave vector of the reflected light, *i.e.*, $k_x = \partial \varphi/\partial x$, can be simply described as $2\pi/4p$.

Because the deflection angle is relative to the parallel wave vector of the reflected light, the deflection of the reflected light can be adjusted using metasurfaces with different periods, p. To demonstrate this, we discuss four systems with different phase gradients. For an infinite p, that is, a pure film without a metasurface, the phase gradient equals zero in the *x*-direction, as shown in Fig. 2(a). In this case, the reflected beam obeys Snell's law and propagates along the *z*-direction. The corresponding far-field scattering intensities at various angles are shown in Fig. 2(b), in which the scattering maxima can be found at $\theta = 0^{\circ}$. Figure 2(c) depicts the electric field



Fig. 2. (a) Designed reflection phase, (b) far-field intensity of scattering, and (c) E_x field distribution on the *x*–*z* plane of a pure perovskite film under the illumination of normally incident light with $\lambda = 700$ nm. (d) Designed reflection phase, (e) far-field intensity of scattering, and (f) E_z field distribution on the *x*–*z* plane of a perovskite film-metasurface assembly with a *p* of 240 nm under the illumination of normally incident light with $\lambda = 700$ nm. (g) Designed reflection phase, (h) far-field intensity of scattering, and (i) E_z field distribution on the *x*–*z* plane of a perovskite film-metasurface assembly with a *p* of 200 nm under the illumination of normally incident light with $\lambda = 700$ nm. (g) Designed reflection phase, (h) far-field intensity of scattering, and (i) E_z field distribution on the *x*–*z* plane of a perovskite film-metasurface assembly with a *p* of 200 nm under the illumination of normally incident light with $\lambda = 700$ nm. (j) Designed reflection phase, (k) far-field intensity of scattering, and (l) E_z field distribution on the *x*–*z* plane of a perovskite film-metasurface assembly with a *p* of 200 nm under the illumination of normally incident light with $\lambda = 700$ nm. (j) Designed reflection phase, (k) far-field intensity of scattering, and (l) E_z field distribution on the *x*–*z* plane of a perovskite film-metasurface assembly with a *p* of 150 nm under the illumination of normally incident light with $\lambda = 700$ nm.

distribution along the x-z plane, from which it is obvious that the reflected beam propagates with a wave vector along the z-direction. By contrast, for a finite p of 240 nm, a phase variation from 0 to 2π can be achieved at a spatial range of 960 nm in the x-direction, as shown in Fig. 2(d). Such a phase gradient results in a deflection angle, θ , of 31.84°, which can be obtained from the far-field scattering intensity angular distribution, as shown in Fig. 2(e). Compared with the case illustrated in Fig. 2(c), the electric field distribution along the x-z plane in Fig. 2(f) has an obvious deflection along the direction of propagation of the reflected beam. The electric field distribution shown in the enlarged area reveals localized electric fields at the corners of the gold nanostripes, indicating the excitation of LSPR in the nanostructures. The phase gradient can be further increased by decreasing p to 200 nm, as shown in Fig. 2(g). The far-field scattering intensity angular distribution shown in Fig. 2(h) reveals that this metasurface possesses a larger deflection angle ($\theta = 41.94^{\circ}$) and a lower scattering intensity than the metasurface with p = 240nm. Deflection along the propagation direction of the reflected beam is more apparent in the electric field distribution along the x-z plane, as shown in Fig. 2(i), in which LSPR can also be observed in the enlarged area. Furthermore, a larger phase gradient can be observed by reducing p to 150 nm (Fig. 2(j)). In this case, the reflection beam propagates primarily along the x-direction and is trapped within the perovskite film, causing the far-field scattering to mostly disappear, as shown in Fig. 2(k). The electric field distribution in Fig. 2(l) reveals that the electric field intensity in free space is nearly zero, with the electric field confined to the proximity of the perovskite film, indicating that the incident light is completely trapped within the film. These results demonstrate that the LSPRs are excited and the deflection of the reflected light is achieved through the use of an appropriately designed metasurface, which can be advantageous in terms of improving the absorption of the HOIP film.

3. Metasurface-assisted broadband optical absorption enhancement

We used the reflection and absorption spectra to demonstrate the optical absorption enhancement of a perovskite film following the introduction of a designed metasurface. Figure 3(a) presents the reflection spectra of perovskite films combined with metasurfaces (p = 150 nm) and gold gratings with different widths, all of which exhibit obvious reflection suppression in the long-wavelength range. Obvious dips are observed in the reflection curves of the grating structures, with the dip location gradually red-shifting as the width increases owing to changes in the excited LSPR modes. For the metasurface, however, the decrease in reflection in the long-wavelength range is broadband and even stronger than the decreases occurring in the grating structures, especially at wavelengths of approximately 670-750 nm, thereby demonstrating that the combined effect of LSPR excitation and light deflection can effectively suppress reflection in the perovskite film over a broadband range. The absorption (A) of the system can be calculated using the relationship A = 1 - R - T, where R and T are the reflection and transmission, respectively. Because of the presence of the Au substrate, T can be ignored in the considered wavelength range, thereby producing the absorption spectra plotted in Fig. 3(b). Compared with the pure film, all the gratings and metasurface systems exhibit obvious enhancements in absorption in the long-wavelength range, with the absorption of the metasurface being nearly 100% at a wavelength of approximately 670-750 nm. Since both metasurfaces and gratings are made of gold, the total absorption of systems contains the contributions from not only the perovskite layer but also the gold. To evaluate the absorption only from perovskite layer in metasurface and grating systems, we try to calculate the absorption of the selected regions by the following equation [13,41]

$$A(\lambda) = \omega \times Im(\varepsilon) \int |E|^2 dV, \tag{1}$$

where λ is incident wavelength, ω is corresponding angular frequency, $Im(\varepsilon)$ is the imaginary part of permittivity of materials and E is the electric field. Figure 3(c) shows absorption from the perovskite layer in grating and metasurface systems, and absorption of pure film calculated

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by Eq (1). It reveals that after subtracting the absorption from gold structures, the absorption of perovskite layers in gratings and metasurface systems are indeed enhanced at least 42% in gratings with width of 20 nm compared with pure film at a wavelength of 600-750 nm. These results demonstrate that the absorption of a perovskite film can be effectively enhanced by the excitation of LSPR in Au nanostripes and can be further enhanced by the deflection effect caused by the metasurface.



Fig. 3. (a) Reflection and (b) absorption spectra of pure perovskite film, film assembles gratings with four widths, and film-metasurface assembly with p = 150 nm. Here, the period of the gratings is fixed at 150 nm. (c) Absorption only from perovskite layer in grating systems, metasurface system (p = 150 nm) and absorption of pure film. (d) Reflection and (e) and absorption spectra of pure perovskite film and film-metasurface assembly with different p; here, the thickness of the perovskite film is fixed at 100 nm. (f) Absorption contributed by perovskite layer in metasurface systems with different p.

The absorption property of a perovskite film with a metasurface can be further varied by changing the structural parameters of the metasurface. Considering that the deflection angle can be altered by using a different p, we calculated the reflection spectra of perovskite films with metasurfaces with different values of p, as shown in Fig. 3(d). It is observed that decreasing pfrom 220 to 150 nm causes an obvious decrease in the reflection at longer wavelength owing to the enlarged optical path in the perovskite film at larger deflection angles. The corresponding absorption spectra shown in Fig. 3(e) reveal that the metasurfaces with the values of p = 150 nm can promote the system nearly total absorption at a wavelength 670-750 nm. We also calculate the absorptions contributed by perovskite layers in film-metasurface assemblies, and absorption of pure film as depicted in Fig. 3(f). The absorptions of perovskite layers are enhanced more than 106% in metasurface system with p = 220 nm and further to 146% in metasurface system when p = 150 nm at a wavelength 600 - 750 nm compared with those in pure film, thereby confirming the complete entrapment of the reflected beam within the perovskite film, as demonstrated in Section 2.

Metasurface-induced absorption enhancement can be effective over a wider range of perovskite film thickness because the excited deflection and LSPR effects are rarely dependent on the thickness of the film. We compared the absorption spectra of pure perovskite films with those of perovskite film-assembled metasurfaces (p = 150 nm) that have film thicknesses ranging from 80

to 140 nm, as shown in Figs. 4(a) and 4(b), respectively. For the pure film shown in Fig. 4(a), the absorption decreases rapidly with decreasing thickness, particularly at long wavelengths. This effect occurs because the absorption coefficient of MAPbI₃ gradually decreases with increasing wavelength. For the films with metasurfaces, by contrast, the total absorption maintains a high value even at thicknesses below 100 nm, particularly at long wavelengths. This result demonstrates that the absorption enhancement induced by the metasurface can be achieved at film thickness ranging from 80 to 130 nm. For thinner films, the reduction in the volume of perovskite owing to the presence of the metasurface will weaken the absorption enhancement effect; for thicker films, the difference in absorption between a pure film and a film with a metasurface will no longer be apparent because the absorption path will be sufficient.



Fig. 4. Absorption spectra of (a) pure perovskite film and (b) and film-metasurface assembly at various perovskite film thicknesses. The red-, black-, and blue-dashed lines correspond to wavelengths of 650, 700, and 750 nm, respectively. (c–e) Absorption of a pure film, total absorption of film-metasurface assembly (p = 150 nm) and absorption contributed by the perovskite layer in metasurface system at wavelengths of (c) 650, (d) 700, and (e) 750 nm as functions of film thickness. Here, the structural parameter p of the metasurface is fixed at 150 nm.

Despite the fact that the response wavelength of a designed metasurface is designed at 700 nm, the broadband property of the metasurface will enable its absorption enhancement to occur over a broadband wavelength range at various film thicknesses. To demonstrate this, we plotted the absorption of a pure film, total absorption of film-metasurface assembly (p = 150 nm) and absorption contributed by the perovskite layer in metasurface system (calculated from Eq (1)) at three different wavelengths 650, 700 and 750 nm as functions of film thickness (Figs. 4(c)–4(e), respectively). The absorption from perovskite layer is always stronger than that of the pure film in the film thickness range of 80 to 130 nm. For example, the absorption at 650, 700 and 750 nm of 100-nm thick perovskite layer in metasurface systems are enhanced 119%, 161% and 229%, respectively, compared with those in pure film. These results demonstrate that the introduction of a metasurface can achieve broadband absorption enhancement over a range of film thicknesses.

4. Wide-angle absorption enhancement

The discussions shown above only focus on the normal incident condition. However, for practical applications involving photoelectrical devices, the incoming light will not be incident only along

the normal direction; accordingly, the incidence angle tolerance of absorption enhancement should be considered. Here, we consider the perovskite film-metasurface (1D hybrid gratings) assembly at TM incidence as an example to study the wide-angle absorption enhancement of perovskite layer. Total absorption spectra of a pure film (t = 100 nm) and a film with metasurface (t = 100 nm, p = 150 nm) at various incidence angles (θ_i) are calculated, as shown in Figs. 5(a) and 5(b). In this case, incident light with positive and negative θ_i would possess parallel wave vector components along the -x and x directions, respectively. For the pure film, an obvious decrease in the absorption in the long-wavelength range is observed at θ_i values ranging from -60° to 60° owing to the aforementioned reduced absorption coefficient in this wavelength range. In addition, the absorption is slightly enhanced as θ_i increases in a manner that is symmetrical for positive and negative angles. This property arises from the enlarged optical path within the perovskite film when light is incident on an oblique plane. By contrast, the film with the metasurface effectively suppresses the decrease in absorption in the long-wavelength range, as shown in Fig. 5(b). Additionally, the absorption enhancement has an asymmetrical characteristic, with stronger absorption observed at negative incidence angles (the dark region above the white dashed line).



Fig. 5. Absorption spectra of (a) pure perovskite film and (b) film-metasurface assembly with p = 150 nm at various incidence angles. White dashed line indicates location of 0°. (c) Absorptions of pure perovskite film, film-metasurface assembly and from perovskite layer on metasurface at wavelength of 700 nm as functions of incident angle. (d) Total absorption of perovskite film-metasurface assembly and absorption only from perovskite layer on metasurface at three different incidence angles. (e) Fixed-angle absorption enhancement factor F_a as a function of incidence angle. (f) Total absorption spectra of pure perovskite film and film-metasurface assembly for wide-angle incident light. Corresponding wide-angle absorption enhancement factor F_w is plotted as a blue curve.

To demonstrate the wide-angle absorption enhancement provided by the metasurface, we plotted the absorptions of pure perovskite film, film-metasurface assembly and absorption contributed by perovskite layer on metasurface at wavelength of 700 nm as functions of the incidence angle in Fig. 5(c). For the metasurface system, the total absorption reaches approximately 100% within a range of θ_i from -40° to 10°, and absorption only from perovskite layer is over 60% from -60° to 60°, higher than the absorption attained by the pure film. The asymmetric enhancement occurring in the metasurface system can be explained in terms of the general Snell's law, under which the presence of a phase gradient differentiates incident light with positive and negative

incidence angles; for light with a negative angle, the deflection angle will be larger than that for light with a positive angle, leading to stronger absorption in the former case. In Fig. 5(d), the absorption spectra of the metasurface system at three different θ_i (-40°, 0°, and 40°) are compared. Although the absorption at long wavelength is enhanced at $\theta_i = -40^\circ$ compared with that at $\theta_i = 40^\circ$, it should be noted that, even in the latter case, the absorption is obviously stronger than that in pure film. The absorption of perovskite layer in metasurface system calculated by Eq (1) at $\theta_i = -40^\circ$, 0° , and 40° are similar to the total absorption of the system and estimated about 80% of total absorption. Thus the absorption of perovskite layer is enhanced more than 87%at $\theta_i = 40^\circ$ (113% at $\theta_i = -40^\circ$) compared with that in pure film at the wavelength of 600 - 750 nm, demonstrating the wide-angle absorption enhancement introduced by the metasurface at designed wavelength. To measure the enhancement of total absorption in metasurface system at visible wavelength region quantitatively, we calculated the absorption enhancement factor $F_{a}(\theta)$ at specific incident angle θ , defined as $F_a(\theta) = \frac{\int_{400}^{750} A_m(\lambda)d\lambda}{\int_{400}^{750} A_f(\lambda)d\lambda}$, $A_m(\lambda)$ and $A_f(\lambda)$ mean total absorption of metasurface system and pure film for incident light with wavelength λ . It represents the ratio between the integrated areas in the total absorption spectra (400 - 750 nm) of film-perovskite assembly and pure perovskite film. The F_a values at various incidence angles are plotted in Fig. 5(e), from which it can be observed that the values of F_a are larger than one in the angular range of -60° to 60° , confirming that the total absorption in the visible regime of the metasurface system is stronger than the pure film over the wide-angle range.

The broadband and wide-angle absorption enhancement properties can work together to enhance the absorption performance of perovskite film-metasurface assembly in practical application scenarios. As an example, we consider the absorption properties of perovskite films with and without metasurfaces under a wide-angle incident light. For simplicity, we assume that the light is incident over an angular range from -60° to 60° with a uniform angular distribution I_0 . In this case, the total absorption at a specific wavelength can be calculated as

$$Abs(\lambda) = \frac{\int_{-\pi/3}^{\pi/3} A(\theta) I_0 d\theta}{\int_{-\pi/3}^{\pi/3} I_0 d\theta},$$
(2)

where $A(\theta)$ is the absorption at wavelength λ and angle θ . Using the absorption spectra at the different incidence angles shown in Figs. 5(a) and 5(b), we calculated the total absorptions of the respective systems, as shown in Fig. 5(f). The absorption of the metasurface system is obviously stronger than that of the pure film in the long-wavelength range, which is conducive to the metasurface-induced LSPR and deflection phenomena in the metasurface. We could define the wide-angle absorption enhancement factor, $F_w(\lambda)$, as absorption enhancement factor for wide incident angle when incident wavelength is λ . It is defined as $F_w(\lambda) = Abs_m(\lambda)/Abs_f(\lambda)$, wherein $Abs_m(\lambda)$ and $Abs_f(\lambda)$ mean total absorption of metasurface system and pure film for incident light with a uniform angular distribution from -60° to 60° , respectively. This parameter represents the ratio between the total absorptions of perovskite film-metasurface assembly and pure film under a wide-angle incident light. The values of F_w as a function of wavelength are plotted as a blue curve in Fig. 5(f), from which it can be observed that the enhancement factor is more than one at wavelengths > 600 nm and gradually increases with the wavelength, achieving a value of three at wavelengths of approximately 750 nm. For practical applications the absorption contributed by gold should be subtracted, the absorption contributed by perovskite layer is estimated about 80%of total absorption in metasurface system based on our data, which is enhanced 82% compared with that in pure film under a wide-angle incident light at 600 - 750 nm. These results can be attributed to the enhancement of absorption in the long-wavelength range by the metamaterials. At shorter region far from target wavelength, the absorption of perovskite is not enhanced.

5. Absorption enhancement in two-dimensional metasurface case

All metasurfaces above are designed based on one-dimensional (1D) stripes, thus only the absorption enhancement of TM (i.e. x-polarized) incident light is examined. For potential applications of proposed metasurface-assisted absorption enhancement approach, a structure insensitive to the polarization is needed. Here we have tried to extend the 1D case to twodimensional (2D) case. We use four 40 nm-thick gold squares with side lengths of w1 = 20 nm, $w^2 = 60$ nm, $w^3 = 80$ nm and $w^4 = 120$ nm to build the 2D metasurface. Different blocks are arranged in a 4×4 layout with a separation of p = 150 nm, as depicted in Fig. 6(a). The thickness of perovskite film is fixed at 100 nm. To investigate the polarization independence of designed metasurface, we calculate reflection and total absorption of 2D perovskite film-metasurface assembly as shown in Figs. 6(b) and 6(c). The total absorption of 2D metasurface system is larger than that in pure film at 600-750 nm wavelength for both TE and TM incident light. The absorption enhancement factor plotted in Fig. 6(d) is introduced to evaluate the enhancement quantitatively and defined as the ratio between the integrated areas in total absorption of metasurface systems and absorption of pure perovskite film for different polarized light at the 600 - 750 nm wavelength. As shown in Fig. 6(d), the absorption enhancement in perovskite-1D gratings assembly (marked as "1D meta" in Fig. 6(d)) is sensitive to incident polarization, while in 2D system (marked as "2D meta" in Fig. 6(d)) changes little under different polarized light. Figure 6(e) depicts the absorption only from perovskite layer in 2D metasurface system and absorption of pure perovskite film, calculated by Eq (1). The absorption spectra in Fig. 6(e) is similar to that in Fig. 6(c), illustrating the equal absorption enhancement in perovskite layer for TE and TM polarization.



Fig. 6. (a) Schematic of 2D perovskite film-metasurface assembly. (b) Reflection and (c) total absorption spectra for pure perovskite film and 2D metasurface system with *TE/TM* incident illumination. Here the absorption is calculated by A=I-R. (d) Absorption enhancement factor of total absorption in 1D and 2D perovskite film-metasurface assembly for different polarization states, respectively. (e) Absorption of pure perovskite film and of perovskite layer on 2D metasuface with *TE/TM* incident illumination. Here the absorption enhancement factor of absorption contributed by perovskite layer in 1D and 2D perovskite film-metasurface assembly for different polarization states, respectively. (e) Absorption of pure perovskite film and of perovskite layer in 2D metasuface with *TE/TM* incident illumination. Here the absorption is calculated from Eq (1). (f) Absorption enhancement factor of absorption contributed by perovskite layer in 1D and 2D perovskite film-metasurface assembly for different polarization states, respectively.

The enhancement factor plotted in Fig. 6(f) presents the ratio between the integrated areas in absorption contributed by perovskite layer on metasurface and of pure film at the 600 - 750 nm. The result demonstrates that absorption enhancement of perovskite layer on 2D metasuface is nearly uniform for different polarized incident light.

6. Conclusion

In this work, we have proposed the plasmonic metasurface that can effectively enhance the absorption of ultrathin perovskite films in a broadband wavelength range under wide-angle incident light. Our FDTD simulation results reveal that the metasurface-induced excitation of LSPRs and deflection of reflected light within an ultrathin HOIP film can cause a significant enhancement of the absorption of the film. The far-field intensities, deflection angles, and electric field distributions can be effectively altered using metasurfaces with various geometric parameters. Compared with a pure HOIP film, the HOIP with an elaborately designed metasurface will have a significant enhancement in broadband optical absorption. We further demonstrate that this absorption enhancement is effective over a wide angular range. And the absorption enhancements insensitive to the polarization states have been obtained as well by extending metasurface structure to a 2D case. This metasurface-assisted absorption enhancement effect can be used to produce high-performance photoelectrical devices, such as solar cells and photodetectors.

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