Broadband antireflection and light-trapping enhancement of plasmonic solar cells

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In this work, we demonstrate broadband extraordinary transmission and antireflection in two-dimensional periodic metallic cuboids at optical frequencies. These phenomena originate from nonresonant excitations of surface plasmons, and represent high antireflection simultaneously for a broad spectral band and a wide angular range of incidence with polarization insensitivity. Based on this principle, we further introduce such metallic cuboids arrays into silicon solar cells. It is shown that high performance of light trapping in the cells can be achieved with a significant enhancement of the ultimate quantum efficiency. This study shows promising applications of plasmonic nanostructures to high-efficiency photovoltaic devices.

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I. INTRODUCTION

Enhancing the efficiency of solar cells has been of great interest in recent years. In general, there are three basic requirements for solar cells to achieve high efficiency.1–4 First, the cell surface must be highly antireflective for the broadband solar spectrum over a wide angular range such that the solar energy can be sufficiently absorbed and trapped in the cell. Second, the absorbed energy must efficiently produce photocarriers instead of generating heat or other effects. Third, the solar cells should also have high conductivity for the photocarriers to be effectively collected with minimum combination in the bulk.

Flat and homogenous metallic films are generally extremely reflective to light, although they satisfy the high-conductivity requirement of solar cells. However, from intense studies in recent decades, nanostructured metallic films have been demonstrated to have completely different properties. Such metallic nanostructures can support surface plasmons (SPs) (i.e., excitations of collective conduction-electron waves at the interface between a metal and a dielectric5) or spoof surface plasmons6 in the long-wavelength regime. A typical effect of the SP excitations is that they may lead to extraordinarily enhanced optical transmission through metallic films with significantly reduced reflection.7–9 Meanwhile, SPs can trap and confine light in dimensions much smaller than the wavelength near the metal/dielectric interface, which may greatly enhance the interaction of light with thin active dielectric (or semiconductor) layers. Therefore, properly engineered metal/dielectric nanostructures in principle can meet all the above three requirements for making ultrahigh-efficiency thin-film solar photovoltaic (PV) devices.10–14

Nevertheless, one of the drawbacks of SP excitations is the inherently narrow bandwidth of operation, which is directly associated with the plasmonic resonance mechanisms.7–9 Very recently, it was found both theoretically and experimentally that simple metallic gratings consisting of narrow slits can become transparent and completely antireflective for extremely broad bandwidths under oblique incidence.15–18 This unnatural and fascinating phenomenon can be explained either by the nonresonant excitation mechanism of SPs and SSPs,15 or by the anomalous impedance match between the grating and free space (or dielectric material) at the plasmonic Brewster angle.16 One of the various potential applications of the ultrabroadband transmission mechanism is to make long-dreamed-of transparent metals.15,17,19–22 But, for applications to antireflective photovoltaic devices, one-dimensional (1D) metallic gratings will suffer from two limitations. First, perfect ultrabroadband transmission can be achieved only for transverse-magnetic (TM) polarization while transverse-electric (TE) polarization is almost completely reflected.23 Second, even for the TM polarization, high ultrabroadband transmission occurs only within a narrow angular range of extremely inclined incidence.

In this paper, we extend the principles of ultrabroadband transmission through 1D metallic gratings to 2D cases for both TM and TE waves. The 2D structures are periodic metallic cuboids on dielectric or semiconductor substrates. Note that, in the literature, 2D subwavelength hole arrays perforated in metallic films have been widely used to study extraordinary optical transmission phenomenon.7,9,24 but the absolute transmission of these structures is usually very low. One of the reasons is that the waveguide modes in the holes have a cutoff frequency below which the wave fields inside the holes are evanescent. By contrast, the cuboid structures do not have this limitation. Therefore, here we demonstrate that such structures can achieve polarization-insensitive high transmission and antireflection simultaneously for a broad spectral band at optical frequencies and a wide angular range of incidence. Furthermore, by applying the periodic metallic cuboids as a broadband antireflection coating layer on silicon solar cells, we show that high performance of light trapping in the cells can be achieved with significant enhancement of the ultimate quantum efficiency. The investigation presents promising applications of plasmonic nanostructures to high-efficiency photovoltaic devices.

The paper is organized as follows. In Sec. II, we first show the experimental demonstration of broadband transmission for both TM and TE waves in 2D metallic cuboids in the optical frequency range. The experimental measurements agree reasonably well with numerical calculations based on the finite-difference time-domain method. A nonresonant feature has also been explored that depends on the spatial...
The advantage of this plasmonic approach is also discussed. Finally, a summary is given in Sec.VI.

Further, the efficiency comparison between this plasmonic solar cell is evaluated in Sec.IV. The quantum efficiency for this plasmonic solar cell is measured to range from 0° to 72° can be found in Fig.A1 in the Supplemental Material.

The phenomenon of high ultrabroadband transmission has been observed in 2D periodic metallic cuboids for λ > λ_{WDx1}. This is different from the effect in 1D metallic gratings, where TE waves are almost completely reflected in the long-wavelength range. In 2D periods, the transmission spectra become nearly flat at θ = 64° for long wavelengths λ > λ_{WDx1}, and the experimentally measured flat transmittivity is nearly 80%. Therefore, the high ultrabroadband transmission in 2D periodic metallic cuboids is similar to that of 1D metallic gratings under oblique incidence.

For TE polarization (with the electric field E parallel to the Y axis), significant transmission still occurs in Figs.1(e) and 1(f) for long wavelengths λ > λ_{WDx1}. This is different from the effect in 1D metallic gratings, where TE waves are almost completely reflected in the long-wavelength range. In Figs.1(e) and 1(f), the transmission only slightly decreases when θ increases towards 90°. In particular, the transmission spectra become nearly flat at θ = 64° for long wavelengths λ > λ_{WDx1}, and the experimentally measured flat transmittivity is nearly 80%. Therefore, the high ultrabroadband transmission in 2D periodic metallic cuboids is similar to that of 1D metallic gratings under oblique incidence.

For different ratios of w_x/d_x along the X axis, the maximum ultrabroadband transmission occurs around different optimal incident angles (θ_m). The calculated and measured θ-dependent TM transmission spectra with three different w_x/d_x ratios are shown in Fig.2. The experimentally observed optimal angles are θ_m = 68° in Fig.2(b) for w_x/d_x = 0.25, θ_m = 62° in Fig.2(d) for w_x/d_x = 0.375, and θ_m = 56° in Fig.2(f) for w_x/d_x = 0.5. Obviously, the angular range (area) of the incident angle corresponding to high ultrabroadband transmission broaden when the ratio w_x/d_x increases. For small w_x/d_x, high transmission occurs only near the optimal angle, but under conditions such as w_x/d_x = 0.5, significant transmission is observed in a wider angular range. Thus, we have demonstrated that high transmission and low reflection of 2D metallic cuboids can be achieved for both broad spectral bands and wide angular ranges.

The phenomenon of high ultrabroadband transmission has a nonresonant feature, which can be understood from the \(|\mathbf{E}|^2\) distribution. Figure 3(a) shows the calculated transmission
spectra of the sample under incidence angles of 0° and 68°, and the corresponding \([\textbf{E}]^2\) fields are shown in Figs. 3(c)–3(f) at two specific wavelengths. For normal incidence, one can see that a Fabry-Pérot (FP) resonance peak appears at \(\lambda = 1020\) nm. This wavelength satisfies \(\beta = 2h/N + \Delta_N\), where \(N \geq 0\) is an integer and \(\Delta_N\) is the redshift of the peak.\(^{15,27–29}\) Since the FP peak is due to the resonance effect, we can see that the electric field shows a standing-wave pattern in Figs. 3(c)–3(f) for comparison. This nonresonant feature can reduce the loss from the metal layer. [Here the scale is the same in Figs. 3(c)–3(f) for comparison.] This nonresonant feature can reduce the loss from the metal layer.

III. ANALYTICAL ANALYSIS OF BROADBAND ANTIREFLECTION ON 2D PERIODIC METALLIC CUBOIDS

The phenomenon of high broadband transmission and antireflection for 2D periodic metallic cuboids can be understood from the detailed analytical solutions of Maxwell’s equations in the Supplemental Material.\(^{26}\) From these analyses, the reflection coefficient can be expressed as

\[
r = \frac{A - \frac{n_s \beta_s}{k_0 \cos \theta} B \tan(\beta_s h) + i \frac{(n_s - 1) \beta_s}{k_0 \cos \theta} C}{A + \frac{n_s \beta_s}{k_0 \cos \theta} B \tan(\beta_s h) + i \frac{(n_s + 1) \beta_s}{k_0 \cos \theta} C},
\]

where

\[
A = \begin{cases} 1 & \text{(for TM)}, \\
|w_x/d_x|^2 & \text{(for TE)}, \\
|w_y/d_y|^2 & \text{(for TE)}\end{cases},
\]

\[
B = \begin{cases} w_x/d_x & \text{(for TM)}, \\
1 & \text{(for TE)}\end{cases},
\]

\[
C = \begin{cases} w_x/d_x & \text{(for TM)}, \\
|w_y/d_y| & \text{(for TE)}\end{cases},
\]

\[
k_0 = 2\pi/\lambda, \quad \beta_s = \text{the wave number in the slit. For TM polarization, the minimum reflection appears at } (\beta, w_x)/(k_0 d, \cos \theta) = 1 \text{ or } \tan(\beta h) = 0. \text{ Note that the FP resonance corresponds to } \tan(\beta_s h) = 0. \text{ Therefore, the optimal incident angle for high ultrabroadband transmission and antireflection satisfies}
\]

\[
\theta_m = \cos^{-1}[\tan(\beta h)/(k_0 d)]
\]

in 2D periodic metallic cuboids. In fact, when TM light is incident at the optimal angle \(\theta_m\), the reflection coefficient reaches the minimum:

\[
r_{\text{min}} = \frac{(1 - n_s) \tan(\beta h) + i(n_s - 1)}{(1 + n_s) \tan(\beta h) + i(n_s + 1)}.
\]
As indicated in Fig. 3(b), the $\theta_m$ calculated from Eq. (2) reasonably agrees with the simulated one from the FDTD method, particularly for the cases with low $w_x/d_x$. Most importantly, the experimentally measured results agree well with both the calculations for the FDTD method and the predictions from Eq. (2). All these results justify the current theory of the optimal angle for 2D periodic metallic cuboids.

For TE polarization, the absorbance decreases with increasing incidence angle, so the highest absorbance respectively. For TE polarization, the absorbance decreases with increasing incidence angle because of the high absorbance in the semi-infinite single-crystalline silicon layer acting as the absorber.

In our design, the aluminum cuboids have the periods $d_x = d_y = 160$ nm, the gap widths $w_x = w_y = 80$ nm, and the thickness $h = 80$ nm. The silicon oxide layer is required in order to avoid additional surface combination, since the dielectric spacer layer between the aluminum cuboids and the semiconductor can cause electrical isolation. The silicon oxide layer (20 nm) meanwhile offers a graded-index layer, which also reduces the reflection. The absorbance of the structure at a given wavelength ($\lambda$) is evaluated by $A(\lambda) = \int \text{Im}(\varepsilon) |E|^2 dV$, where $\text{Im}(\varepsilon)$ is the imaginary part of the permittivity, $\omega$ and $E$ are the angular frequency and electric field strength, respectively, and the integral is over the entire semi-infinite silicon layer. The normalized absorbance in the semi-infinite single-crystalline silicon layer at different incident angles are shown in Figs. 4(b) and 4(c) for the TM and TE polarization states, respectively. For TE polarization, the absorbance decreases with increasing incidence angle, so the highest absorbance value is at 0°. For TM polarization, however, the absorbance increases with increasing incidence angle because of the high ultrabroadband transmission mechanisms discussed above. In addition, the reflection spectra of this solar cell are given in Figs. A2(a)–A2(b) in the Supplemental Material, where the high antireflection of the solar cell can be observed. Because of the nonresonant feature of high transmission in the long-wavelength range, the loss from the metal layer is reduced, as indicated by the absorbance of the metal layer in Figs. A2(c)–A2(d) in the Supplemental Material. Since the antireflection effect and nonresonant transmission both significantly reduce the loss from the metal layer, most of the light energy can be trapped in the active layer. Consequently, high absorbance in the semi-infinite single-crystalline silicon layer can be achieved.

It is worthwhile to discuss the absorbance enhancement of the present solar cell design, which can be defined by the factor $\Lambda(\lambda) = A(\lambda)/A_{\text{ref}}(\lambda)$. Here $A(\lambda)$ is the absorbance of the silicon layer in this plasmonic solar cell, and $A_{\text{ref}}(\lambda)$ is the absorbance of the same layer of silicon without adjacent coating, i.e., the naked silicon layer. The absorbance enhancement for both TM and TE polarization states at different incident angles is shown in Figs. 4(d) and 4(e). For TM polarization, most of the enhancement factors $\Lambda(\lambda)$ are greater than unity, which indicates high performance of light trapping in the active layer. It is also shown that the enhancement factors for TE polarization are even better than those for TM polarization.

To further demonstrate that the present design is suitable for high-performance solar cells, we calculate its ultimate quantum efficiency (QE) which assumes that every absorbed photon with energy larger than the electronic band gap produces exactly one electron-hole pair. The ultimate QE then evaluates the fraction of the incident solar photons which are absorbed and converted to electron-hole pairs under the

IV. BROADBAND LIGHT-TRAPPING ENHANCEMENT OF PLASMONIC SOLAR CELLS BASED ON 2D PERIODIC METALLIC CUBOIDS

In the following, we will demonstrate that 2D periodic metallic cuboids can be used to design broad-angle and ultrabroadband solar cells based on the above high-transmission and antireflection effects. Consider the structure of the solar cell in Fig. 4(a) that consists of three layers. On the top is a 2D periodic array of metallic cuboids made of (low-cost) aluminum. In the middle is a 20-nm-thick silicon oxide layer, below which is the semi-infinite single-crystalline silicon layer acting as the absorber.

![Schematic of the plasmonic solar cell that consists of three layers. On the top is a 2D periodic array of metallic cuboids made of (low-cost) aluminum. In the middle is a 20-nm-thick silicon oxide layer, below which is the semi-infinite single-crystalline silicon layer acting as the absorber. Normalized absorbance in the silicon layer of this solar cell: (a) for TM polarization and (b) for TE polarization. The absorbance enhancement in the silicon layer of this solar cell: (c) for TM polarization and (d) for TE polarization. Here the parameters for this plasmonic solar cell are $d_x = d_y = 160$ nm, $w_x = w_y = 80$ nm, and $h = 80$ nm.](image)
large-angle incidence, the factor still exceeds 1.1. Therefore, high performance of the solar cell can be achieved for a wide angular range of incidence. If we assume that the incidence angle is from \( \theta_1 = -68^\circ \) to \( \theta_2 = 68^\circ \) for sunlight, the total quantum efficiency enhancement of the solar cell can be expressed as

\[
Q_{\text{EHM}} = \frac{\int_{\theta_1}^{\theta_2} Q_{\text{E}}(\theta) \cos \theta d\theta - \int_{\theta_1}^{\theta_2} Q_{\text{E}}_{\text{ref}}(\theta) \cos \theta d\theta}{\int_{\theta_1}^{\theta_2} Q_{\text{E}}_{\text{ref}}(\theta) \cos \theta d\theta} \times 100\%.
\]

Our calculations based on this equation show that the current plasmonic solar cell can achieve an enhancement of the ultimate QE by about 29.5\% due to the fact that 2D periodic metallic cuboids have the properties of wide-angle and broadband antireflection.

V. EFFICIENCY COMPARISON BETWEEN THE PLASMONIC SOLAR CELL AND TRADITIONAL SOLAR CELLS

It is well known that the traditional solar cells usually contain six parts. From bottom to top, they are the rear electrode, the back reflector, the active layer, the passivation layer, the top electrode, and the antireflection (AR) coating, respectively. For simplification, we ignore both the rear electrode and the back reflector, and consider only the other four parts, which play the following roles. (i) The active layer (such as the silicon layer) absorbs the sunlight and produces the photocarriers; (ii) the passivation layer efficiently reduces the surface recombination velocity and enhances the lifetime of the charge carriers; (iii) the top electrode collects the electrons and links to the external electrical circuit; and (iv) the AR coating, such as dielectric \( \lambda/4 \) AR coating, is used to enhance the light absorption in the active layer. Nowadays, in high-performance photovoltaic devices, the standard c-Si surface passivation scheme uses thermal SiO$_2$ in order to yield outstanding characteristic. Due to its long-term stability, SiO$_2$ is also a good choice. However, the top electrode, which is made of metallic material, suffers from its opaque covering and suppresses the solar energy coming into the device. For example, in a cell device where a metal grid electrode of 20 \( \mu \)m width and 200 \( \mu \)m period is used, 10\% of the solar energy is wasted due to the covering of the electrode.

The advantage of the present plasmonic solar cells is that 2D periodic metallic cuboids can combine the AR coating and the top electrode together, which inhibits the reflection of sunlight and reduces the loss caused by the previously used electrode covering at the same time. Actually, as shown in Figs. 6 and 7, the light trapping of this plasmonic solar cell is comparable with that of the traditional silicon solar cell using dielectric \( \lambda/4 \) AR coating. As we mentioned, the traditional solar cell uses \( \lambda/4 \) thick SiO$_2$ or SiN$_x$ film as the AR coating [as shown in Figs. 6(a) and 6(b)]. For example, 100-nm-thick SiO$_2$ film or 70-nm-thick SiN$_x$ film can significantly suppress the reflection of the light around the wavelength of 600 nm. (The refractive index of SiO$_2$ is taken as 1.47 and the refractive index of SiN$_x$ as 2.1 in the
calculation.) For a realistic comparison, the metal electrode should be taken into account and it covers about 10% of the surface area.\textsuperscript{42} Then based on Eqs. (4) and (5), the ultimate QE enhancement of the silicon solar cells containing these two dielectric AR coatings can be evaluated, as shown in Figs. 6(d) and 6(e), respectively. As a result, the total QE enhancement of the cells can reach 20.2\% for a SiO\textsubscript{2} AR coating and 27.7\% for a SiN\textsubscript{x} AR coating (as illustrated in Fig. 7). Interestingly, the total QE enhancement for the plasmonic solar cell can achieve 29.5\%, which is slightly higher than for the traditional ones using dielectric AR coatings (see Fig. 7). It is common that optical resonances may happen in metallic structures, but they usually depend on the incident angle. Thanks to the nonresonance mechanism we mentioned in Sec. II, this plasmonic solar cell based on 2D periodic metallic cuboids is not sensitive to the incident angle, which is comparable with the traditional solar cell using dielectric $\lambda/4$ AR coating. In particular, if the incident angle is within the range of $\theta = 0^\circ$–$45^\circ$, the ultimate QE enhancement of the plasmonic solar cell is obviously higher than one made with SiO\textsubscript{2} or SiN\textsubscript{x} AR coating. This feature originates from the fact that the 2D nanocuboids of metal can achieve polarization-insensitive high antireflection and light trapping simultaneously for a broad spectral band at optical frequencies and a wide angular range of incidence.

VI. SUMMARY

In summary, we have demonstrated high ultrabroadband transmission and antireflection of 2D periodic metallic cuboids at optical frequencies for both TM and TE polarized light, and, accordingly, these plasmonic structures can make crystalline silicon achieve very efficient polarization-insensitive light trapping for a broad spectral band in a wide angular range of incidence. It has been verified that such 2D plasmonic structures can be used to design wide-angle and ultrabroadband silicon solar cells with significant enhancement of the ultimate quantum efficiency achievable. These mechanisms may be applied to improve the performance of various other photovoltaic devices.

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FIG. 6. (Color online) (a) Schematic and (d) ultimate QE enhancement of a solar cell containing a SiO\textsubscript{2} $\lambda/4$ AR coating and a metal grid electrode. (b) Schematic and (e) ultimate QE enhancement of a solar cell containing a SiN\textsubscript{x} $\lambda/4$ AR coating and a metal grid electrode. (c) Schematic and (f) ultimate QE enhancement of the plasmonic solar cell based on 2D Al cuboids. The thickness of the SiO\textsubscript{2} AR coating in (a) is 100 nm, and the thickness of the SiN\textsubscript{x} AR coating in (b) is 70 nm. The parameters of the 2D Al cuboids are $d_x = d_y = 120$ nm, $w_x = w_y = 60$ nm, and $h = 80$ nm and the thickness of the passivation layer (SiO\textsubscript{2}) in (c) is 20 nm. The metal grid electrode accounts for 10\% of the surface area.

FIG. 7. (Color online) The total quantum efficiency enhancement of three types of silicon solar cells. S1 is the cell with 100-nm-thick SiO\textsubscript{2} AR coating; S2 is the cell with 70-nm-thick SiN\textsubscript{x} AR coating; and S3 is the plasmonic solar cell with 2D Al cuboids, where $d_x = d_y = 120$ nm, $w_x = w_y = 60$ nm, and $h = 80$ nm, and the passivation layer is 20-nm-thick SiO\textsubscript{2}. 
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