Design principles for morphologies of antireflection patterns for solar absorbing applications

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Two-dimensional surface texturing is a widespread technology for imparting broadband antireflection, yet its design rules are not completely understood. The dependence of the reflectance spectrum of a periodically patterned glass film on various structural parameters (e.g., pitch, height, shape, and fill factor) has been investigated by means of full-vectorial numerical simulations. An average weighted reflectivity accounting for the AM1.5G solar spectrum (λ = 300–1000 nm) was sinusoidally modulated by a rod pattern’s height, and was minimized for pitches of 400–600 nm. When a rationally optimized cone pattern was used, the average weighted reflectivity was less than 0.5%, for incident angles of up to 40° off normal. The broadband antireflection of a cone pattern was reproduced well by a graded refractive index film model corresponding to its geometry, with the addition of a diffraction effect resulting from its periodicity. The broadband antireflection ability of optimized cone patterns is not limited to the glass material, but rather is generically applicable to other semiconductor materials, including Si and GaAs. The design rules developed herein represent a key step in the development of light-absorbing devices, such as solar cells.

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1. INTRODUCTION

Antireflection (AR) techniques are used to suppress the reflection that occurs at interfaces between dielectrics; AR structures are incorporated into a variety of photonic applications, including solar cells [1], light-emitting diodes [2], and imaging systems [3]. The use of a single dielectric coating is the most traditional AR strategy, but it is effective for only a narrow range of wavelengths and angles [4]. To impart broadband AR, graded refractive index films [2,5] and two-dimensional (2D) surface patterning [6–10] have been proposed. In particular, 2D surface patterning is considered a facile and effective method; its AR properties depend on various structural parameters, such as pitch, height, shape, and fill factor [10,11]. For example, Lu and Lal reported Si conical pattern arrays with a few-micrometer thickness, exhibiting a near-unity absorbance over wavelengths 400–1100 nm and demonstrated a power conversion efficiency of ~10% for solar cells using the patterned arrays [12]. Yamada et al. fabricated moth-eye patterns composed of acrylic resin and acquired their reflectance lower than 1% in the wavelength range from 400 to 1170 nm [13]. Also, a number of simulation works on optimizing antireflecting patterns have been reported thus far [8,10,11,14,15]. For example, Song et al. calculated reflectance spectra of GaAs films with cone and parabolic patterns as a function of their heights, by using a rigorous coupled-wave analysis (RCWA) method [14]. Similarly, Choi et al. used a RCWA method to design parabolic patterns in polymer films by changing their heights [15]. However, no systematic strategy has been articulated thus far for designing these patterns to impart broadband AR reflection; unanswered questions include which shape is optimal for the objects within a pattern, why one object is more beneficial to antireflection than others, and how each parameter describing a pattern interacts with the incident light. Notably, finite-difference time-domain (FDTD) simulations used in this study are capable of accurately describing any complex morphology [4], which provides optimal antireflection patterns by exploring all of their structural parameters.

In this study, we investigated the reflection spectra of 2D-patterned glass films by conducting three-dimensional FDTD simulations (Fig. 1). We examined all available structural parameters for a 2D periodic pattern and studied how each parameter influences the reflectance spectrum. To understand the AR effects of a periodic pattern, we constructed an effective refractive index film model. By applying design rules, we
determined an optimal pattern morphology that imparts AR over a broad range of incident wavelengths and incident angles. The design rules for AR conditions are generic, and can be applied to other optical media; we demonstrated that the broadband AR effect could also be achieved in Si and GaAs films, which will facilitate the development of efficient semiconductor light absorbers.

2. DESIGN OF ROD PATTERNS: PITCH, HEIGHT, AND FILL FACTOR

To examine the AR effect of a periodic pattern, we studied a glass (n = 1.5) film with 2D square-lattice patterning. In fact, a lattice type (e.g., square lattice and triangular lattice) does not have significant impact on reflectivity [16]. For FDTD simulations, the spatial resolution along three orthogonal directions was set to be 10 nm. Periodic boundary conditions were applied to the horizontal directions to generate an indefinite periodic pattern, and perfectly matched layers were employed at the top and bottom extremes of the calculated structure. To focus on the effect of pitch, height, and fill factor independently, we first examined simple rod patterns. First, to investigate the effect of pitch (a), we calculated the reflectance spectra (λ = 300–1000 nm) of patterned glasses with various a, fixing the height (b) at 200 nm [Fig. 2(a), inset and upper panel]. The calculated reflectivity in the simulations was determined by R = 1 − T, where R is reflectivity and T is transmittance. Note that the reflectivity includes both diffuse and specular reflections. The calculated reflectance spectra revealed three key aspects of the effect of a [Fig. 2(a), upper panel]. First, the reflection spectra of the structures shifted to longer wavelengths with increasing a. Second, the overall amplitude tended to decrease monotonically with increasing a up to 400 nm. Third, a sharp minimum appeared at the wavelength equal to a (e.g., a minimum at λ = 400 nm when a = 400 nm).

These pitch-dependent reflection features can be partly understood by acquiring near- and far-field profiles at certain wavelengths [Figs. 2(b) and 2(c), respectively]. In the case of a = 200 nm, for a normally incident plane wave, the near-field profile was marginally distorted at λ = 400 and 900 nm, except for an intensified electric field in the vicinity of individual objects in the pattern [Fig. 2(b), upper two panels] [17,18]. In contrast, for a = 400 nm, the near-field profile acquired at λ = 400 nm (i.e., the wavelength for which there is a sharp minimum in reflectivity) showed a horizontally alternating electric field, which is indicative of a grating momentum [Fig. 2(b), lower left panel] [10]. For λ = 900 nm, the normal plane wave was nearly preserved [Fig. 2(b), lower right panel], identical to the two cases illustrated for a = 200 nm. Far-field simulations of both reflected and transmitted light yielded multiple spots with fourfold symmetry, evidence of a diffraction effect [Fig. 2(c)] [19]. Notably, the diffraction spots for a = 400 nm [Fig. 2(c), lower panels] were more intense than those for a = 200 nm [Fig. 2(c), upper panels]. Therefore, a sharp minimum in reflectivity at a certain wavelength is associated with the diffraction effect resulting from a periodic pattern. The diffraction is also responsible for the spectrum shift with changing a, according to the following one-dimensional grating equation [20]:

\[
a \sin \theta_{\text{incident}} - \sin \theta_{\text{diffracted}} = m \lambda.
\]  

Fig. 2. (a) Calculated reflectivity spectra of rod patterns of (top) 200 nm height and various pitches, and (bottom) 400 nm pitch and various heights. Inset, schematic of the simulated structure. (b) Electric field intensity maps illustrating the interaction between normally incident plane waves of the wavelengths (left two panels) 400 nm (right two panels) 900 nm, and patterned glasses with pitches of (upper two panels) 200 nm and (lower two panels) 400 nm. (c) Far-field representation of (left two panels) reflected and (right two panels) transmitted light for (upper two panels) a = 200 nm and (lower two panels) a = 400 nm. (d) Average weighted reflectivity of patterned glass, Si, and GaAs films as a function of fill factor. Inset, schematic illustration of a rod pattern’s fill factor.

Next, we investigated the effects of the pattern’s fill factor upon the reflectivity spectrum [black line, Fig. 2(d)]. The fill factor was defined as a volume ratio between the rod object and the whole unit cell [Fig. 2(d) inset]. For these simulations, a and b were fixed at 400 and 200 nm, respectively. To quantitatively assess the AR performance of periodic patterns, we defined the solar spectrum–average weighted reflectivity (Rave) as
where \( S(\lambda) \) is the spectral irradiance of the AM1.5 G spectrum at 1-sun solar intensity and \( R(\lambda) \) is the calculated reflectivity for a given wavelength \([4,10]\). The calculated results showed that \( R_{\text{ave}} \) changed gradually with increasing fill factor and was minimized at a fill factor of approximately 0.4. Using Si and GaAs, \( R_{\text{ave}} \) was investigated as a function of the pattern’s fill factor [red and blue lines, Fig. 2(d)]. For the high-refractive index optical media, the minimum \( R_{\text{ave}} \) was found at a fill factor of approximately 0.2. Taken together, one infers that an optimum fill factor depends on the bulk’s antireflection condition; the effective refractive index of a patterned layer should equal the geometric mean between the refractive indices of the material and air. As a result, an optimum fill factor is shifted to a smaller value with an increase in the refractive index of the bulk material.

Also, we conducted FDTD simulations to examine the effects of the pattern’s height \( h \), fixing \( a \) at 400 nm for a glass film [Fig. 2(a), lower panel]. Overall, the reflectivity spectra shifted to longer wavelengths as \( h \) increased. However, a sharp minimum of almost zero reflectivity appeared at approximately 400 nm for all these patterns, which as noted above is an effect of their 400 nm pitch. The redshifting with increasing \( h \) can be understood by constructing an effective refractive index film model, in which a patterned layer is replaced by a planar film with an average refractive index [Fig. 3(a) inset]. For three different pitches \((a = 100, 300, \text{ and } 500 \text{ nm})\), we compared the reflectivity spectra of patterned glasses with those of their equivalent film models [Figs. 3(a)–3(c)]. This effective refractive index film model reproduced the reflectivity spectra of the patterned glasses fairly well, particularly for wavelengths larger than \( a \) (i.e., \( \lambda > 300 \text{ nm for } a = 300 \text{ nm} \)); however, the pattern with \( a = 500 \text{ nm} \) exhibited a reflectivity minimum at \( \lambda = 500 \text{ nm} \), and the effective index model did not fit the spectrum for \( \lambda < 500 \text{ nm} \). The effective refractive index film model accounts well for the reflectivity spectrum features of a periodic pattern, except for short wavelengths where the diffraction occurs effectively. The effective refractive index model can be applied to quickly justify an optimal set of parameters to first-order, prior to more refined full-vectorial simulations.

To determine an optimum combinatorial set of \( a \) and \( h \), we calculated \( R_{\text{ave}} \) for rod patterns while varying \( a \) and \( h \) simultaneously for the three different materials: glass, Si, and GaAs. For the patterned glass film, the minimum \( R_{\text{ave}} \) of approximately 0.01 was found for \((a, h) = (600, 500) \text{ nm} \). The optimum range of \( a = 400–600 \text{ nm} \) arises because solar irradiance is most intense for green wavelengths \((\lambda = 400–600 \text{ nm}) \) \([10,19]\). On the other hand, when \( a \) is fixed, \( R_{\text{ave}} \) tends to oscillate with \( h \) [Fig. 4(a)]. The same overall trend on pitch and height is observed for patterned Si and GaAs films [Figs. 4(b) and 4(c)]. For both structures, \( R_{\text{ave}} \) tended to oscillate with \( h \), whereas it was minimized at \( a \approx 400 \text{ nm} \), regardless of \( h \). Interestingly, the overall \( R_{\text{ave}} \) with varying \( a \) and \( h \) was nearly the same for the semiconductor materials because they have similar material dispersion [Fig. 4(d)] \([21]\). The optimum range of \( a \) investigated herein is consistent with other works \([10,22]\). For example, Yu et al. claimed that the periodicity of a patterned surface should be slightly smaller than the wavelength range of interest to maximize the light absorption because a normally incident plane wave readily couples to the first reciprocal points in the momentum space with the maximum number of resonances \([22]\). These numerical findings on rod patterns are utilized to rationally design conical patterns for imparting broadband antireflection, as will be discussed in the next section.

\[
R_{\text{ave}} = \frac{\int S(\lambda)R(\lambda)d\lambda}{\int S(\lambda)d\lambda},
\]

Fig. 3. Calculated reflectivity spectra of rod patterns with \( a \) of (a) 100, (b) 300, and (c) 500 nm, compared to their corresponding models of effective refractive index film structures, for two different pattern heights or film thicknesses. Inset, schematic of a modeled effective refractive index film structure.

Fig. 4. (a)–(c) Calculated average weighted reflectivity of rod patterns with various pitches \((a)\) and heights \((h)\) for patterned (a) glass, (b) Si, and (c) GaAs films. Inset of a, schematic illustration of the simulated structures. (d) Optical constants \((n, k)\) of the used Si and GaAs materials.
3. EFFECTS OF CLOSELY PACKED CONICAL PATTERNS

We investigated the effects of the patterned shapes upon the periodic patterns’ AR performance [Fig. 5(a)]. It has been reported that moth-eye patterns, that is to say, closely packed cone patterns, are beneficial to broadband AR [9,23]. To rigorously study the effect of shape, we conducted FDTD simulations for patterned glass [Fig. 5(b)], Si [Fig. 5(c)], and GaAs [Fig. 5(d)] films with various shapes: rod, box, hemisphere, ellipsoid, cone, pyramid, and truncated patterns. For all shapes except for hemisphere, \((a, b)\) was fixed at (428, 400) nm. For the hemisphere shape, \((a, b)\) was (428, 214) nm. The calculated reflectance spectra showed that the conical shapes including cone and pyramid provided superior antireflection over the considered wavelengths 300–1000 nm, contrasting with the nonconical shapes including rod and box [Figs. 5(b)–5(d)]. The other shapes with a relatively gentle slope (e.g., hemisphere and ellipsoid) exhibited an intermediate antireflection ability. Notably, the shape effect was nearly identical for the three different bulk materials. These results on the effect of shape explain well the findings of previous works using conical or moth-eye patterns [9,12,13,23].

Next, we investigated the reflectivity of the cone patterns while varying their taper angles. For these simulations, the taper angle of the cones \((\theta)\) was varied from 20° to 86° for a glass film [Fig. 6(a) inset]. For a given \(\theta\) and \(h/a\), the closely packed cone pattern (fill factor = 0.26) is expressed as follows:

\[
a = 2h \tan(\theta/2)
\]

The calculation results showed that \(R_{ave}\) was significantly reduced for sharp \((\theta < 60°)\) cone patterns [Fig. 6(a)]. For instance, \(R_{ave}\) was less than 0.001 at \((h, \theta, a) = (400 \text{ nm}, 56°, 430 \text{ nm})\). This structural condition allows for a closely packed cone pattern to have \(a\) of approximately 500 nm (i.e., within an optimum range of pitch for diffraction). When sharper cones \((\theta < 60°)\) are used, \(R_{ave}\) is minimized at a larger \(h\) because the use of sharper cones allows them to be taller while still fitting within the optimum range of pitch.

![Fig. 5.](image1)

![Fig. 6.](image2)

It is noteworthy that for the cone patterns, \(R_{ave}\) did not exhibit clear modulation with \(b\). For a more complete investigation of parameters, we obtained a two-dimensional surface plot of \(R_{ave}\) as a function of pitch and height for different cone angles [Fig. 6(b)]. This surface plot clearly reveals the optimization strategies of the cone shapes toward broadband antireflection. The overall \(R_{ave}\) decreases with a decreasing taper angle. For a fixed taper angle, the \(R_{ave}\) approaches a minimum value for pitches in the range of 400–600 nm. To elucidate the improved AR performance of the periodic cones, we introduced a model of films of graded refractive index, in which cones were replaced by multilayer films of monotonically decreasing refractive indices. The reflectivity spectra of the cone patterns with \((h, \theta, a) = (200 \text{ nm}, 86°, 370 \text{ nm})\) and \((500 \text{ nm}, 39°, 350 \text{ nm})\) were compared with those of their equivalent film models with the same thickness \((t)\) [Fig. 6(c)]. The multilayer films were composed of 50 nm thick stacks with graded refractive indices corresponding to the cone geometry [Fig. 6(c) inset]. The graded refractive index film model reproduced the cone patterns’ broadband AR well, except for wavelengths shorter than 400 nm, which could be modeled better by considering the diffraction effect resulting from the patterns’ periodicity. Essentially, the cone and pyramid patterns provide the same antireflection ability as long as their fill factors are the same [Fig. 6(d)]. For close-packed arrangement, the fill factor of a pyramid structure is higher than that of a cone structure, which results in slight difference in reflectivity [Figs. 5(b)–5(d)].

4. ANTIREFLECTION PERFORMANCE AT OFF-NORMAL ANGLES

The reduced \(R_{ave}\) from the cone pattern was maintained even when off-normal incident angles were used, with both p- and s-polarizations [Figs. 7(a)–7(c)]. The angular \(R_{ave}\) of the cone
pattern was also reproduced well by the graded refractive index film model [black line, Fig. 7(a)]. For the patterned glass film, a cone pattern with \((b, \theta, a) = (400 \text{ nm}, 56^\circ, 430 \text{ nm})\) exhibited almost zero \(R_{\text{ave}}\) for both polarizations as long as the incident angle was less than 40° [red lines, Fig. 7(a)]. In contrast, for a rod pattern with the same height, pitch, and fill factor, the \(R_{\text{ave}}\) increased steadily as the incident angle increased, particularly for s-polarized light [blue lines, Fig. 7(a)]. The broadband AR of a cone pattern is not limited to a glass material but rather is a generic effect that can be used with other materials, including semiconductors [9,12,13,24]. We compared the reflectivity spectra and the angular \(R_{\text{ave}}\) of cone and rod patterns, for both Si [Fig. 7(b)] and GaAs [Fig. 7(c)]. For these simulations, we employed the same rod and cone patterns as used in Fig. 7(a) example. As discussed above regarding the Fig. 5 examples, the cone patterns dramatically reduced reflectivity over a broad range of wavelengths and incident angles. The exact reflectivity of each material depends on the details of its optical characteristics [22]; however, the same overall trend is observed for the semiconductor materials. The enhanced antireflection from the cone patterns is clearly visualized in the near-field profiles acquired at the wavelengths where the reflectivity is the minimum in Figs. 5(b)–5(d) [Fig. 7(d)]. For all the considered materials, the electric field is intensified within the cone, thereby alleviating the reflection. The design rules developed herein for AR patterns will be useful for enhancing the light absorption efficiency of solar cell devices, regardless of their material’s refractive index.

5. CONCLUSIONS

We studied design principles for the broadband AR performance of patterned glass films. The effects of various structural factors, such as pitch, height, fill factor, and shape of patterned objects, were investigated by means of FDTD simulations. The numerical simulation and the use of an effective refractive index film model suggested that the AR effect of a periodic pattern comprises a diffraction effect in conjunction with a geometrical effect. When a pattern of closely packed cones was used, the average reflectivity of the patterned glass film was less than 0.005 for a broad range of incident angles up to 40°. The design rules extend to semiconductor materials, such as Si and GaAs, and thus will provide a platform for designing high-efficiency light absorbers.

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