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Research Article

Broadband Low Reflection Surfaces with Silicon Nano-tube Square Arrays and Quantum Dot Layers

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Abstract

In this study, we take as a starting nanostructure which is already optimized in terms of the silicon nanopillar arrays' structure pillar height, pillar diameter, and filling ratio such that the optical reflection from its surface is very low (weighted average reflection 3.75 percent). Full-field Finite Difference Time Domain method is used to simulate electric and magnetic fields and calculate the reflection from the modified nanostructured substrate surfaces in 400nm-1100nm spectral range. In this paper, we present the optimization of the structure in terms of the silicon nanotube structure cavity diameter, step coverage of the dielectric thin film. As a result, the weighted reflection is decreased to 3.35 percent. We also want to simulate the quantum dot solution layer deposited over the nanostructure. We modeled the quantum dots as Lorentz dielectric and decreased the optical reflection even lower level of 3.1 percent. Optimization recipe is clearly presented, and the developed method is not only useful for square arrays but also for regular arrays of nanopillars in general for photovoltaic devices.

Keywords: Silicon, Nanopillars, Low reflection, Solar energy.

Geniş Bantta Düşük Yansıtmalı Kare Örgü Yapılı ve Kuantum Nokta Kaplı Silisyum Nanotüp Kaplı Yüzeyler

Öz

Bu çalışmada daha önceki çalışmamızda optimize edilmiş boyutları başlangıç nanoçubuk yapının mimarisi olarak aldık. Nanositun yüksekliği, çapı, doldurma oranı özelliklerine göre optimize edilmiş ve ağırlıklı ortalama ile hesaplanmış yüzey yansıması yüzde 3.75'tir. Tam alanlı sonlu farklar zaman düzlemi metodu kullanılarak 400nm-1100nm spektrumda nanoyapılı yüzeylerden yansıyan ışığın elektrik ve manyetik alanları simüle edilmiştir. Bu makalede, nanoçubukların içindeki oyukların çapı ve kaplanan ince film dielektrik kaplamanın düzensiz kaplanmasının yüzeyden optik yansıma üzerindeki etkisinin sonuçları sunulmuştur. Bu iki parametrenin optimizasyonu ile ağırlıklı ortalama yansıma yüzde 3.35 düzeyine indirilmiştir. Nano örgü yapının üzerine kaplanan kuantum nokta katmanının da etkisi simüle edilmiştir. Bu çalışmada kuantum noktaları Lorentz dielektrik olarak modellenmiştir ve simülasyonlar yansımanın yüzde 3.1 seviyesine indiğini göstermiştir. Optimizasyon reçetesi açık bir şekilde sunulmuştur ve geliştirilen bu metod sadece kare örgülü yapılar için değil fotovoltaikte kullanılan diğer örgülü nanoyapılar için de kullanışlı olacaktır.
Anahtar Kelimeler: Silisyum, Az yansıtma, Güneş Enerjisi.

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1. Introduction

Generally, crystalline silicon based photonic devices such as photodiodes, solar cells, phototransistors need low surface reflectance over a wide spectrum of light in order to have high external quantum efficiency and energy harvesting through outer circuits. One of the conventional methods to reduce surface reflection is to use single layer dielectric at specific wavelength but this does not reduce reflectivity for broadband [1,2] . SiO_2, SiN_x, TiO_2, Al_2O_3 dielectric materials are the most popular thin film materials for anti-reflection coatings. When double dielectric layers are used for this purpose, there are two reflection minimum in the reflection spectrum [1]. Unfortunately, in order to achieve a broadband low reflection, multi-layer band pass filter should be used but this requires many thin film layers and that increases the production costs. However, this is not an optimum solution to the broadband low reflection problem. Surface modification is needed in order to obtain broadband low reflection from the optoelectronic substrate surfaces. This modification can be achieved by using micro [3] or nano-size structures [4-15] over the device surfaces. In order to build such small features, researchers used mainly wet etching and dry etching methods. For example, industrial pyramidal surface texturing for crystalline silicon uses anisotropic wet-etching method. In other methods, dry etching can be used with ionized gases in the plasma vacuum chambers. For some studies both etching methods are used in combination [16,17].

Several research groups add some degree of randomness [8,15] in the periodic structures to decrease the optical reflection further. However, for large area applications, this can have some advantages as well as disadvantages in having a standard method of production.

Structure sizes show differences in these applications. Some applications use micro-meter size pillars [3] and some applications use nano-meter size pillars. For applications that need thin silicon film layers, nanometer size structures have to be used in order to achieve low reflection in broadband. To summarize, recently several types of surface nanostructures have been investigated using a variety of methods such as reactive ion etching (RIE), metal assisted chemical etching, laser etching, and low-frequency inductively coupled plasma etching [18-24].

If the pillars are tall then this can lead to quantum efficiency loss due to charge recombination in the pillar structure and the generated electron hole pairs cannot be extracted from the devices for energy harvesting [25]. Therefore, the height of the pillars should be optimized to get low reflectivity. When the height of the pillars is too short then the coupling of incident light into the silicon substrate is low. Moth-eye nano-pyramidal pillars use graded index refractive index to reduce reflection. Nanowires and nanocylinders that use Mie resonances (scattering) which result in overall decrease in reflection. Metallic nanoparticles use their plasmonic effect to direct electromagnetic energy through the substrate effectively and to decrease the reflection from the surface [26-31].

Optimization studies of the nanostructured surfaces in optoelectronic devices are still popular since the energy sector is very crucial for the economy of the countries. Even one percent increase in energy efficiency in solar cells mean millions of dollars saving in countries' economies. In order to obtain high efficiency in solar cells, low reflection from the surface irrespective of incident angles in wide wavelength spectrum is needed.

In this work, we optimized nanopillars filling ratio, height, apex angle, and calculated the weighted average reflection with respect to wavelength to get the overall efficiency improvement for the optoelectronic devices. We focused on square array type of nanopillars. In contrast with other structures such as black-silicon [16,25] or several photonic crystals (PC) [32,33], the optimization process we applied in this study is nonconformal antireflection coating and light trapping mechanisms presented not only let broadband reflection minimization over a broad angle of incidence but enhances absorption [34-36]. In addition, recombination losses are decreased by holding the total area of total area lower compared to micro pyramidal random texturing [22,37] or black-silicon [16,22,25]. We also use quantum dots to down-conversion of the

2. Methods and Simulations Results

In this letter, we present the optimization of the nanopillar square arrays in order to achieve broadband low reflection surfaces. When compared to a bare planar crystalline silicon wafer surface, the modified surface has significant reduction in reflection in 400nm-1100nm range. As a first step, we take the pillar period **a**=270nm, pillar diameter as **d**=190nm, pillar height as **h**=120nm which has 3.91 percent weighted average reflection using equation (1) in 400nm-1100nm spectral range (Figure 1).

We used Finite Difference Time Domain (FDTD) method to simulate photonic nanostructures. We placed monitors just below the nanopillars to calculate the transmission to the bulk substrate. We also placed a monitor above the structure to simulate the EM fields and calculate the reflected power. We also calculate the light absorbed in the nanostructure. The optical constants of crystalline structure and thin films are taken from [38]. We first optimized the cavity diameter in the nanopillar in the range from 40nm to 160nm and the minimum is obtained at 60nm cavity diameter. As seen Figure 2. Several different reflection curves for several different cavity diameter is also shown in Figure 3. Red curve has the lowest weighted total reflection of 4.65 percent. Over this Silicon nanotube square array structure, SiO_2 thin film is coated (placed) and since the lowest reflection occurs at 5nm thick SiO_2. We take s=5nm and varied the thickness of t. In this way, we simulate the step coverage effect, which is the result of the nature of etching processes, the vertical pillars can be obtained under special physical and chemical conditions [39,40]. It is important to have vertical pillars to get optimum reflection from such structures since as the pillar angle gets smaller, the reflection starts to increase. For large area applications, nano-imprint lithography can be used with a previously prepared template using nano-fabrication methods or nanosphere lithography as to include some randomness effect. We also realized that in order to increase the efficiency of photonic devices, we have to increase the optical power absorbed in the bulk of the active silicon layer. The absorbed light in the nanostructures part could be lost due to recombination and cannot contribute to the photocurrent with large percentage. Therefore, we have to maximize the absorption not in the nanostructures but the optical absorption in the bulk silicon active device region. We will pursue the study for applications that need the absorption maximized in the active region in another paper. **s/t** ratio varied from 0.1 to 1.0 increasing the t thickness parameter but keeping the **s**=5nm. When **s/t**=0.1 is used, we obtained weighted average reflection of 3.35 percent Figure 4. In order to decrease this reflection loss even further, step coverage concept is used. Since these pillars form nano steps and PECVD (Plasma enhanced chemical vapor deposition), LPVD (Low pressure vapor deposition), Sputtering (Coating of target material using generally argon plasma) coatings generally are nonconformal in nature. We also investigated its effect on reflection. The anisotropy can be increased further by optimization of the deposition conditions of the dielectric thin films e.g. gas flow rates, chamber pressure, substrate temperature, RF power. We used the cavities for two purposes, first is to get a better light trapping such that the reflection is decreased lower. Secondly, this will give a better stable low reflectivity when the incidence angle of light is changed. We will investigate this feature in future study. In Figure 5, spectral optical reflections for **s/t**=0.1 and for **s/t**=1.0 are shown and it is clearly noticed that the reflection is lower when the ratio is 0.1 over the 400nm-1100nm wavelength range.

Solar irradiance differs with wavelength; we can use the total reflection of light from the surface weighted with solar irradiance. This way, the performance of the low reflection loss performance can be quantified better. ASTM Air Mass 1.5 direct solar irradiance is used for the calculation. In this formulation, irradiance is multiplied with the wavelength and the integral is taken with wavelength. The numerator is composed of irradiance multiplied with wavelength and reflection. For a random polarization condition, both TE and TM polarizations are calculated and the average is taken to get result that is more accurate. We used the formula given below in calculation of the weighted average reflection in 400nm-1100nm wavelength range.

$$Weighted\ Average\ Reflection = \frac{\int_{400}^{1100} \frac{R_{TE}(\lambda)+R_{TM}(\lambda)}{2} I(\lambda)\lambda d\lambda}{\int_{400}^{1100} I(\lambda)\lambda d\lambda} \quad (1)$$

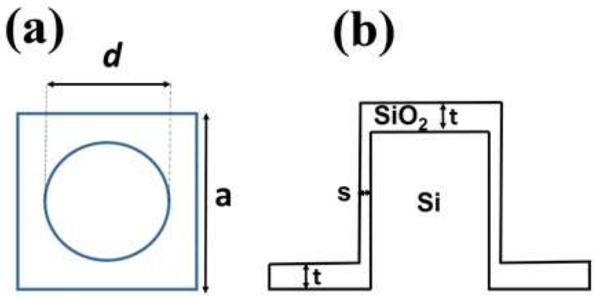

Figure 1: a) Unit cell of pillar array, b) Cross section of nonconformal coating of dielectric thin film.

The absorption in the UV region of the spectrum at the nanopillar array structure is particularly high, this is also a big issue if there is recombination in the nanostructure for energy extraction from an active device. The reflection from this range is also high from figure 5. The reflection in the UV region can be decreased by using quantum emitters like quantum dots for example. These nano materials can be prepared in solutions and coated by spin coating over the substrate. After drying a clean environment like glovebox systems the structure can be produced. In our simulations, we modeled these quantum dots as Lorentzian dielectric with oscillator strength of 0.6. **Weg**=3.72x10^{15} rad/s and lifetime of **1ns** [47]. Weg is chosen to correspond to 350nm wavelength so that the quantum dot layer absorbs in UV and scatter strongly with lower wavelengths too. As a result, the emitted radiation will be strongly absorbed by the nanostructure photonic crystal structure. By using this concept in the simulations, we decreased the weighted average reflection from 3.33 percent to 3.1 percent. This can be even decreased by using thicker layers of Qdots. In Figure6, the reflection reduction is especially noticeable in the UV region as our strategy in using the Qdots.

To our knowledge, this result is one of the best results for this type of nanostructured square array surfaces reported in the literature. The recent studies and their comparisons are given in Table I.

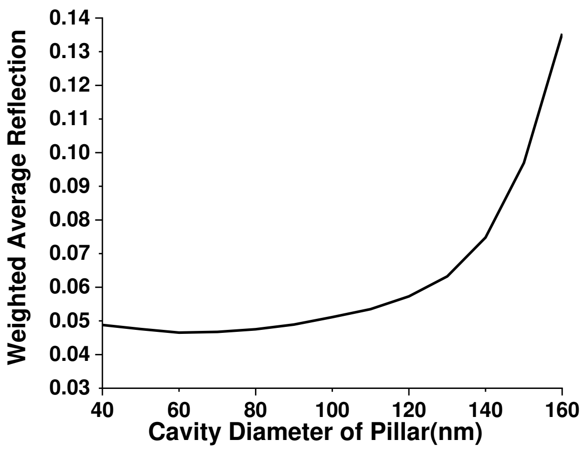

Figure 2: Reflection versus Cavity diameter of the silicon nanopillars.

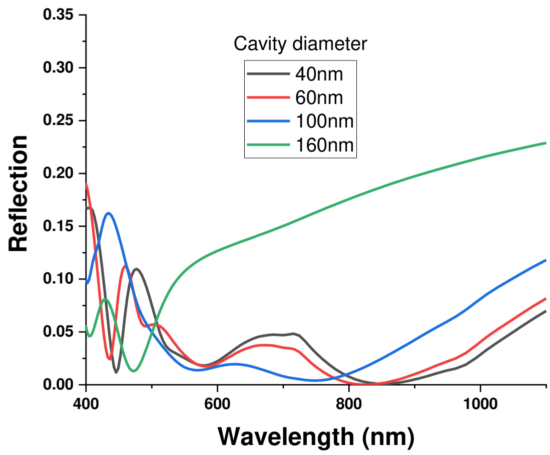

Figure 3: Reflection spectrum for several cavity diameters.

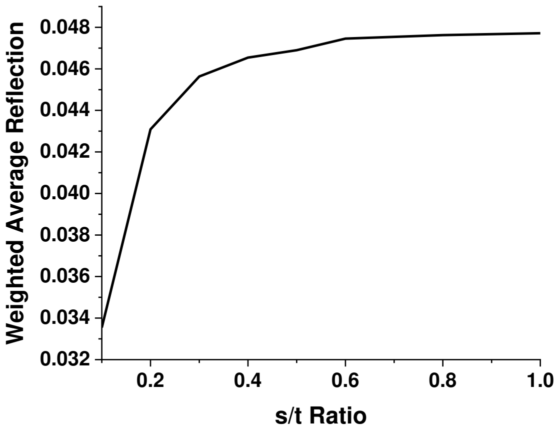

Figure 4: Weighted average reflection versus s/t ratio.

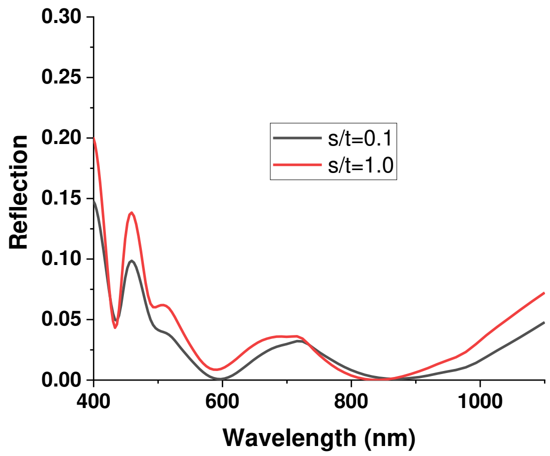

Figure 5: Reflection spectrum for s/t=0.1 and s/t=1.0 over 400nm-1100nm wavelength range.

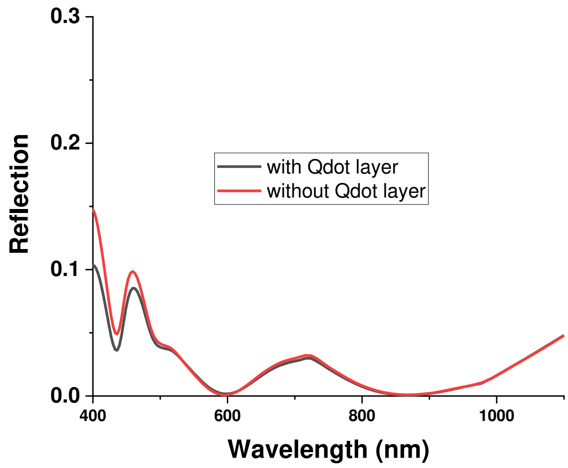

Figure 6: Reflection spectrum for surfaces with and without Qdot layer for comparison

3. Discussion

Nanostructured photonic arrays can be employed to achieve surfaces with low optical reflection. Use of step coverage and

Quantum emitters can have a great potential to decrease the optical reflection even further for broadband light absorbers and fabrication of photovoltaic devices with high efficiency. The table 1 shows the results of previous works related to nanostructures array surfaces. As noticed, our result of weighted average reflection of 3.1 is the lowest result when compared to the results in the Table 1.

Table 1. Comparison of recent results for the average reflections from regular square nanopillar arrays

Author	Year	R (Average)
J. Li et al. [43]	2009	>10 %
C. Lin et al. [44]	2011	4.64 %
J. Proust et al. [45]	2016	4 %
J. Kim et al. [46]	2021	7.1%

4. Conclusions and Recommendations

The reflection properties of the nanostructured silicon surfaces have been investigated and understood physically through weighted average reflection as a function of wavelength.

We optimized the silicon based square nanopillar arrays which is already optimized from our previous study and then we study further their reflection properties when we use silicon nanotube structure and optimized the cavity diameter in the pillar structure. We then used the step coverage concept which is the naturally occurring phenomenon in nanofabrication process and it also decreases the reflection from the surface of these nanostructures.

In the last step, we used Quantum emitters and modelled it as Lorentz dielectric layer. We obtained 3.1 percent weighted average reflection and to our knowledge this is the lowest reflection from this type of regular array of square array of nanostructures.

These modified nanostructured surfaces concept is useful for photovoltaic device applications having thin film silicon or active absorbing semiconducting material for optical energy harvesting through low surface reflection over a broadband optical spectrum [42]. In broad sense, thin film solar cells, photodetectors, phototransistors are the potential application of these nanostructured surfaces with high quantum efficiency.

In future work, we will experimentally realize the optimized nonconformal coatings of dielectric films and these periodic nanopillar array structure on a photodetector and demonstrate the quantum efficiency with respect to a control photodetector micro device.

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