Modeling of Optical Properties of Black Silicon/Crystalline Silicon

Introduction

The optical properties of crystalline silicon (c-Si) is one of the most well studied areas in the literature [1] due to its applications in photovoltaics [2], detectors [3], microlenses [4], filters [5] and silicon based optoelectronics [6]. One of the major disadvantages of applications of c-Si in optoelectronics is its significant reflectance (R) and low absorptance (A) in the visible range of wavelengths, in addition to its inherent indirect bandgap [7]. A surface modification of c-Si was introduced in the 1980’s by an unwanted side effect of Reactive Ion Etching (RIE). This modified c-Si had needle-shaped surface structure where needles were made of single-crystal silicon. As a result, the surface of Si appeared black in color, and hence the name “Black Silicon” (b-Si). Black silicon exhibits properties of very low reflectance and high absorptance in both the visible and infrared wavelength regions.

In recent years, several other methods of processing b-Si, such as electrochemical etching [8-10], stain etching [11-19], metal-assisted chemical etching using gold nanodots or silver [20-24], laser treatment, etc., have been developed. Of these, the laser treatment of Si has been developed by Eric Mazur and his group at Harvard University [25-30]. As a result of pulsed laser processing, the novel material created has surface texture that facilitates enhanced light absorption. The enhanced light absorption and low reflection of b-Si has prompted b-Si to become an active area of research in renewable sources of energy. Apart from the application of b-Si in solar cells, it has potential applications in image sensors [31], biosensors [32, 33], Micro Electro Mechanical Structures (MEMS) [34], light-emitting devices, antibacterial coatings [35] and gas sensors [36]. B-Si, owing to its high absorptance in the IR region, can be used for IR detection [37]. Photodetectors made of b-Si are expected to have increased sensitivity in the NIR and SWIR range of wavelengths. With the IR sensing properties of b-Si, true digital day-night imaging is possible; this has significant applications in aerospace, defense, security and transportation industries. Recently, a novel method to fabricate black poly-silicon (BPS) has been reported by Fekete et al [38]. In this method, BPS was fabricated on the surface of thermally oxidized Si by RIE at ~ cryogenic temperatures (~110°C to ~90°C). BPS is formed in Low Pressure Chemical Vapor Deposition (LPCVD) deposited poly-silicon thin film by Deep Reactive Ion Etching (DRIE) in the presence of SF₆ and O₂ plasma at ~ cryogenic temperatures. The potential applications of BPS extend to biological and chemical sensors due to the highly adjustable morphology and integrable fabrication technology. The theory developed in the present study can be extended to BPS by considering its spectral properties.

Abstract

Black Silicon, a surface modification of silicon has low reflectance and correspondingly high absorptance in the visible range of wavelengths making it more viable than crystalline silicon for applications in silicon optoelectronics. Like crystalline Si, black silicon has potential applications in solar cells, sensors, antibacterial surfaces and offers new opportunities. In this study, the optical properties of black silicon, in the visible, near and short wavelength infrared region, are simulated based on the effective medium approximation. The application of Helmholtz’s law of optical reciprocity to the two layer medium of black silicon on crystalline silicon is examined. In the long wavelength infrared region, the reported values of emissivity of black silicon are compared with those in the literature. In general, the emissivity of black silicon is higher than that of crystalline silicon.

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Experimental Methods in the Literature

Reactive ion etching

This method of producing b-Si uses SF₆ and O₂ gases to generate free radicals (F* and O*) [39-43]. These free radicals are responsible for etching c-Si, producing volatile products such as SiFₓ. These products, particularly SiFₓ, react with the free radicals to form a passivation layer of SiOₓFᵧ on a cooled Si surface [41]. Reflectivity can be controlled well by RIE. This way of texturing is ideal for very fragile substrates. The optimal reflectivity of uncontrolled textured cells is around 15% [32].

Figure 1 [44], shows the reflectance spectra of the Si samples, in the wavelength range of 200 to 1500nm, etched for different durations. The spectra of polished c-Si before etching are also shown for comparison purposes. It can be observed from the spectra that the optical reflectance spectra of c-Si is reduced significantly right from the onset of structure formation i.e., after 2 mins. The reflectance spectra further decreases with pore growth. After 5 mins, the reflectance spectra are almost completely suppressed and b-Si is formed. The decreased reflectance results in significant increase of absorptance in b-Si [44].

Mazur’s method

A Harvard University group [7] has developed a process in which b-Si is produced by irradiating Si with femtosecond laser pulses. After irradiation of Si surface with a gas containing mixture of SF₆/O₂ and other species, the surface of Si develops a self-organized microscopic structure of micrometer-sized cones. Sulfur(S) atoms are forced to the Si surface, creating a structure with a lower bandgap and therefore the ability to absorb longer wavelengths. The excitation of electrons from the bonding to anti-bonding states causes repulsive forces and disorder in the lattice structure while remaining thermally cold [45].

The roughened surface decreases the surface reflection greatly, but the sulfur atoms and structural defects, introduced by laser treatment, create more absorbing states in the sub-band gap region of Si [46, 47]. Also, the laser induced damage renders the surface of Si less active electronically [48]. Careful annealing process reduces the number of defects in the Si surface and improves the carrier mobility [28].

From Figure 2 [34], it can be observed that when only SF₆ is used as the plasma gas, the reflectance is very high. There is a significant decrease in reflectance when a mixture of SF₆ and O₂ is used. This is due to the formation of needle like structures that result in enhanced light trapping. The average reflectance of b-Si over the wavelength range of 200nm to 1100nm is 2.12%. The decrease in reflectance can be explained by diffraction effects in the shorter wavelength range and by moth ball effect in larger range of wavelengths [34].

Theory & modeling

Optical properties of materials are of importance for a variety of applications. These include applications in non-contact process monitoring and control as well as in the design of coatings, detectors, light emitting devices and waveguides.

Helmholtz’s reciprocity principle

The Helmholtz’s Reciprocity Principle describes the interaction of a ray of incoming light and the corresponding outgoing light with matter. These interactions involve reflections, refractions and absorption in a passive medium or at an interface. The bidirectional reflectance distribution function [49] represents such interactions. The 2-D model of a c-Si and b-Si system, considered for examining the reciprocity principle, in this study, is shown in Figure 3.
we use Multi-Rad.

of the multi-layers and its reciprocity, i.e., c-Si/b-Si and b-Si/c-Si, in order to simulate the wavelength dependent optical properties and polarization.

respectively. The depolarization factor \( g \) depends on the geometry for the dilute composite.

We consider a layer of vertically aligned b-Si array on a semi-infinite Si substrate, as shown in Figure 4 [52]. It should be noted that the assumption of the Si substrate being treated as opaque is a good one because the penetration depth of Si is usually small, except at wavelengths close to the indirect bandgap (around 1.1 µm wavelength). The basic assumption is that the characteristic geometric dimensions are much smaller than the wavelength of the electromagnetic waves.

Based on Bruggeman’s effective medium approximation, the effective dielectric function \( \varepsilon_{\text{eff}} \) of the b-Si layer can be calculated by solving the following equation:

\[
\frac{(1-f)(\varepsilon_{\text{air}}-\varepsilon_{\text{eff}})}{\varepsilon_{\text{eff}}} + \frac{f(\varepsilon_{\text{Si}}-\varepsilon_{\text{eff}})}{\varepsilon_{\text{eff}}} = 0
\]

(1)

where, \( \varepsilon_{\text{air}} \) and \( \varepsilon_{\text{Si}} \) are the dielectric functions of air and Si respectively. The depolarization factor \( g \) depends on the geometry and polarization.

In order to simulate the wavelength dependent optical properties of the multi-layers and its reciprocity, i.e., c-Si/b-Si and b-Si/c-Si, we use Multi-Rad.

**Multi-Rad**

Multi-Rad was created at Massachusetts Institute of Technology by Hebb et.al. [54]. Multi-Rad is a PC-based modelling software that enables the calculations of the radiative properties of thin-film stacks, with emphasis on semiconductor applications. The matrix method of multi layers forms the basis of the calculations. This model assumes that the layers are optically smooth and parallel and that the materials are optically isotropic. Thus the interfaces are considered to be abrupt. For a given multilayer stack, at specific temperature, wavelength and angle of incidence dependent radiative properties are obtained.

The radiative properties of the multilayer stacks are calculated using the matrix method of multilayers [55-57]. Radiation at a given wavelength is treated as coherent; therefore, interference effects are taken into account. The main assumption of the theory is that the layers are parallel and optically isotropic, the surface is optically smooth, and the area of the multilayer stack is much larger than the wavelength of the incident radiation.

Thermal radiation is usually well approximated as unpolarized. The directional transmittance and reflectance are calculated as simple average of s and p wave properties. This theory makes it general such that the predicted properties do not depend on classifying the radiation in a layer as coherent or incoherent.

The spectral absorbance (A) is calculated from:

\[
A(\lambda) = 1 - R(\lambda) - T(\lambda)
\]

(2)

It should be noted that the measurement of the optical properties, in the infrared range of wavelengths, is complex. This is in spite of the significant development in instrumentation relating to these measurements. In particular, the estimation of the wavelength dependent extinction coefficient, \( k \), and its reported values in the literature are sparse and sometimes, in a wide range, for the same wavelength. The measurement becomes more complicated for rough surfaces, multi-layers and highly infrared transparent materials. While spectral emissometry [58] provides a methodology for hemispherical measurements of the optical properties, Fourier Transform Infrared Spectrophotometry (FTIS) has become a standard tool for such measurements [59].

**Kirchhoff’s law**

In general, any object at a temperature \( T \) radiates electromagnetic energy. A perfect black body in thermodynamic equilibrium absorbs all light that strikes it, and radiates energy according to the law of radiative emissive power for temperature \( T \), universal for all perfect black bodies. Kirchhoff’s law [60] states that:

“For an object of any arbitrary material, emitting and absorbing thermal electromagnetic radiation at every wavelength in thermodynamic equilibrium, the ratio of its emissive power to its dimensionless coefficient of absorption is equal to a universal function only of radiative wavelength and temperature, the perfect black-body emissive power.”

In other words, Kirchhoff’s law describes the optical reflectance reciprocity as a material or structural property. The reflectance of an optical system is measured by detecting the light reflected off of the system when a source of light is incident on the system. If reciprocity holds for the material, the detector response is the same when the source and detector positions are switched.

**Emissivity**

Emissivity (\( \varepsilon \)) of the surface of a material is its effectiveness in emitting energy as thermal radiation. All objects do not radiate infrared (thermal) energy equally. An ideal black body has an emissivity of 1.0 and no other material can radiate more thermal energy at a given temperature. Materials have value of \( \varepsilon \) between 0 and 1. The emissive and reflective behavior of most materials is similar in the visible and IR regions. However, some materials may differ in their emissive and reflective behavior.
Hemispherical Emissivity:

\[ \epsilon = \frac{M_e}{M_{oe}} \]

where,

\( M_e \) - radiant flux emitted by the surface
\( M_{oe} \) - radiant flux emitted by a black body at the same temperature as the surface.

Directional Emissivity:

\[ \epsilon_\Omega = \frac{L_{e,\Omega}}{L_{oe,\Omega}} \quad (4) \]

where,

\( L_{e,\Omega} \) - radiant flux emitted by the surface
\( L_{oe,\Omega} \) - radiant flux emitted by a black body at the same temperature as the surface.

For the modeling purposes in this study, the samples considered are b-Si coated on c-Si wafers of 200 mm diameter (usually referred to as “8 inch”), thickness 725 µm. The doping is p-type with dopant concentration of 1 x 10\(^{16}\) cm\(^{-3}\). The thickness of b-Si is varied in the range of 1 – 10 µm. In order to investigate the reciprocity in the optical properties, R, T and A of c-Si/b-Si are compared, with those of b-Si/c-Si, for b-Si thickness of 5 µm, in Table 1.

Results and Discussion

The unique features of b-Si—improved light trapping and decreased reflectance make it an ideal candidate for solar cell applications. The low reflectance of b-Si indicates that the refractive index is lower than that of c-Si. Just as in the case of porous silicon (p-Si) [59, 61, 62], the effective medium approximation [50-51] can be used to determine the optical constants of b-Si. The optical constants thus obtained are effective refractive index (\( n_{\text{eq}} \)) and effective extinction coefficient (\( k_{\text{eq}} \)).

Figure 5 [63] illustrates the optical constants, i.e., the effective refractive index and effective extinction coefficient of b-Si obtained by using the effective medium approximation. An abrupt change in the extinction coefficient in the wavelength range of 900 – 1100 nm is observed; this corresponds to the energy gap of crystalline silicon.

From Figure 6(a), for b-Si/c-Si, it can be seen that with increase in thickness of b-Si layer, the reflectance of the sample is strongly influenced. For 10 micron thick b-Si layer, the reflectance is below 1% (R10), and is very close to zero. In Figure 6(b), for c-Si/b-Si, there is no effect of change in thickness of b-Si on the reflectance of the multilayer. Since reflectance is a surface property, it remains the same as that of c-Si. In the lower wavelength region, i.e., from 0.4 to 1.0 µm, the reflectance of c-Si/b-Si sample is almost uniformly changing. Beyond the wavelength of 1.1 µm, the reflectance behaves similarly with change in wavelength. The fluctuations in R are the result of interference effects.

By comparing Figures 7(a) and 7(b), we observe that the transmittance for b-Si/c-Si and c-Si/b-Si are the same and behave similarly with wavelength. With increase in thickness of b-Si, the transmittance decreases. The transmittance becomes almost zero when the thickness of b-Si is 5µm (T5) and is ~ zero for samples with 10 µm (T10) thick b-Si, throughout the entire range of wavelengths considered in this study. Though the influence of b-Si on the reflectance of c-Si/b-Si is negligible [Figure 6(b)], the transmittance is affected by the presence of b-Si in c-Si/b-Si [Figure 7(b)]. In the region of the energy gap of c-silicon (1.1µm), there is a change in the transmittance which becomes less pronounced as the thickness of b-Si layer increases [Figure 7(b)]. From Figure 8(a), it is clear that b-Si has positive effect on the absorptance. As the thickness of b-Si increases, the absorptance of b-Si/c-Si increases and approaches almost unity [Figure 8(a)]. Though the change in the absorptance with respect to wavelengths below the energy gap (1.1 µm) is very small, the absorptance in the wavelength regions beyond the energy gap has improved. However, in the case of c-Si/b-Si, the influence of varying thickness of b-Si is minimal [Figure 8(b)]. The influence of interference effects and diffraction effects is less prominent in the lower range of wavelengths.

In these calculations, all the values of reflectance, transmittance and absorptance are approximated to 3 decimal places. In Table 1, a comparison of the R, T and A of c-Si/b-Si and b-Si/c-Si, with respect to c-Si is presented. The thickness of b-Si, considered in this comparison in Table 1, is 5 microns. The R for c-Si/b-Si is higher than that of b-Si/c-Si. Correspondingly, A for c-Si/b-Si is lower than that of b-Si/c-Si. This result is independent of the wavelengths considered in this study.

In Table 2, the emissivity values of n type Si and b-Si, from the literature, are compared for selected long wavelengths in the infrared. As can be seen in this table, the emissivity of b-Si is generally higher than that of n-Si. The emissivity of b-Si decreases with increasing temperature. Due to free carrier absorption, the emissivity of n-Si, increases with increase in temperature. At

![Figure 5](https://example.com/figure5.png)
higher temperatures and longer wavelengths, the emissivity of n-Si is higher than that of b-Si (T = 873 K, 973 K; Wavelength = 12.5 µm, 15.0 µm).

From a solar cell perspective, the National Renewable Energy Laboratory (NREL) has reported the best efficiency of b-Si solar cell; the reported efficiencies are 16.8% [64-66] and 18.2% [67]. Based on the decrease in reflectance losses of b-Si, the anticipated improvement in efficiency, assuming the device performance to be similar to that of c-Si [68], should be about 30%.

**Conclusion**

Optical properties of black silicon have been investigated in the above study. Effective medium approximation and the Helmholtz’s optical reciprocity have been examined for a two layer model that consists of black silicon and crystalline silicon. The results show significant differences in reflectance and absorptance for this two-layer model. Comparison of the emissivity of n-Si and b-Si, in the infrared long wavelength range, has been presented.
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