Investigation of mis-estimation of structure of amorphous silicon films in ellipsometric modeling


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Abstract

Dielectric functions for thin films of hydrogenated amorphous silicon (a-Si$_{1-x}$:H$_x$) with varying hydrogen content $x$, void concentration and surface roughness may be constructed from measured dielectric functions for amorphous silicon (a-Si) by use of the tetrahedron model, scaling procedures, dielectric formulation, and the effective medium approximation (EMA). The measured dielectric functions may correspond to relaxed or unrelaxed (ion-implanted) a-Si. Ellipsometric measurements on thin films of a-Si$_{1-x}$:H$_x$ are often fitted with such dielectric functions to obtain film parameters such as void concentration and surface roughness, with the fit quality being assessed from the value of the unbiased estimator $\sigma$. Due to the strong preparation dependence of the a-Si$_{1-x}$:H$_x$ lattice structure, it may not be clear whether it is relaxed or unrelaxed dielectric functions which are appropriate for the fit. In this work, dielectric functions are constructed for a-Si$_{1-x}$:H$_x$ thin films using unrelaxed a-Si dielectric functions, and fitted using relaxed a-Si$_{1-x}$:H$_x$ dielectric functions and Levenberg–Marquardt non-linear regression. It is demonstrated that a small value of $\sigma$ may be obtained despite the incorrect choice of relaxation state. Comparison of the input and output void concentration and surface roughness shows significant mis-estimation in the fits. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Hydrogenated amorphous silicon (a-Si$_{1-x}$:H$_x$) has received considerable attention in scientific research due to its attractive electrical and optical properties. Numerous structural, optical, and electrical measurements have been conducted on amorphous materials in an attempt to understand the materials and thereby tailor their properties to best suit different applications. Spectroscopic ellipsometry (SE) [1–3] can be used as a non-destructive technique to determine the microstructure of amorphous materials, especially silicon-based materials such as non-hydrogenated amorphous silicon (a-Si) [4–9] and a-Si$_{1-x}$:H$_x$ [10–13]. For instance, by analyzing the complex dielectric functions of an amorphous thin film with appropriate models, information regarding the surface roughness and void concentration of the film can be obtained with relatively high accuracy.

In analyzing the SE spectra of a-Si$_{1-x}$:H$_x$, the Bruggeman effective medium approximation (EMA) [13,14] is usually used to model a sample as a mixture of different materials. In the standard approach, the surface roughness of the film can be

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modeled as a mixture of a-Si$_{1-x}$,H$_x$ and voids, while the bulk film is usually treated as a mixture of a-Si$_{1-x}$,H$_x$ and voids. In these models, however, precise knowledge of the dielectric functions of the various components is required in order to obtain a reliable estimation of the structural parameters. This requirement is particularly critical for the amorphous component because the structure of a-Si, depends strongly on the preparation procedures [10,13]. So far, the dielectric functions used for modeling a-Si$_{1-x}$,H$_x$ have been based on certain references in the literature without consideration of the preparation method. It is known that ion-implanted a-Si is structurally and optically distinct from annealed or relaxed a-Si [4–9]. Fried et al. [8,9] attributed this difference to the presence of a significant amount of defects such as dangling bonds and distorted tetrahedral bond-angles in the unrelaxed material. It therefore may not be clear, particularly for thin films of amorphous hydrogenated silicon prepared by novel methods, whether it is relaxed or unrelaxed dielectric functions which are appropriate for the fit.

In the present work, the scope of the problem is assessed by constructing dielectric functions for a-Si$_{1-x}$,H$_x$ thin films using unrelaxed a-Si dielectric functions, and fitting using relaxed a-Si$_{1-x}$,H$_x$ dielectric functions and Levenberg–Marquardt non-linear regression on the roughness and void concentration parameters. The values of the parameters at input and output, and of the mean-square deviation $\sigma$ are compared and discussed.

2. Method

The tetrahedron model [13] is applied to construct the complex dielectric functions of relaxed (unrelaxed) a-Si$_{1-x}$,H$_x$, by using the dielectric functions for relaxed (unrelaxed) a-Si [8]. This model constructs the optical response of the amorphous alloys by use of the scaling procedure, dielectric formulation, and EMA [13,14]. For details we refer to the work of Mui and Smith [13,15,16]. The complex dielectric functions constructed for relaxed and unrelaxed a-Si$_{1-x}$,H$_x$ components with various hydrogen concentrations are shown in Figs. 1(a) and (b), respectively. It is seen that the relaxed and unrelaxed spectra exhibit different peak position and amplitude. Moreover, as the hydrogen content increases, the complex dielectric functions shift to higher photon energy with lower peak values.

The complex dielectric functions for the unrelaxed a-Si$_{1-x}$,H$_x$ film are constructed for the structure illustrated in Fig. 2, consisting of three layers. The first layer is a surface roughness layer, modeled using the EMA, consisting of 50% voids, 50% layer 2. Layer 2 is modeled also using the EMA, consisting of a mixture of unrelaxed
a-Si_{1-x}:H (see Fig. 1(b)) and voids. The substrate is smooth silicon dioxide (SiO\textsubscript{2}). Fractional void concentrations in layer 2 were varied from 0.01 to 0.26 in the simulations. The thickness of layer 1 was allowed to vary from 0 to 0.005 \(\mu\)m, while the film thickness (layer 2) was fixed at 1 \(\mu\)m. The incidence angle was set to be 75°. The results, however, do not depend on the incidence angle. The spectral range was set to 1.7–5.0 eV, where 1.7 eV is slightly higher than the band gap of a-Si:H. Due to the large absorption coefficient above the band gap and the large film thickness, the films behave like bulk materials and the influence of the substrate is minimal. This is important in order to avoid interference caused by reflection of light from the film/substrate interface.

The dielectric functions constructed as described above were then fitted using complex dielectric functions for relaxed a-Si:H (see Fig. 1(a)), applying a non-linear regression based on the Levenberg–Marquardt method [3]. The void concentration and surface roughness are numerically determined by minimizing the mean-square deviation \(\sigma\). The fits were done for the case (1) that the thickness of surface roughness is known, and only the void concentration is a fitting parameter, and (2) for the case that both surface roughness and void concentration, are unknowns and are fitting parameters.

### 3. Results

We denote the void concentration and the thickness of the surface roughness by \(C_{\text{void}}\) and \(RT\), respectively. The fitting error of the void concentration and the thickness of surface roughness are denoted by \(\Delta_{\text{void}}\) and \(\Delta RT\), respectively, i.e.; \(\Delta_{\text{void}} = |C_{\text{void}} - C_{\text{fit}}|\), similarly for \(\Delta RT\). Although several structures with different hydrogen contents were studied, we will concentrate on the structure of a-Si\textsubscript{0.9}:H\textsubscript{0.1} for the sake of brevity, because it is found that presence of hydrogen in the structure barely changes the magnitudes of \(\Delta_{\text{void}}\) and \(\Delta RT\).

#### 3.1. Case (1): RT known and \(C_{\text{void}}\) varies

The mean-square deviations \(\sigma\) and the \(\Delta_{\text{void}}\) deduced from the best fits are shown in Fig. 3 as a function of \(C_{\text{void}}\). For \(C_{\text{void}} = 0.01\) and \(RT = 0\), \(\sigma\) is found to be \(~0.077\). In SE analysis, \(\sigma\) also called the unbiased estimator of the deviation of the templates and the fitting results, is often used as a figure of merit to quantify the quality of the fit. The smaller is \(\sigma\), the better the fit. From this point of view, it might be concluded that a good fit to an unrelaxed structure can be obtained with relaxed dielectric functions. But the corresponding error value \(\Delta_{\text{void}}\) is 0.13, considerably greater than the void concentration itself. For void fractions < 0.1, \(\sigma\) is smallest for small \(C_{\text{void}}\), but the error in estimated void fraction is largest. For \(C_{\text{void}} > 0.1\), \(\sigma\) improves with increasing \(C_{\text{void}}\); however, \(\Delta_{\text{void}}\) tends to saturate to a value of \(~0.08\), indicating a limit for the resulting \(C_{\text{void}}\). It is known that a variation of the void concentration can change the amplitudes of the dielectric functions [17], modifying the shape of the dielectric functions for...
relaxed a-Si$_{1-x}$:H$_x$ to be closer to that of the unrelaxed spectra. However, it does not change their peak position. The accuracy limit is thus probably due to the intrinsic differences in peak locations of the spectral shape between the relaxed and unrelaxed dielectric functions.

We note that the presence of surface roughness improves $\sigma$ considerably. However, the improvement of $\sigma$ is accompanied by an increase in $\Delta$void.

### 3.2. Case (2): RT and $C_{\text{void}}$ vary

In this case, both surface roughness and void concentration are allowed to vary in the regression. The $\Delta$void and $\Delta RT$ of the best fit are shown in Fig. 4(a). The amplitudes of $\Delta$void are approximately half in comparison with those in Fig. 3, ranging from 0.05 to 0.07; moreover, increasing the surface roughness displaces the entire curve to lower values of $\Delta$void without saturation and overlapping between individual curves. In contrast, $\Delta RT$ increases monotonically as a function of $C_{\text{void}}$ and shifts to higher values with increasing surface roughness over the whole range of $C_{\text{void}}$.

The corresponding mean-square deviations are shown in Fig. 4(b). The amplitudes of $\sigma$ are about one order smaller than in Fig. 3. For $RT \leq 0.002 \mu m$, $\sigma$ decreases significantly with increase in void concentration from $\sim 0.017$ to $\sim 0.0095$. Increasing surface roughness also causes a decrease in $\sigma$. However, for $RT = 0.005 \mu m$, the general profile of $\sigma$ deviates from other curves and increases markedly with increasing void concentration.

### 4. Discussion

In analysis of SE data, the structure of a sample composed of a-Si: H, voids and surface roughness is commonly determined by minimizing the mean-square deviation $\sigma$ in a non-linear numerical fitting process, and the size of the minimum $\sigma$ is taken as an indication of the reliability of the fit [3,13,18]. However, in our fitting results, small values of $\sigma$ may be obtained despite the incorrect choice of relaxation state. Additionally no correlation is observed between the mean-square deviation and the errors $\Delta$void and $\Delta RT$. For any given a-Si film, and in particular for films prepared by novel methods, there is an ambiguity in the whether relaxed or unrelaxed dielectric functions are the appropriate choice for the fitting procedure. We have demonstrated that the magnitude of $\sigma$ is not a reliable indicator.

### 5. Conclusion

Complex dielectric functions for structures containing a-Si$_{1-x}$:H$_x$ with various void concentrations and thickness of surface roughness were
constructed using the tetrahedron model and EMA from dielectric functions for unrelaxed a-Si. The constructed spectra were then fitted using dielectric functions for relaxed a-Si$_{1-x}$:H$_x$ and Levenberg–Marquardt regression. Small values of the mean-square deviation $\sigma$ were obtained in the fit despite the incorrect choice of relaxation state. Additionally, no correlation between $\sigma$ and the errors between input and output film parameters $\Delta$void and $\Delta RT$ was found. The results indicate that it is unreliable to use mean-square deviation $\sigma$ as a measure for the reliability in determining the real structure of an a-Si$_{1-x}$:H$_x$ film of unknown composition.

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References