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Complete parameterization of the dielectric function of microcrystalline silicon fabricated by plasma-enhanced chemical vapor deposition

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The dielectric functions of microcrystalline silicon (μc-Si:H) layers deposited by plasma-enhanced chemical vapor deposition have been determined by applying real-time spectroscopic ellipsometry (SE) in an attempt to construct an optical database for μc-Si:H. The μc-Si:H dielectric functions have been parameterized completely by a dielectric function model that employs two Tauc-Lorentz peaks combined with one Harmonic oscillator peak. This parameterization scheme has been applied successfully to describe the structural variation from hydrogenated amorphous silicon (a-Si:H) to highly crystallized μc-Si:H. Moreover, to express the microstructure of μc-Si:H, the μc-Si:H structural factor k has been defined based on the amplitude of the E2 optical transition with a critical point energy of 4.3 eV. From the value of k, a variety of Si microstructures, including complete a-Si:H phase (k = 0), μc-Si:H with a-Si:H-rich grain boundaries (k ~ 0.5), and μc-Si:H with void-rich grain boundaries (k = 1), can be distinguished. The μc-Si:H structures estimated from the above SE analyses show excellent correlation with those deduced from the Raman spectroscopy. From the SE analysis procedure developed in this study, the layer thickness as well as the microstructure of μc-Si:H can be characterized rather easily. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4704158]

I. INTRODUCTION

Microcrystalline silicon (μc-Si:H) layers, generally composed of nanometer-scale crystalline Si grains (~20 nm) with hydrogenated amorphous silicon (a-Si:H) phase at the grain boundaries,1–4 have been applied widely to fabricate a-Si:H/μc-Si:H tandem-type solar cells.5–10 For the deposition of μc-Si:H by plasma-enhanced chemical vapor deposition (PECVD), SiH4 highly diluted with H2 is used as the source gas.1–10 With the variation of the hydrogen dilution ratio R = [H2]/[SiH4], the microstructure and volume fraction of μc-Si:H grains change drastically1–3,5,6. The performance of μc-Si:H solar cells is quite sensitive to R, and the maximum efficiency is obtained when the μc-Si:H layer is prepared in the phase transition region where a fully a-Si:H phase changes to a μc-Si:H/a-Si:H mixed phase.5 Unfortunately, the optimum R value is not universal and the μc-Si:H microstructure varies depending on other deposition conditions, including a growth temperature,1,11 pressure,4 and rf power.1,6 Thus, the characterization of the μc-Si:H microscopic structure is of significant importance for the optimization of μc-Si:H solar cells.

Nevertheless, the nucleation and the following grain growth processes of μc-Si:H are highly complicated,5,11,12 and the structural characterization of μc-Si:H layers is in general difficult. To determine the crystallographic orientation of μc-Si:H grains, x-ray diffraction has been employed13 although the a-Si:H phase cannot be characterized from this technique. To estimate the volume fractions of the μc-Si:H and a-Si:H components, Raman spectroscopy has been employed widely.14 However, this technique is rather insensitive to the μc-Si:H microstructures, and the performance of μc-Si:H solar cells differs significantly even when the Raman spectra are quite similar.4 In particular, recent studies confirm that the presence of the a-Si:H phase at the grain boundaries is quite important to prevent the post-oxidation and to achieve higher solar cell efficiencies.4,5 For the accurate determination of μc-Si:H microstructures, it is necessary to develop a more reliable characterization technique.

Several studies have shown that, depending on the μc-Si:H microstructures, the optical properties in the visible/ultraviolet (UV) region change largely.15–17 Specifically, the optical constants (or dielectric function) of μc-Si:H show intermediate features between single crystalline silicon (c-Si) and a-Si:H, and the optical response is roughly proportional to the c-Si and a-Si:H fractions.16 Moreover, in the μc-Si:H dielectric function,15,16 there exist two absorption peaks in the UV region. These peaks correspond to the critical points in c-Si and are generally referred to as E1 and E2 peaks.18 The width of these transition peaks is inversely proportional to the size of μc-Si:H grains and allows the characterization of grain sizes from the dielectric function.19 Thus, from the μc-Si:H dielectric function, detailed evaluation of the μc-Si:H microstructure is possible.

In the last two decades, various μc-Si layers, fabricated by PECVD,15,16,20–22 CVD,23 magnetron sputtering,17 and solid phase crystallization,24,25 have been studied intensively by applying spectroscopic ellipsometry (SE). From SE, the μc-Si:H layer thickness can also be characterized, in addition to the optical properties resulting from μc-Si:H microstructures. Accordingly, SE can basically be utilized as a fast and reliable measurement technique for μc-Si:H layers. However, to extract various physical properties from SE spectra, the
parameterization of dielectric functions is required. So far, the c-Si dielectric function has been parameterized using various models, whereas the Tauc-Lorentz (TL) model and Cady-Lorentz model have been employed widely to show the dielectric function of a-Si:H layers. Although the TL model is expressed by only five parameters, the c-Si dielectric function has been described using a large number of model parameters (~20 parameters). In such cases, it is quite difficult to determine the μc-Si:H properties based on these model parameters. For the extraction of μc-Si:H microstructures from the dielectric functions, therefore, it is necessary to develop a more simple parameterization scheme.

In this study, we have determined the dielectric function of μc-Si:H layers prepared at different R using real-time SE. The resulting μc-Si:H dielectric functions have been expressed by a simple model that has three transition peaks with only nine variables. From this model, the structural variation from a-Si:H to highly crystallized μc-Si:H has been described successfully. As a result, we have developed a SE analysis method, which allows the characterization of the layer thickness as well as the microstructure of μc-Si:H.

II. EXPERIMENT

We prepared Si:H layers (μc-Si:H or a-Si:H) by a conventional PECVD system equipped with real-time SE. The Si:H deposition was carried out using a substrate temperature of 190 °C, a pressure of 300 mTorr, and a SiH4 flow rate of 5 SCCM (SCCM denotes cubic centimeter per minute at STP). In this study, a series of Si:H layers were deposited by varying R. In this case, a H2 flow rate was changed from 50 SCCM (R = 10) to 500 SCCM (R = 100), and the Si:H layers were deposited using an rf plasma (13.56 MHz) with an input power of 64 mW/cm² and an electrode spacing of 4 cm. These growth conditions yield relatively slow deposition rates that decrease from 0.11 nm/s (R = 10) to 0.01 nm/s (R = 100). We deposited quite thin Si:H layers with a thickness of 20 nm on Si(100) substrates covered with 3-nm-thick native oxides for the SE characterization.

It has been shown that the Si:H layer deposition at low R leads to the a-Si:H phase (incubation layer) formation at the initial deposition stage, and the thickness of the a-Si:H incubation layer reduces with increasing R, accompanied by the increase in the μc-Si:H nucleation density. In the above deposition conditions, the deposited layers are initially amorphous at 20 ≤ R ≤ 30, whereas the immediate μc-Si:H nucleation occurs on the substrate at R ≥ 50. To suppress the a-Si:H phase formation at low R (20 ≤ R ≤ 30), we prepared μc-Si:H layers on a highly crystallized μc-Si:H layer (R = 50). In this case, the samples have a slightly complicated structure of μc-Si:H (R = 20–30, 20 nm)/μc-Si:H (R = 50, 15 nm)/SiO2 (3 nm)/c-Si substrate. On the other hand, the fully a-Si:H (R = 10) and μc-Si:H (R ≥ 50) layers were prepared directly on the SiO2/c-Si substrate.

In order to investigate the validity of our SE analysis method, we also prepared a series of the μc-Si:H layers with different thicknesses ranging from 39 to 1912 nm using R = 50. For this series, to obtain a higher deposition rate (0.2 mm/s), the μc-Si:H layers were deposited using a VHF plasma (30 MHz) with an input power of 255 mW/cm² and an electrode spacing of 2 cm. When the thick μc-Si:H layers (≥ 250 nm) were deposited on glass substrates, however, the deposited layers peeled off from the substrates due to high compressive stresses in the μc-Si:H layers and the poor adhesion to the substrates. Thus, to improve the μc-Si:H layer adhesion, we deposited the μc-Si:H on ZnO:Al-coated glass substrates. The ZnO:Al layers with a thickness of 100 nm were prepared by rf magnetron sputtering using a ZnO target (Al2O3: 1 wt. %). The sputtering deposition was carried out at room temperature using a pressure of 0.6 Pa, an rf power of 50 W, and an Ar flow rate of 9 SCCM.

The SE measurements were performed using a rotating-compensator instrument (J. A. Woollam, M-2000). For the μc-Si:H layers deposited on the SiO2/c-Si substrates using different R, real-time SE spectra were collected with a measurement repetition time of 10 s to determine the dielectric function of the Si:H layers. The real-time SE spectra were analyzed by using a global error minimization (GEM) scheme, in which the overall analysis is made self-consistently under the assumption that the dielectric function of the deposited layer is uniform toward the growth direction. In this analysis, the structural uniformity toward the growth direction is essential for the accurate determination of dielectric functions. To minimize such structural non-uniformities, we have analyzed quite thin Si:H layers with a thickness of 20 nm. On the other hand, the SE spectra of the μc-Si:H layers (R = 50) formed on the ZnO:Al/glass substrates were obtained from conventional ex-situ measurements. In this case, the Scotch tape was pasted onto the roughened backside of the glass substrate (0.5 mm in thickness) to suppress the influence of the backside reflection. Raman spectroscopy using an Ar laser at 514.5 nm was used to characterize the structures of the Si:H layers.

III. RESULTS AND DISCUSSION

A. Dielectric function of μc-Si:H

Figure 1 shows the dielectric functions of the Si:H layers deposited with different R. As a reference, the c-Si dielectric function reported previously is also shown. The dielectric functions of the Si:H layers in Fig. 1 were obtained from the SE analysis using GEM. For the analysis, we employed an optical model consisting of ambient/surface roughness layer/Si:H bulk layer/SiO2 (3 nm)/c-Si substrate. In the case of R = 20 ≤ R ≤ 30, however, an optical model of ambient/surface roughness layer/μc-Si:H bulk layer/μc-Si:H bulk layer (R = 20–30)/μc-Si:H bulk layer (R = 50)/SiO2 (3 nm)/c-Si substrate was employed. The room-temperature dielectric functions shown in Fig. 1 have been obtained using a procedure established previously.

When R = 10, the ε2 spectrum indicates a single broad peak due to the formation of the a-Si:H phase. At R ≥ 20, the layer structure becomes μc-Si:H, and the ε2 spectra show intermediate features between a-Si:H and c-Si. The two sharp peaks in the ε2 spectrum of c-Si show the E1 and E2 optical transitions. These transitions arise from the direct interband transitions at critical points. The E1 critical point (3.4 eV) is due to the transition from Λ(3) (valence band) to Λ1 (conduction band) in the first Brillouin zone (BZ), whereas the E2 critical point (4.3 eV) does not correspond to a single well-defined
B. Construction of $\mu$c-Si:H model

From the $\mu$-c-Si:H dielectric functions shown in Fig. 1, we have developed the optical database for $\mu$c-Si:H. This optical database has been constructed, so that (1) the smooth variation of the dielectric function from a-Si:H to $\mu$-c-Si:H phase can be expressed and (2) the number of analytical parameters becomes as small as possible to avoid complications. In particular, to express the $\mu$-c-Si:H dielectric function numerically, each dielectric function in Fig. 1 was fitted using various dielectric function models. The dielectric function of a-Si:H can be expressed simply using the TL model. We also employed this TL model to describe the fundamental light absorption in $\mu$-c-Si:H. This is quite important to realize the smooth variation of the dielectric function from a-Si:H to $\mu$-c-Si:H. In addition to the fundamental transition, there are $E_1$ and $E_2$ transitions in $\mu$-c-Si:H. To minimize the numerical parameters, we have modeled the $\mu$-c-Si:H dielectric function by considering three optical transitions of the $E_1$, $E_2$, and fundamental transitions.

For the modeling of the $E_1$ and $E_2$ transitions in $\mu$-c-Si:H, various combinations of dielectric function models, including the Lorentz model,\textsuperscript{31} Gaussian model,\textsuperscript{37} Harmonic oscillator (HO) model,\textsuperscript{31,38} and TL model, have been investigated. As a result, we found that the best fitting to the experimental spectra can be obtained when the $E_1$ and $E_2$ peaks are expressed by the TL and HO peaks, respectively, with the fundamental peak expressed also by the TL model. In the TL model, the dielectric function is described by five independent parameters: namely, the amplitude parameter ($A$), broadening parameter ($C$), band gap ($E_0$), peak position of an $E_2$ peak ($E_2$), and an $e_1$ value at high energies ($\epsilon_1(\infty)$).\textsuperscript{27} On the other hand, the expression for the HO model used for the $E_2$ transition ($\epsilon_{E2} = \epsilon_1 - i\epsilon_2$) is given by

$$\epsilon_{E2}(E) = \frac{A_{E2}C_{E2}E_{E2}}{E_{E2}^2 - E^2 + ic_{E2}E + 0.25C_{E2}^2},$$

(1)

where $A_{E2}$, $C_{E2}$, and $E_{E2}$ denote the amplitude parameter, broadening parameter, and peak position of the $E_2$ peak, respectively.\textsuperscript{39} In our case, the dielectric function of $\mu$-c-Si:H is modeled by

$$\epsilon(E) = \epsilon_{\text{fun}}(E) + \epsilon_{E1}(E) + \epsilon_{E2}(E).$$

(2)

In Eq. (2), the subscripts of “fun” and “$E_1$” denote the TL model used for the fundamental and $E_1$ transitions, respectively. Since the TL and HO models have five and three parameters, respectively, the total number of the parameters in Eq. (2) becomes 13.

Figure 2 shows the $\epsilon_2$ spectra of (a) $\mu$-c-Si:H prepared at $R = 100$ and (b) c-Si analyzed by the dielectric function model of Eq. (2). In the fitting analysis, we fixed the peak positions of the $E_1$ ($E_{E0,E1} = 3.360$ eV) and $E_2$ ($E_{E2} = 4.250$ eV) transitions. In the case of the TL model, the $\epsilon_2(\infty)$ values are also fixed to $\epsilon_2(\infty) = 1$ (fundamental transition) and $\epsilon_2(\infty) = 0$ ($E_1$ transition). It can be seen that the dielectric function model developed in this study provides excellent fitting to the $\mu$-c-Si:H layer as well as c-Si. We observed similar fitting for the other $\mu$-c-Si:H dielectric functions shown in Fig. 1.
FIG. 2. ε2 spectra of (a) μc-Si:H prepared at R = 50 and (b) c-Si analyzed by the μc-Si:H dielectric function model. In this figure, the open circles show the experimental spectra shown in Fig. 1, and the solid lines indicate the results of the fitting analyses using the two TL peaks with one HO peak.

μc-Si:H model, there are only nine free parameters, if we exclude the E1 and E2 peak positions and ε1(∞) values. However, it should be emphasized that our model is quite practical and has been developed to provide a simple parameterization scheme for μc-Si:H.

Table I summarizes the TL and HO parameters extracted from the fitting analysis of the μc-Si:H dielectric functions. The result for the c-Si shown in Fig. 2(b) is also listed in Table I. For the E1 and E2 peak positions and ε1(∞), the fixed values identical to the analysis of Fig. 2 were also used in the analysis. As confirmed from Table I, the parameters obtained show rather complicated variations with R. Although the microstructure of μc-Si:H layers changes largely with R, the μc-Si:H structure also varies with the other deposition conditions. Thus, the μc-Si:H microstructure cannot be defined simply from the R value.

In Table I, on the other hand, we find that the Aσ values increases monotonically with R. This is based on the experimental fact that the amplitude of the E2 peak increases gradually with increasing R, as evidenced from Fig. 1. In this study, in an attempt to characterize the microstructure of various μc-Si:H layers, we defined the μc-Si:H structural factor k from the Aσ values in Table I. In particular, the k values have been determined by normalizing the Aσ values in Table I using Aσ = 16.750 obtained from R = 100; i.e., k = Aσ/16.750. The resulting k values for all the μc-Si:H layers have also been shown in Table I.

Figure 3 shows the TL and HO parameters shown in Table I, plotted as a function of k. In Fig. 3, the solid circles show the experimental values in Table I, and the solid lines indicate the results of the fitting to the experimental data. From the fitting analyses, we obtained the following expressions:

\[ A_{\text{fit}}(\kappa) = 135.0 + 555.5 \exp \left[ -\frac{(\kappa + 0.666)^2}{0.243} \right] \text{eV}, \]  

\[ C_{\text{fit}}(\kappa) = 2.322 - 3.568 \kappa + 7.145 \kappa^2 - 6.382 \kappa^3 + 2.112 \kappa^4 \text{eV}, \]  

\[ E_{\text{fit}}(\kappa) = 1.733 \text{ eV} \quad (\kappa \leq 0.477), \]  

\[ = 2.060 - 0.328 \exp \left[ -\frac{(\kappa - 0.477)}{0.209} \right] \text{ eV} \quad (0.477 < \kappa \leq 1), \]  

\[ E_{0,\text{fit}}(\kappa) = 3.666 + 2.114 \kappa \text{ eV} \quad (0 \leq \kappa < 0.099), \]  

\[ = 3.875 \text{ eV} \quad (\kappa \geq 0.099), \]  

\[ A_{E1}(\kappa) = -0.7 + 347.3 \kappa + 366.4 \kappa^2 - 478.5 \kappa^3 - 30.5 \kappa^4 \text{ eV}, \]  

\[ C_{E1}(\kappa) = 0.474 + 1.026 \exp \left[ -\frac{\kappa}{0.125} \right] \text{ eV}, \]

**TABLE I.** Tauc-Lorentz(TL) and Harmonic oscillator (HO) parameters extracted from the fitting analysis of the μc-Si:H and c-Si dielectric functions using two TL and one HO peaks.

<table>
<thead>
<tr>
<th>R = [H2][SiH4]</th>
<th>Fundamental transition (TL model)*</th>
<th>E1 transition (TL model)*</th>
<th>E2 transition (HO model)*</th>
<th>μc-Si:H structural factor k = Aσ/4Eσ(R = 100)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R = 10</td>
<td>224.9 2.321 1.733 3.666</td>
<td>35.1 0.941 2.338</td>
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</tr>
<tr>
<td>R = 20</td>
<td>184.6 2.038 1.733 3.875</td>
<td>106.9 0.605 2.855</td>
<td>43.22 0.452</td>
<td>0.258</td>
</tr>
<tr>
<td>R = 30</td>
<td>152.6 1.771 1.733 3.875</td>
<td>193.6 0.491 3.041</td>
<td>7.990 0.777</td>
<td>0.477</td>
</tr>
<tr>
<td>R = 50</td>
<td>137.2 1.666 1.733 3.875</td>
<td>251.1 0.481 3.085</td>
<td>11.742 0.645</td>
<td>0.701</td>
</tr>
<tr>
<td>R = 70</td>
<td>135.2 1.642 1.948 3.875</td>
<td>203.9 0.476 3.096</td>
<td>16.750 0.691</td>
<td>1.000</td>
</tr>
<tr>
<td>R = 100</td>
<td>135.1 1.629 2.033 3.875</td>
<td>1277.7 0.261 3.124</td>
<td>25.189 0.361</td>
<td></td>
</tr>
<tr>
<td>c-Si</td>
<td>131.1 1.084 2.334 3.875</td>
<td>1277.7 0.261 3.124</td>
<td>25.189 0.361</td>
<td></td>
</tr>
</tbody>
</table>

*ε1(∞) = 1 (fixed).

Eσ(E1) = 3.360 eV and ε1(∞) = 0 (fixed).

Eσ(E2) = 4.250 eV (fixed).
\[ E_{E1}^{E1}(\kappa) = 3.095 - 1.534 \exp\left( -\frac{\kappa}{0.140} \right) \text{eV}, \quad (9) \]
\[ C_{E2}^{E2}(\kappa) = 0.711 - 0.600 \exp\left( -\frac{\kappa}{0.313} \right) \text{eV}. \quad (10) \]

For the parameters of the \( E_1 \) and \( E_2 \) transitions (\( A_{E1}, C_{E1}, E_{E1}, \) and \( C_{E2} \)), we extrapolated the fitted functions toward \( \kappa = 0 \), as these transitions do not occur when \( \kappa = 0 \). The dotted lines in Fig. 3 show the calculated results obtained from the extrapolation. In Fig. 3, the amplitude and broadening parameters of the fundamental peak (\( A_{\text{run}} \) and \( C_{\text{run}} \)) gradually reduce with increasing \( \kappa \). In contrast, the amplitude parameter of the \( E_1 \) transition (\( A_{E1} \)) gradually increases up to \( \kappa = 0.7 \). At high \( \kappa \), however, the value of \( A_{E1} \) decreases, due to the post-oxidation described in Sec. III A.

If we apply Eqs. (3)–(10), all the model parameters can be calculated for an arbitrary \( \kappa \) value. Figure 4 shows the dielectric functions of various Si:H layers calculated with the variation of \( \kappa \). It can be seen that the dielectric function of a-Si:H changes smoothly to that of \( \mu\)-Si:H as \( \kappa \) increases. This result indicates that the optical database for \( \mu\)-Si:H layers can be prepared successfully from the \( \mu\)-Si:H model using two TL peaks, combined with one HO peak.

Figure 5 shows the schematic microstructures of \( \mu\)-Si:H layers (\( \sim 20 \) nm), depicted for the variation of the \( \kappa \) value. As shown in this figure, the layer structure is completely a-Si:H when \( \kappa = 0 \), and the \( \mu\)-Si:H volume fraction gradually increases up to \( \kappa \sim 0.5 \). By the increase in the \( \mu\)-Si:H component, the light absorption in the visible region (\( 2.0 \leq E \leq 3.2 \) eV) decreases rapidly. It should be noted that the underlying layer is \( \mu\)-Si:H when \( 0.099 \leq \kappa \leq 0.258 \) (\( 20 \leq R \leq 30 \)). From the real-time SE analysis for \( \mu\)-Si:H growth, we confirmed that the direct \( \mu\)-Si:H nucleation on the substrate occurs at \( \kappa = 0.477 \) (\( R = 50 \)). Above this critical value, the \( \mu\)-Si:H layers have high density \( \mu\)-Si:H grains with the a-Si:H phase at the grain boundaries. At \( \kappa \sim 1 \), however, the void-rich structures
are formed at the grain boundaries, and the E\textsubscript{1} transition becomes weaker due to the formation of the n-type µc-Si:H layer by the post-oxidation. In Fig. 5, the µc-Si:H nuclei density is assumed to increase with κ (or R), as reported previously.\textsuperscript{11,16}

C. Application of the µc-Si:H optical database

To confirm the validity of the µc-Si:H optical database established in Sec. III B, we have applied our dielectric function model for the SE analysis of the µc-Si:H layers formed on the ZnO:Al/glass substrates. Figure 6 shows the optical model used for the SE analysis of the µc-Si:H(R = 50)/ZnO:Al/glass multilayer structures.

The optical constants of the glass substrates were modeled by the modified Cauchy model \( (n = A + B/\lambda^2 + C/\lambda^4 - D/\lambda^6) \) with \( A = 1.500 \), \( B = 3162 \text{ nm}^2 \), \( C = 8.371 \times 10^7 \text{ nm}^4 \), and \( D = 4.101 \times 10^{-19} \text{ nm}^{-6} \). In this model, the term \(-D/\lambda^6\) is added to express the rapid reduction in the refractive index below 1.0 eV.\textsuperscript{32} The structure and properties of the ZnO:Al layer were determined in advance from the SE analysis of the ZnO:Al/glass samples. For the SE analysis, we employed an optical model consisting of ambient/surface roughness layer (\( \sim 2 \text{ nm} \))/ZnO:Al (\( \sim 100 \text{ nm} \))/glass substrate. In this analysis, the dielectric function of the ZnO:Al layer was expressed from the combination of the TL and Drude models.\textsuperscript{40,41} The parameter values for the TL model are A = 72.28 eV, \( C = 12.00 \text{ eV} \), \( E_2 = 2.54 \text{ eV} \), \( E_0 = 7.00 \text{ eV} \), and \( \varepsilon_2(\infty) = 1.74 \), whereas the two parameters for the Drude model\textsuperscript{41} are A = 0.34 eV and \( \Gamma = 0.27 \text{ eV} \). In the ZnO:Al layers deposited at room temperature, the carrier concentration is quite low (\( < 10^{20} \text{ cm}^{-3} \)) and the free carrier absorption is rather small. In the optical model of Fig. 6, the interface layer (\( \sim 2 \text{ nm} \)) is provided between the ZnO:Al and µc-Si:H layers. The interface thickness corresponds to the surface roughness layer thickness obtained from the SE analysis of the ZnO:Al/glass structure. The optical properties of the interface layer were modeled as a 50/50 vol.% mixture of the µc-Si:H and ZnO:Al layers using the Bruggeman effective medium approximation (EMA).\textsuperscript{31}

The optical constants of the µc-Si:H layers were calculated from the κ value by applying our µc-Si:H optical database. In the optical model shown in Fig. 6, the structural variation of the µc-Si:H layer toward the growth direction has been neglected, and the single κ value is used to represent the dielectric function of the µc-Si:H bulk layer. It has been reported that, when the µc-Si:H layer thickness increases, the µc-Si:H shows extensive surface roughening.\textsuperscript{3,16} To express these rough surfaces, we have introduced two surface roughness layers having different void volume fractions (\( f_{\text{void1}} \) and \( f_{\text{void2}} \)) into the optical model. In the top layer (1st surface roughness), we also included the SiO\textsubscript{2} component with a volume fraction of \( f_{\text{SiO2}} \) to show the surface oxidation, as reported previously.\textsuperscript{2,22} In the 2nd roughness layer, however, the \( f_{\text{SiO2}} \) is assumed to be zero to reduce the number of the analytical parameters. The volume fractions of the µc-Si:H component in these roughness layers are given by \( f_{\text{µc-Si1}} = 1 - f_{\text{SiO2}} - f_{\text{void1}} \) (1st surface roughness) and \( f_{\text{µc-Si2}} = 1 - f_{\text{void2}} \) (2nd surface roughness), and the optical constants of the roughness layers were obtained from these volume fractions using EMA. In this case, the µc-Si:H dielectric function was calculated simply from the κ value of the µc-Si:H bulk layer. As a result, the total number of the analytical parameters in the optical model of Fig. 6 becomes seven (κ, \( f_{\text{SiO2}} \), \( f_{\text{void1}} \), \( f_{\text{void2}} \), \( d_{\text{1}} \), \( d_{\text{2}} \), and \( d_{\text{b}} \)). When the µc-Si:H layer is quite thin (39 nm), however, the 2nd surface roughness layer was removed. In addition, we observed \( f_{\text{SiO2}} = 0 \) for the thin µc-Si:H layers (\( \leq 163 \text{ nm} \)). In these cases, a fixed value of \( f_{\text{SiO2}} = 0 \) was employed in the analysis.

Figure 7 shows the (\( \psi \), \( \Delta \)) spectra obtained from the SE measurement of the µc-Si:H layer (\( \sim 1900 \text{ nm} \)) formed on the ZnO:Al/glass substrate. In this figure, the two small peaks that appear in the \( \psi \) spectrum show the \( E_1 \) and \( E_2 \) transitions. For the characterization of the µc-Si:H layer, therefore, the SE spectra at the high energy region (\( \sim 4 \text{ eV} \)) are quite important. In this region, however, the number of the data points in the (\( \psi \), \( \Delta \)) spectra becomes smaller, as the SE
spectra have been measured with equal spacing for wavelengths. In this case, if we perform the conventional SE fitting analysis, the low energy region with more data points is fitted predominantly, which in turn degrades the fitting quality at higher energies. To determine $\kappa$ values accurately using the high-energy spectral region, we have employed the following fitting error function in the SE analysis

\[
\chi^2 = \frac{1}{2N-M}\sum_{j=1}^{N} \left\{ \left[ \frac{\psi_{\text{ex}}(E_j) - \psi_{\text{cal}}(E_j)}{\delta\psi(E_j)} \right]^2 + \left[ \frac{\Delta E(E_j) - \Delta E_{\text{cal}}(E_j)}{\delta\Delta E(E_j)} \right]^2 \right\} H^2(E_j),
\]

(11)

where $H(E)$ is a weighting function that shows the separation of data points in photon energies

\[
H(E) = \frac{1239.8}{\lambda} - \frac{1239.8}{\lambda + \Delta \lambda}.
\]

(12a)

\[
= \frac{\Delta \lambda E^2}{1239.8 + \Delta \lambda E^2}.
\]

(12b)

In Eq. (11), $N$ and $M$ show the number of measured ($\psi$, $\Delta$) pairs for photon energies and the number of analytical parameters, respectively. The subscripts “ex” and “cal” in Eq. (11) denote experimental and calculated values at the photon energy of $E_j$, respectively. In addition, we included measurement errors ($\delta \psi$, $\delta \Delta$) in the analysis, as reported previously. In Eq. (12a) and (12b), on the other hand, $\lambda$ shows the wavelength and $\Delta \lambda$ denotes the interval for the measured wavelengths ($\Delta \lambda = 4.77$ nm in our case). By the introduction of the function $H(E)$, the error values for ($\psi$, $\Delta$) reduce at the lower energy region, whereas those increase at high energies. If we employ Eq. (11) in the linear regression analysis, therefore, the SE spectra at the high energy region are fitted predominantly, which in turn allows for the accurate determination of $\kappa$ from the $E_1$ and $E_2$ transitions. It should be noted that the fitting error for $\Delta$ is divided by four in Eq. (11), since the measurement range for $\Delta$ is $0^\circ \leq \Delta \leq 360^\circ$ and is four times larger than that for $\psi$ ($0^\circ \leq \psi \leq 90^\circ$).

The solid lines in Fig. 7 show the fitting result obtained using Eq. (11). It can be seen that the calculated result shows the excellent fitting to the experimental spectra particularly in the high energy region. From the SE analysis, we deduced $\kappa = 0.62 \pm 0.01$ for a $\mu$-Si:H layer thickness of $d_\theta = 1912 \pm 3$ nm. The other parameters extracted from the analysis are $f_{\text{so2}} = 0.22 \pm 0.02$, $f_{\text{ovdd}} = 0.32 \pm 0.02$, $f_{\text{red2}} = 0.142 \pm 0.003$, $d_{\lambda} = 10.5 \pm 0.2$ nm, and $d_{\Delta} = 24.0 \pm 0.4$ nm. If we perform the fitting analysis using $H(E) = 1$ in Eq. (11), the fitting in the interference region ($E < 2.2$ eV) improves, but with relatively large deviation of the calculated spectra at higher energies. The rather poor fitting quality in the interference pattern is mainly caused by the variation of the $\mu$-Si:H properties toward the growth direction, as confirmed below. It should be emphasized that the structural non-uniformity toward the growth direction is negligible at the high energy region, as the penetration depth of light is small in this region due to high absorption coefficients ($\sim 10^7$ cm$^{-1}$) at 3.0 eV.

Figure 8 shows the variation of the Raman spectrum with the $\mu$-Si:H bulk layer thickness ($d_\theta$). A broad peak at $\sim 480$ cm$^{-1}$ and a sharp peak at $\sim 520$ cm$^{-1}$ in the Raman spectra have been assigned to the TO mode of a-Si:H (Ref. 14) and the TO/LO mode of the c-Si component. In the case of the thin $\mu$-Si:H layer (39 nm), the $\mu$-Si:H layer formed initially on the ZnO layer contains a relatively large amorphous component, while the a-Si:H component rapidly decreases with increasing the layer thickness. This confirms the change in the $\mu$-Si:H structure with thickness.

Figure 9 shows $\kappa$ obtained from the SE analysis and the Raman peak intensity ratio, plotted as a function of $d_\theta$. The Raman peak ratio was simply determined from the Raman peak intensities at 480 cm$^{-1}$ and $\sim 520$ cm$^{-1}$ using $I_{480}/(I_{520} + I_{480})$. In this figure, the $\kappa$ value increases with increasing $d_\theta$, whereas the Raman peak intensity ratio also increases with $d_\theta$. Thus, the $\kappa$ value deduced by applying the $\mu$-Si:H optical database shows good correlation with the Raman peak ratio that corresponds to the volume fraction of the crystalline component. The above result demonstrates that the microstructure and the thickness of $\mu$-Si:H layers can be determined simultaneously from the SE analysis.

As described earlier, our dielectric function model assumes that the $\mu$-Si:H optical properties are expressed completely by the $\kappa$ value, which is defined by the $E_2$ peak amplitude. Nevertheless, all the $\mu$-Si:H dielectric functions in this study have been obtained using an identical substrate.
temperature of 190 °C. As known widely,1,11 the growth temperature has large influence on the μc-Si:H microstructures. Thus, our model may not provide reasonable results when the μc-Si:H preparation temperature is quite different from 190 °C. In this study, the substrate temperature was fixed to 190 °C, since the temperature region of 160–200 °C is used commonly for the fabrication of μc-Si:H solar cells.6,10 Our model could show some limitations for other μc-Si:H deposition conditions. Although such limitations have not been found, the application range of our model should be investigated further. It should also be noted that the absorption coefficient z calculated from our model is not reliable when z < 10⁴ cm⁻¹, as the modeling of the low z region has not been performed in this study. To apply our model for the optical simulation of μc-Si:H solar cells, further investigation is necessary.

IV. CONCLUSION

From real-time SE measurements, we have determined the dielectric functions of μc-Si:H layers fabricated by PECVD using different hydrogen dilution ratio R = [H₂]/[SiH₄] at 190 °C. The μc-Si:H dielectric function shows systematic variation with R, and the E₂ direct-transition peak at 4.3 eV increases monotonously with R, whereas the E₁ transition at 3.4 eV shows a rather complicated variation for R. To parameterize the μc-Si:H dielectric function, a μc-Si:H model that employs two Tauc-Lorentz models and one Harmonic oscillator model has been developed. To characterize μc-Si:H microstructures, we introduced the μc-Si:H structural factor k defined by the amplitude of the E₂ transition. From our model, the optical database for the μc-Si:H layers has been constructed, and the variation of the dielectric function with the a-Si:H/μc-Si:H phase structure has been expressed successfully using only nine variables. The developed model has been applied further to determine the structural properties of μc-Si:H layers deposited on ZnO/glass substrates. For the SE analysis of this structure, an optical model with two surface roughness layers has been developed to model the extensive surface roughening in the μc-Si:H layers. The SE analysis revealed that the crystalline component increases with increasing μc-Si:H layer thickness. Moreover, the microstructure of the μc-Si:H layers obtained from Raman spectroscopy shows a direct relationship with that deduced from the SE analysis. As a result, we have developed a SE characterization method that allows the characterization of the μc-Si:H microstructure and thickness simultaneously.

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39Equation (1) can be derived by replacing A in Refs. 31 and 38 with AT and using C = 2π.
44L. P. Herman, Optical Diagnostics for Thin Film Processing (Academic, San Diego, 1996).