SURFACE ROUGHNESS AND OPTICAL SCATTERING IN MICROCRYSTALLINE SILICON

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ABSTRACT: The size of the features that form the surface of microcrystalline silicon increases with increasing film thickness. By application of atomic force microscopy we detail the increase in root-mean-square surface roughness with increasing thickness for a number of microcrystalline silicon samples deposited by hot-wire chemical vapour deposition. For these, specular reflection and transmission show increased optical scattering with increasing thickness. To test if the surface roughness can be determined by optical means for the variety in the roughness values, we apply optical scattering theory to calculate the rootmean-square surface roughness from specular reflection data. We find good agreement with the atomic-force derived values. We also find an enhanced optical absorption coefficient for the thicker samples which may be beneficial for photovoltaic applications.

Keywords: Optical Properties - 1: Microcrystalline Si - 2: Si Films - 3

1. INTRODUCTION

Optical characterisation and especially a variety of reflectance-measurement techniques as a non-contact method are widely used [1]: It ranges from classifying of metal surfaces [2] to obtaining structural information on plants [3]. These techniques can also be applied to semiconductors that play a role in photovoltaics to study the surface topography or to determine optical scattering that can be advantageous for photovoltaic applications as it increases optical absorption and photocarrier generation. Microcrystalline silicon (µc-Si) that is also called nanocrystalline silicon (nc-Si) because of its grains in the nm-range, is a thin-film semiconductor which will benefit from optical scattering when applied for example in the bottom cell of tandem solar cells in combination with hydrogenated amorphous silicon top cells. As the µc-Si films show an increase in the size of the surface structure with increasing thickness [4] there is some correlation between increased absorption, optical scattering that may be surface scattering and also bulk scattering, and surface roughness [5].

We apply measurements of optical reflection and transmission to correlate the increase in the optical scattering with the increasing thickness of a series of µc-Si samples deposited by hot-wire chemical vapour deposition (HWCVD). All the parameters were fixed for the deposition of the samples for which the different thicknesses were achieved by a variation in the deposition time. With atomic force microscope (AFM) measurements we studied the increase in the size of the structures that form the surface as the thickness increases and calculated the RMS surface roughness R_q . We apply optical scattering theory to determine the value for the RMS surface roughness from specular reflection, labelled R_s . Comparing the values from the different techniques we discuss the applicability of reflectance measurements on µc-Si for surface roughness determination.

2. EXPERIMENTAL

Hot-wire µc-Si samples were deposited at Institut für Physikalische Elektronik, Universität Stuttgart, on Corning



Figure 1: Raman spectra show the same shape for all thicknesses and no significant change in the amorphous contribution to the crystalline volume fraction.

7059 glass in an ultra-high-vacuum (UHV) deposition system for which Ref. 6 gives more details. Standard parameters in the study here are tungsten as filament material with 250 μ m in diameter and a filament current of 11 A, resulting in a filament temperature of about 1700°C. We show results for a thickness series for a substrate temperature of 200°C where the hydrogen gas flow rate was fixed at 60 sccm and the silane gas flow rate [SiH₄] was set to 4 sccm. Different thicknesses of 0.54, 1, 2.5, 4.3, 7.2 μ m were achieved by the different deposition times of 15, 30, 60, 117, 180 min, respectively. In addition we compared the properties of a sample with [SiH₄] = 2 sccm.

Standard constant photocurrent method (CPM) was applied to determine the optical absorption properties [7]. Specular transmission T and reflection R were measured with a double-beam photospectrometer. For total integrated transmission T_t and reflection R_t scattered and specular radiation were collected with an integrating sphere.

The surface topography of the samples was investigared by atomic force microscopy in contact mode (Nanoscope III, Digital Instruments). For each sample, several square areas of $25 \,\mu\text{m} \times 25 \,\mu\text{m}$ were explored, for which the RMS roughness R_q was calculated, leading to the mean values we are presenting. It is worth noting that a particular extension of the Nanoscope called "Resiscope" has been developed at LGEP in order to simultaneously collect topographical and local electrical information [8,9]. Results derived from this technique will be soon reported elsewhere.

3. THEORY

We apply optical scattering theory [10] to determine the value for the RMS surface roughness, labelled R_s , from the expression $R_s = \lambda/4\pi \times [\ln(R_{c-Si}/R)]^{1/2}$ where R is the specular reflection of the sample in the ultraviolet spectral range and R_{c-Si} the reflection of crystalline silicon. It is noted that for simplicity and to test its applicability we use the c-Si reflection rather than that of an ideally reflecting flat sample surface required by the theory.

4. RESULTS

4.1 Structural properties

Raman spectra, obtained by excitation with green light, of Fig. 1 show the typical peak for μ c-Si with no variation in its position for the samples of the thickness series. Further inspection of the curves also indicates that crystallinity of the samples does not change much as there is no significant difference in the amorphous and crystalline contributions to the signal.

4.2 Optical properties

Figure 2 shows that *R* and *T* for samples of the thickness series change to a large degree with increasing thickness. In Fig. 2 (top) the thinner samples exhibit the reflection peaks characteristic of monocrystalline Si in the UV range. These peaks disappear with increasing thickness although there is no hint from Raman that the crystalline fraction close to the surface is reduced. Optical scattering increases which may also be noticed by inspection of the μ c-Si samples with the eye.

For increasing thickness the edge in specular transmission T in Fig. 2 (bottom) would shift to longer wavelengths even if no scattering was present because of thicknessdependent larger absorption. Scattering also plays a role as the interference fringes disappear for the thicker samples.

A 4-micron thick sample, deposited with the higher hydrogen dilution (labelled lines) shows only very low values for both R and T and exhibits a large amount of optical scattering.

Figure 3 displays the absorption coefficient as determined from CPM, shown as raw data, not corrected to lower values for optical scattering effects [5]. There is a clear increase in the sub-gap data with increasing thickness.

4.3 Atomic force microscopy

Figure 4 shows the height variations taken from horizontal scans of three μ c-Si samples with 15, 60 and 180 min. deposition time. There is a large increase in the height variations and thus the surface roughness increases with



Figure 2: Specular reflection R (top) decreases with increasing thickness as optical scattering increases. The edge in specular transmission T (bottom) shifts to longer wavelengths and interference fringes are reduced for thicker samples. The sample, deposited with higher hydrogen dilution, is shown in labelled lines with low values in R and T.

thickness. The R_q values in Fig. 5 clearly show this increase with increasing thickness. The surface roughness of the higher-dilution sample with about 100 nm is clearly larger than the other data for the thickness series.

Figure 6 shows topographical images of three μ c-Si samples with 15, 60 and 180 min. deposition time. Starting with the thinnest sample and noting that the grey scale varies from the nm to μ m range one can see the evolution in three samples to larger-seized features with increasing thickness/deposition time.

4.4 Surface-roughness comparison

Figure 7 compares the RMS surface roughness R_s from optics, evaluated at a wavelength of 330 nm, with the AFM-derived R_q . The data follow the diagonal which represents data when R_s equals R_q .

5. DISCUSSION

Specular reflection and transmission from Fig. 2 change to a large degree as the thickness of the samples increases. It is the optical scattering that reduces the R in the UV and the typical signature peaks or shoulders as a sign for crystallinity disappear, to be found, however, in the integrated reflection, measured with an integrating sphere. With it, one can also identify optical scattering in the T data of Fig.2.



Figure 3: Calibrated CPM-derived absorption coefficient for three samples of the series, showing the increase in sub-gap absorption with increasing thickness. The absorption coefficient for c-Si is shown for comparison.



Figure 4: Height variations increase with increasing deposition time. The corresponding thicknesses are $0.54, 2.5, 7.2 \mu m$.



Figure 5: The AFM surface roughness increases by almost one order in magnitude as the thickness increases for the series. The single point indicates that at higher hydrogen dilution R_q becomes larger, with a value above 100 nm.



Figure 6: Showing the evolution of the surface topography for three with 0.54 (top), 2.5 (middle) and 7.2 (bottom) μ m thick samples, the AFM scans illustrate the development of larger sized features in the thicker samples. Note that the height scale attached to the plots is different in each case.



Figure 7: Good agreement in the comparison of the RMS surface-roughness (R_q) from AFM (abscissa) and from optical measurements (ordinate). The ordinate data are evaluated at 330 nm.

The effect of scattering upon the sample deposited with higher dilution of silane in hydrogen (with 2 sccm silane in 60 sccm) is much larger. Specular reflection remains below a few percent indicating more pronounced scattering.

Relating the decrease in R and increase in optical scattering to the surface properties we see from Figs. 4 and 6 larger-sized features on the surface for the thicker samples. It may be noted that the needle-like structures with widths in the several-hundred-nm range in Fig. 4 consist of many small crystallites. Fig. 5 summarises the findings in terms of an increase in RMS surface roughness of the films. The sample with high hydrogen dilution stands out in Fig. 5 for the value for R_q as it does with its scattering properties. The simple and not surprising qualitative statement from here is that increase in surface roughness increases optical scattering in the UV.

Quantitatively the specular reflection data can be exploited by calculating the surface roughness. We show in Fig. 7 the range for which we found very good agreement with the values deduced from the AFM.

For larger surface roughness on the one hand, as R_q approaches the wavelength of the radiation, the validity of the theory vanishes. Computed values for R_s become smaller than R_q . For smaller R_q on the other hand, the error introduced by applying the c-Si reflection data increases. We find that R_{c-Si} is larger than that for example of a polished surface of our samples. Thus, R_s values that are too large are returned from the formula in that case.

Nevertheless in the large range shown in Fig. 7 the error that one makes when applying R_{c-Si} is minor and surface roughness determination by optical means, as also applied in Ref. 5, may provide an alternative if an AFM is not available.

Increasing the sample thickness increases the absorption coefficient as measured by CPM. Having discussed the increase in optical scattering in the UV range here, one may tentatively conclude that it is the surface scattering that also influences the sub-gap absorption, too. Results on smooth surfaces after polishing of rough samples, however, also show high values in the sub-gap absorption [11,5] so that in this respect also bulk scattering, leading to enhanced optical absorption, cannot be ruled out.

6. CONCLUSION

We monitored the increase in RMS surface roughness with the thickness in a series of μ c-Si samples, finding good agreement between the roughness values calculated from specular reflection data and those from AFM. Comparing with samples from different deposition conditions, as was shown here with a large value for R_q for a highly diluted μ c-Si sample, we conclude that the value of the surface roughness can be adjusted by appropriate deposition parameters. We reported enhanced optical sub-gap absorption that increases with increasing thickness of the films. Surface scattering contributes to the enhanced absorption but is not necessarily its only source.

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REFERENCES

- [1] e.g., J. C. Stover, Optical Scattering: measurement and analysis (McGraw-Hill, New York, 1990).
- [2] e.g., Volumes of J. Modern Optics, Applied Spectroscopy, Applied Optics, Proceedings SPIE.
- [3] J. Qui, W. Gao, B.M. Lesht, Int. J. Remote Sensing 19 (1998) 641.
- [4] R. Brüggemann, W. Bronner, A. Hierzenberger, M. B. Schubert, I. Zrinscak, in J.M. Marshall et al. (Eds.), Thin Film Materials and Devices - Developments in Science and Technology, (World Scientific, Singapore, 1999), p.1.
- [5] A. Poruba, Z. Remes, J. Springer, M. Vanecek, A. Fejfar, J. Kocka, J. Meier, P. Torres, A. Shah, in Proc. 15th European Photovoltaic Solar Energy Conf., Vienna, 1998, edited by J. Schmid et al. (Europ. Commission, JRC, Ispra, 1998), p. 781.
- [6] H.N. Wanka, M.B. Schubert, A. Hierzenberger, and V. Baumung, in Proceedings 14th European Photovoltaic Solar Energy Conference, Barcelona, 1997, edited by H.A. Ossenbrink et al. (Stephens & Associates, Bedford, U.K., 1997), p. 1005.
- [7] M. Vanecek, J. Kocka, J. Stuchlik, A. Triska, Solid State Commun. 39 (1981) 1199.
- [8] F. Houzé, R. Meyer, O. Schneegans, L. Boyer, Appl. Phys. Lett. 69 (1996) 1975.
- [9] O. Schneegans, P. Chrétien, E Caristan, F. Houzé, A. Dégardin, A Kreisler, in Superconducting and Related Oxides: Physics and Nanoengineering III, edited by D. Pavuna and I. Bozovic, Proc. of SPIE Vol. 3481, 1998.
- [10] P. Beckmann, A. Spizzichino, The Scattering of Electromagnetic Waves from Rough Surfaces (Pergamon Press, Oxford, 1963).
- [11] R. Brüggemann, A. Hierzenberger, P. Reinig, M. Rojahn, M. Schubert, S. Schweizer, H. Wanka, I. Zrinscak, J. Non-Cryst. Solids 227-230 (1998) 982.