OPTICAL PROPERTIES OF PECVD a-SI:H LAYER ON GLASS SUBSTRATE

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In the present paper thin a-Si:H films deposited onto glass substrates are characterized using linear (transmission) and nonlinear (second harmonic generation (SHG)) optical techniques. The refractive index at both fundamental and SHG wavelength are estimated. Complicated dependence of SHG on film thickness and deposition temperature is found. The origin of the nonlinear properties is discussed.

Keywords: Hydrogenated Silicon, Second Harmonic Generation.

1. INTRODUCTION

Amorphous silicon has substantially different properties as compared to crystalline silicon. It has become recognized as an important material in its own right, with many interesting applications. Especially in its hydrogenated version a-Si:H, is a material of a considerable technological value for photovoltaic and solar sells applications, liquid-crystall displays and as optical waveguides [1-3]. In the field of the disordered materials the a-Si is often taken as the prototype of a covalent glass. For that reasons deposition and characterization of thin a-Si:H is interesting and important task.

The nonlinear optical techniques are increasingly becoming an important investigation tool of materials because of their sensitivity to symmetry [4-6]. Being non-destructive and contactless, the optical second harmonic generation (SHG) has been applied to study surfaces and interfaces of materials with bulk inversion symmetry. Since in this case no significant response can be gained from the bulk, the main contribution to the second order nonlinearity comes from the surface and interface regions where this symmetry is broken.

In the present paper results are provided on of thin a-Si:H films deposited onto glass substrates through characterization of the linear (transmission) and nonlinear (second harmonic generation (SHG)) optical properties.

2. PROBLEM STATEMENT

The interest to the a-SI:H because of its practical importance and from fundamental point of view has provoked numerous investigations on this material. It has been known for some years that the electrical characteristics of the a-Si:H thin films depend from its structural properties [7]. The latter have often been related to deposition parameters and hydrogen content. The structure of the a-Si:H has been studied theoretically by modeling of the position and connections of the Si-tetrahedra in the amorphous network. Some suggestions have been made from indirect experimental results. In recent years the nonlinear optical techniques are increasingly becoming an important investigation tool of materials because of their sensitivity to symmetry. Recently, observation of second-harmonic generation in a-Si:H thin film has been reported [8]. A simple model has been suggested to explain the source of SHG response being the strained region near the substrate. However, a more detailed insight taking into account the linear properties of the films and their relation to the observed nonlinear effect of the SHG is necessary, which is the purpose of the present study.

3. EXPERIMENTAL CONDITIONS

The a-Si:H films were obtained by plasma-enhanced chemical vapor deposition (PECVD) at 13.56 MHz and 48 mW/cm^2 in a capacitively-coupled planar chemical reactor from hydrogen diluted SiH_4. The substrates were Corning glass 7059 plates. By varying the deposition time, films with thickness between 100 and 600 nm have been obtained. The deposition was carried out at substrate temperatures from 150 to 270^{o}C.

The linear optical transmission spectra have been measured using Specord-M40 spectrophotometer.

The SHG measurements have been performed using YAG:Nd^{3+} laser (λ=1.064 μm, pulse duration of 30 ns and pulse energy of 1 mJ) as a pumping source. The incident fundamental beam was directed onto the a-Si:H film. The SHG signal was detected in transmission by a photomultiplier and fed into a computer-controlled data acquisition system. The probe beam was polarized either parallel (*p*-polarization) or normal (*s*-polarization) to the plane of incidence. The SH intensity was measured as a function of the angle of incidence θ by rotating the sample around an axis lying in the film plane.

The pump energy of 1.17 eV (1.064 μm) is substantially below the absorbtion edge of the amorphous Si:H of 1.8 eV [9]. Nonetheless, the laser beam on the film surface was defocused to a spot with a diameter of about 3 mm to decrease the intensity in order to avoid structural changes.

Modeling of the SHG characteristics of the films has been made by considering the absorbtion of the films at the SHG frequency and the interference in the films.

4. EXPERIMENTAL RESULTS AND DISCUSSION

The measurements of the transmission spectra in the near IR around the basic wavelength λ_1=1064 nm SH frequency λ_2=532 nm have revealed interference patterns. In Fig. 1 the transmission data for around λ_1 are presented. Since the absorption is very weak in this region and the interference fringes are clearly seen, the value of the refractive index n_1 can be obtained. It was estimated to be n_1=3. Values between 2.5 and 3.6 obtained from IR absorbance and spectral ellipsometry measurements for films deposited in the same temperature interval and similar experimental configurations can be found in the literature [7, 10, 11]. Similarly, in the visible range around SH frequency λ_2=532 nm an interference effect combined with strong absorption has been observed. The magnitude of the refractive index at λ_2 has been estimated using respective fitting procedure and the data are given in Table 1. The values are consistent with those found from spectral ellipsometry data for the dielectric function of films deposited at similar conditions [7].

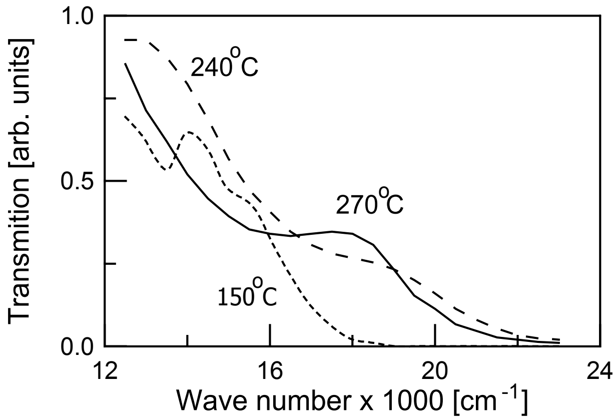

Fig.1 Transmission spectra of a-Si:H film of 230 nm deposited at different substrate temperatures.

Typical experimental angular dependence of the SH intensity is shown in Fig. 2. The observed variation of the SH signal with θ is indicative of ∞m symmetry group of the films deposited on a substrate [8]. This is relevant to the amorphous state of the Si-H films.

In Fig. 3 the SH signals vs. film thickness for different deposition temperatures of the a-Si:H films are plotted. Complicated thickness dependence of the nonlinear response is obvious. The curves exhibit maxima in the SHG intensity, positioned at a different thickness values dependent on the deposition temperature. For 150°C and 240°C the maximum values of the SH intensity are observed at about 300 nm and 220 nm, respectively. It can be assumed that the maximum of the SH signal for 300°C is located below the measured thickness range and corresponds to a film thickness of about 110 nm.

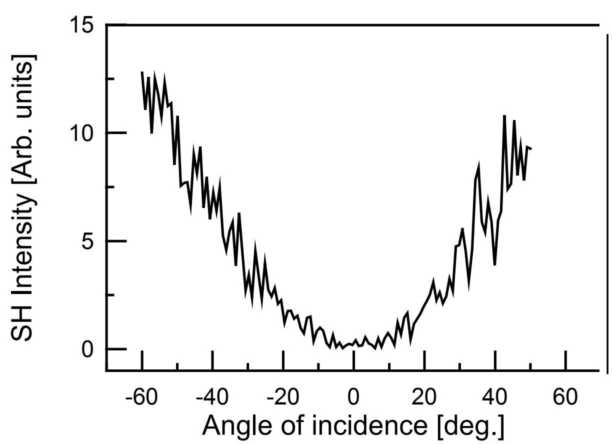

Fig.2 Experimental angular dependence of SHG signal for a-Si:H film of 110 nm thickness.

In our previous study of thin (~100 nm) films [8] it was suggested that the possible origin of the observed SHG is a strained transitional layer between the film and the substrate. The reason was that no significant SHG response is expected in materials with bulk inversion symmetry, where the SHG is forbidden in electric-dipole approximation and the SH signal comes from the surface dipole and bulk quadruple contributions [12]. This is the case with crystalline and amorphous Si.

Therefore, the nonlinear behavior the a-Si:H film should be due to the breaking of the inversion symmetry at the interfaces of the film.

Analysis on the basis of a simple thermal stress model can account for the tendency of enhanced SHG at higher deposition temperature because of the higher total film stress. However, the dependencies in Fig. 3 with the tendency of decrease of the SH signal with thickness, as well as the observed maxima, indicate the need of a more detailed insight considering the contribution of the two film interfaces to the substrate and the air. The interference effects seen in Fig.1 in the films have also to be taken into account. Moreover, the films in ref.[8] with thicknesses around 100 nm are weakly absorbing at the SH frequency λ_2. Increasing of the film thickness up to 600 nm is accompanied by increased absorption in the films. Therefore the absorption in the films has also to be considered.

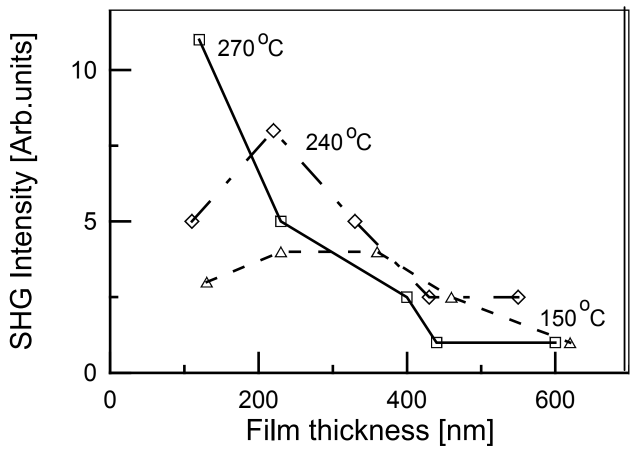

Fig.3 SH signal measured at angle of incidence of 60° as a function of film thickness and deposition temperature.

By the following discussion on the mechanism of the SHG the whole film will be considered as a complex three-layer structure, including the interface film/substrate, the bulk of the film and the interface film/air. Further an attempt will be made to discriminate between these three contributions to the nonlinear process. The conventional point of view is that the bulk of the film, as a centrosymmetrical region, has no contribution to SHG in electric-dipole approximation. At the interfacial regions the inversion symmetry is broken giving rise to the observed SH signal.

The thickness of these regions is smaller than the whole film thickness [13]. To explain the presence of a maximum in the curves in Fig. 3, two models have been considered. *In the first model* the source of SHG was assumed to be only the a-Si:H/glass interface and the interference effect were due to usual linear reflection at the boundaries of the a-Si:H film. *In the second model* an additional SHG source has been included, located at the film/air interface. The assumptions behind the modelling include: substantially smaller nonlinearity at the outer interface in comparison with the nonlinearity at the interface film/substrate; both linear reflection and interference between these nonlinear sources give comparable contributions to SHG signal. At this stage the absorption was not taken into consideration. For that reason the theoretical results are valid only for the relatively transparent thin films with thickness less than 300 nm.

The SHG patterns obtained for the first and the second models have demonstrated very similar behaviour. The variation of the theoretical SH signal taken at angle of incidence of 60° with film thickness is plotted in Fig. 4. It is seen that the SHG signal follows a periodic variation with increasing film thickness. The oscillations appear with a repetition rate of about 60 nm, which is in disagreement with the experimental pattern, observed in Fig. 3. The experimental points, however, follow in a 50 nm step range. Obviously, in order to envisage experimentally the interference phenomenon, a set of samples would be needed at a distance much smaller than 60 nm. This is not to realize experimentally, since it is difficult to rely on determination of the film thickness with an accuracy better than about 20 nm. The reason is the surface roughness which, as found from the AFM image shown [14], is of this order of magnitude. It is obvious that at this stage no discrimination between the two proposed models for the location of the nonlinear source in the films can be made. Additional theoretical simulation of the SH response taking into account the increasing absorbtion with film thickness are supposed to locate the nonlinear source. Most probably the contribution of the bulk of the films has to be considered. In this case the spatial period of the interference is defined by coherence length [15]:

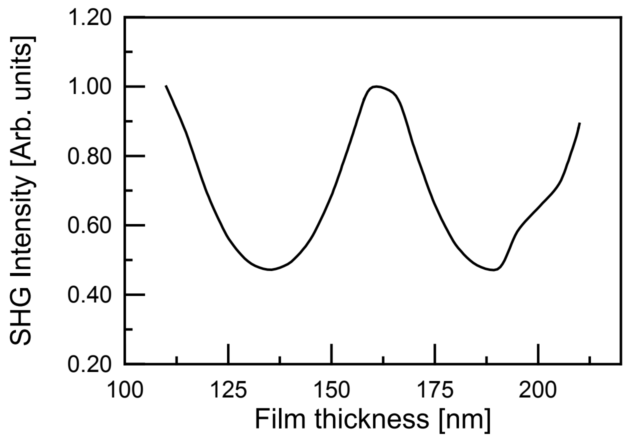

Fig.4 Dependence of the theoretical SH signal on the film thickness, calculated assuming the nonlinear source at a-Si:H/glass interface.

$$l_c = 0.25\lambda_\omega / (n_2 - 2n_1)$$

where λ_ω is laser pump wavelength.

This effect is negligible in transparent thin films because of the small dispersion of the optical index (e.g., l_c of the α-quartz is about 20 microns). However, in strongly absorbing films the dispersion increases and the coherence length can decrease to some hundred nanometers.

A possible indication may be the bulk nature of the stress-induced lowest-order optical nonlinearity [6] under inhomogeneous deformation of deposited a-Si:H films in a direction along the surface normal, so that the symmetry would be violated in this direction. The resulting symmetry of the strained volume of the film is the same as that of the surface, i.e. ∞m m (see Fig. 2).

5. CONCLUSIONS

The refractive index at both fundamental and SHG wavelength are estimated. The thickness dependence of the SHG is discussed modeling the a-Si:H films on the glass substrate as a three layer system. It is shown that the contributions of the two interfaces of the film with the substrate and the outer surface with air are not sufficient to explain the experimental results. The necessity to estimate the contribution of the bulk effects has been underlined. The effect of strained volume of the film due inhomogeneous deformation of deposited a-Si:H films in a direction along the surface normal has been suggested.

6. REFERENCES

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