

# RAMAN SPECTRA AND ATOMIC FORCE MICROSCOPY OF a-Si:H AND a-SiC:H THIN FILMS

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## Abstract

Morphology and structure of hydrogenated amorphous silicon (a-Si:H) and hydrogenated amorphous silicon carbide (a-SiC:H) films were investigated to reveal an influence of the carbon content on the films properties in nanoscale. Thin a-Si:H and a-SiC:H films of different compositions were deposited by plasma enhanced chemical vapor deposition (PECVD) from SiH<sub>4</sub> and SiH<sub>4</sub> + CH<sub>4</sub> gas mixtures. Raman spectroscopy and atomic force microscopy (AFM) were used to analyze the chemical bonds and morphology of the films. It was demonstrated that the carbon introduction in an amorphous network resulted not only in an increase of the optical bandgap of a-Si<sub>1-x</sub>C<sub>x</sub>:H films from  $E_g=1.72$  eV at  $x=0$  to  $E_g=2.18$  eV at  $x=0.2$ , but also it led to a decrease of the effective sizes of nanoroughness on the film surfaces. The Raman scattering spectra verified that the amorphous network of a-SiC:H films even with low carbon content became less ordered on short and intermediate nanoscales.

**Keywords:** amorphous hydrogenated silicon, hydrogenated amorphous silicon carbide, optical gap, Raman spectra, atomic force microscopy.

## 1. Introduction

Amorphous hydrogenated silicon (a-Si:H) and its alloys, including amorphous hydrogenated silicon carbide (a-SiC:H), are widely used in various electronic devices, such as thin-film solar cells [1], detectors of  $\beta$  radiation [2] etc. Physical properties a-Si:H and a-SiC:H strongly depend both on the formation conditions (technological conditions of deposition as pressure and purity of initial gases, temperature of a substrate, charge power, design features) and on some other factors. Therefore the quality control of the properties of amorphous films and their reproduced preparations is an actual problem. In order to prepare disordered or nanostructured material the thermodynamic nonequilibrium process is usually required. According to the basic principles of a self-organization theory (synergetics) a consequence of the nonequilibrium processes can result in formation of a material with nonuniform structure, which influences both the physical properties of the material and devices on its basis. The main aim of our work is to study a-Si:H and a-SiC:H films with different carbon content influenced the morphology and nanostructural properties of the investigated disordered material.

## 2. Experimental

Films of a-Si:H and a-SiC:H were deposited on quartz substrate by using a triode magnetron type reactor under DC decomposition of mixtures of 20% SiH<sub>4</sub> in He and 10 % SiH<sub>4</sub>, 40 % CH<sub>4</sub> in He, respectively. The employed system allows us to reduce an influence of the electronic bombardment on the film properties and to provide a high degree of the gas ionization and then the high deposition velocity under operation at lower pressure. The substrate temperature was maintained nearly constant at 280<sup>0</sup>C and the pressure in the deposition chamber was 2 mTorr. The charge current was about 20 mA. The thickness of films was about 1 μm.

The morphology of films was studied by using a NT-MDT Atomic Force Microscope (AFM). The thickness, refractive index and absorption coefficient of the films were deduced from the optical transmittance spectra measured by using a Lambda-35 spectrometer [3]. The Raman scattering experiments were performed in the range of 100—1200 cm<sup>-1</sup> at room temperature with excitation radiation at 477 nm by using a NT-MDT Integra spectrometer, the incident laser power was about 25 mW.

## 3. Experimental Results and Discussions

The optical bandgap,  $E_g$ , and refractive index,  $n_f$ , accounted  $E_g=1.72$  eV,  $n_f=3.4$  for a-Si:H and:  $E_g=2.18$  eV,  $n_f=2.62$  for a-SiC:H, respectively. The carbon incorporation in a-SiC:H films is known to be accompanied by the optical bandgap increase and refractive index reduction. Characteristic features of amorphous tetrahedral semiconductors with a rigid structural grid (a-Si:H and a-SiC:H) are structural heterogeneities, which influence the electronic properties of material. The morphology of amorphous films is characterized by different type heterogeneities (columns, globules, grains etc.).

Figure 1 shows AFM images of the investigated a-Si:H and a-SiC:H films.

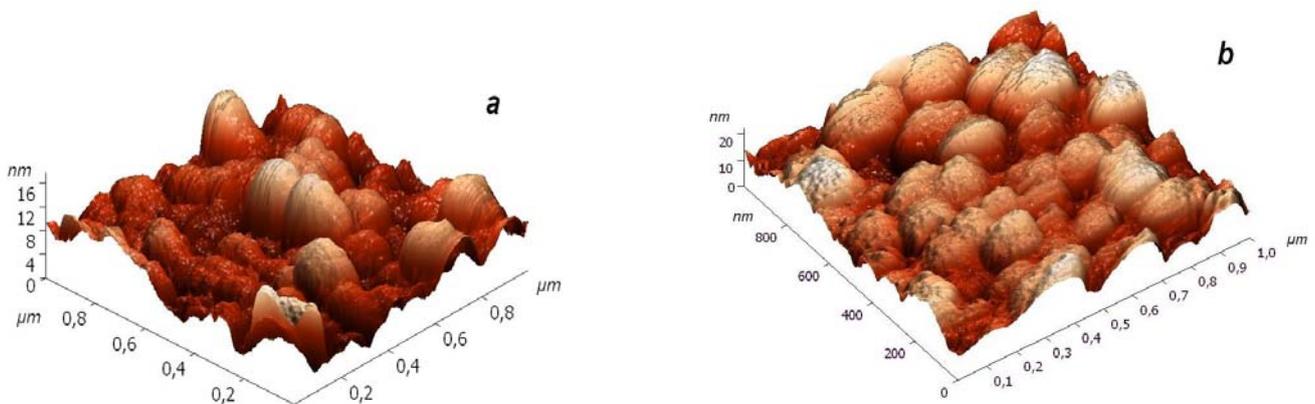
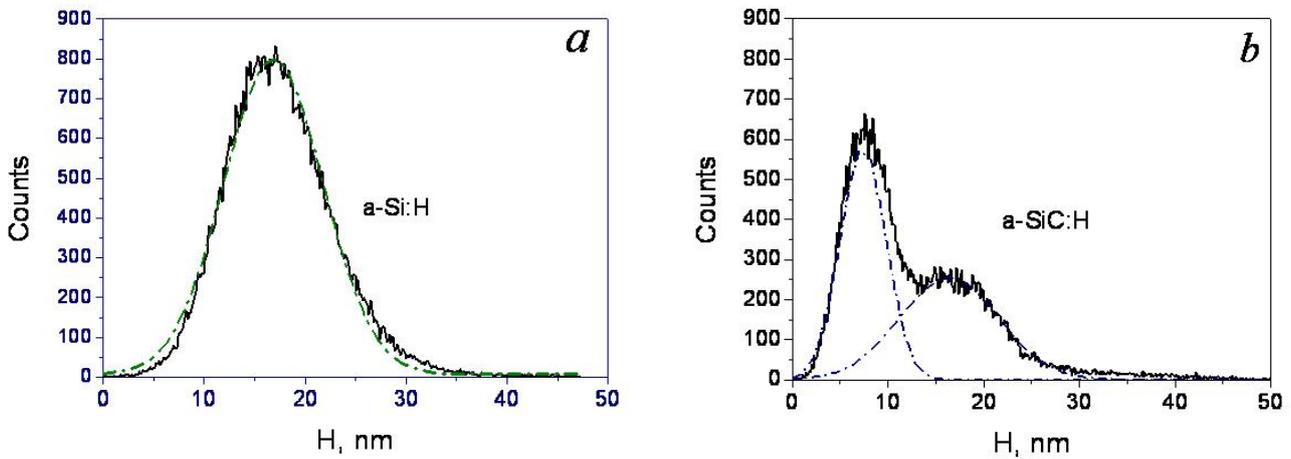


Figure 1. AFM images of a-Si:H (a) and a-SiC:H (b) films surface.

The energy-dispersive analysis of the a-Si<sub>1-x</sub>C<sub>x</sub>:H films showed the carbon content x=0.20. This relatively small concentration of carbon leads to the essential morphology change of the films surface. Taking into consideration the observed islet-like structure of a-SiC:H films, it is possible to assume, that the islet surfaces are formed because of clustering of carbon atoms in the form of C-H<sub>n</sub> bonds.

Figure 2 shows height distribution functions for the investigated films. The function were calculated by considering N=65536 points for surface area of 10x10 μm<sup>2</sup>. The distribution function for a-Si:H is well approximated by a Gaussian function centered at H<sub>1</sub>=16.85±0.03 nm with halfwidth Δ<sub>1</sub>=9.9±0.1 nm. The distribution function for a-SiC:H was fitted (deconvoluted) by two Gaussians with characteristic parameters H<sub>2</sub>=16.44±0.01 nm, Δ<sub>2</sub>=10.2±0.2 nm and H<sub>3</sub>=7.44±0.02 nm, Δ<sub>3</sub>=4.79±0.04 nm. Thereby, it is observed a bimodal structures of the surface morphology in the a-SiC:H films.



**Figure 2. Height distribution function of a-Si:H (a) and a-SiC:H (b) films. Dashed lines are results of a deconvolution with Gaussian functions.**

The sample surfaces consist of many islets (grains). The lateral size and height of islets are seen to be differ essentially for a-Si:H and a-SiC:H films. The a-SiC:H films are characterized by larger scatter of the sizes and height of islets in comparison with that for the a-Si:H films. This fact can be explained by the difference in chemical composition of the films.

The average heights H<sub>1</sub> and H<sub>2</sub>, and halfwidth Δ<sub>1</sub> and Δ<sub>2</sub> practically coincide. Hence it is possible to conclude that the amorphous silicon phase does not change its morphology despite the carbon addition (an effect of “the structure memory”). One can assume that the second distribution peak with parameters H<sub>3</sub> and Δ<sub>3</sub> for the a-SiC:H film is related with the occurrence of carbon and is probably connected with carbon cluster formation.

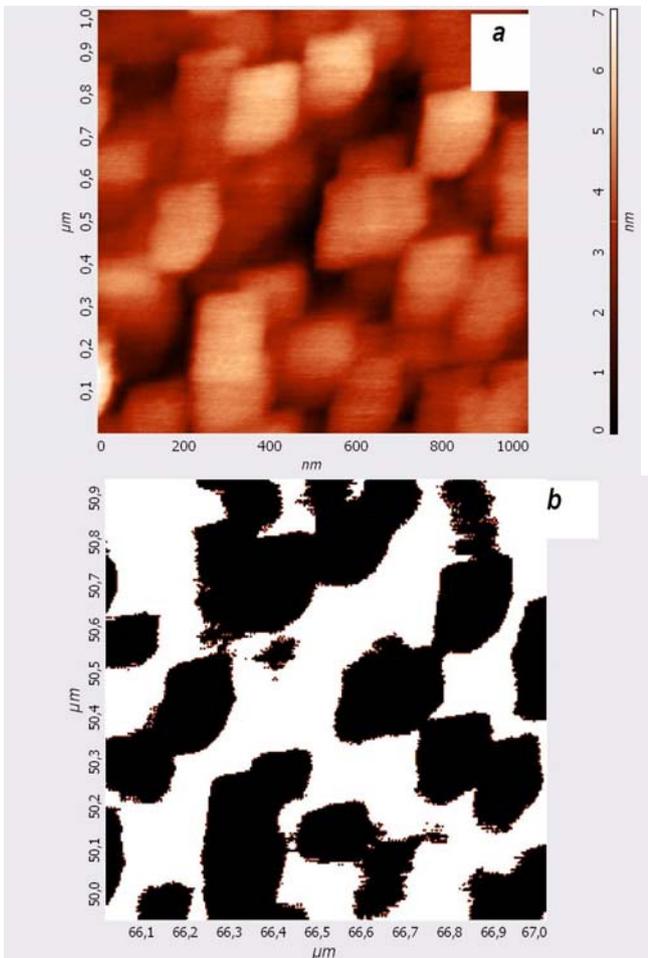
For the mathematical characterization of the surface roughness we used the root-mean-square roughness S<sub>q</sub>, defined as:

$$S_q = \sqrt{\frac{\sum_{i=1}^N (H_i - \hat{H})^2}{N}}, \quad (1)$$

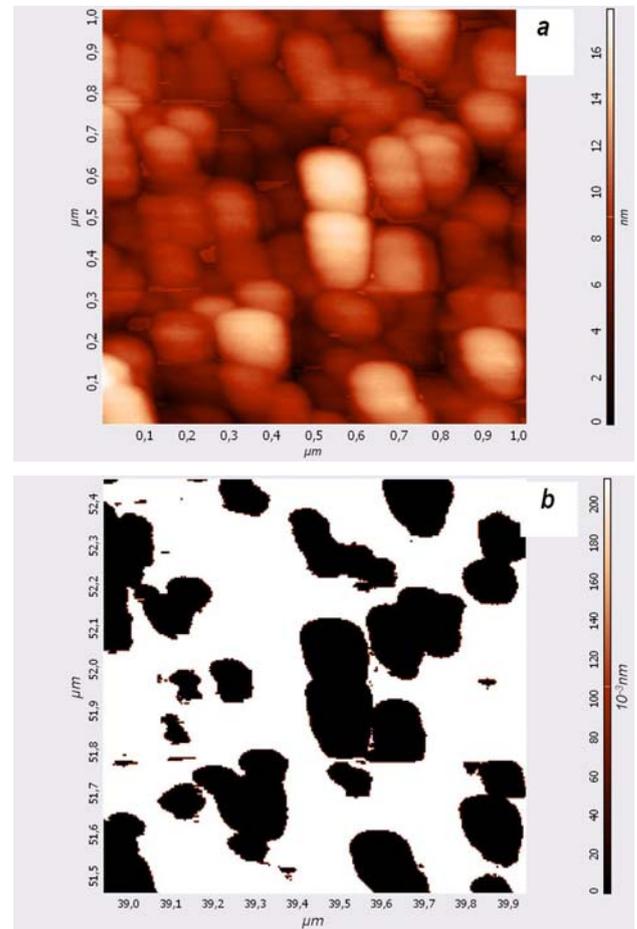
where  $N$  is the number of measured points,  $\hat{H}$  is the average height (zero level),  $(H_i - \hat{H})$  is the relative height of  $i$  points.

On the one hand,  $S_q$  is found to depend on the area of investigated surface, namely with area reduction the average square-law roughness decreased. On the other hand, independently on the scanned area the value of  $S_q$  for a-SiC:H exceed that for a-Si:H. In particular, for the scanning area of  $1 \times 1 \mu\text{m}^2$  one can get  $S_q=3.5$  nm for a-Si:H and  $S_q=3.91$  nm for a-SiC:H.

We used a segmentation of the AFM images as a threshold processing method to allocate borders and to measure precisely the lateral sizes of islets. Figure 3 and Figure 4 show the AFM images of a-Si:H and a-SiC:H films before and after the threshold processing. It is seen that the carbon introduction in a-Si<sub>1-x</sub>C<sub>x</sub>:H leads to a reduction of the average diameter of islets from  $\bar{D}=168$  nm for  $x=0$  to  $\bar{D}=64$  nm for  $x=0.2$ , and the maximum distance  $L_{\text{max}}$  between islets decreases from 259 nm to 98 nm.



**Figure 3.** AFM-image of a-Si:H (a) and a section of this image after processing by threshold method (b).



**Figure 4.** AFM-images of a-SiC:H (a) and a section of this image after processing by threshold method (b).

Fig. 5 shows the Raman spectra of a-Si:H and a-SiC:H films. The characteristic parameters deduced from these spectra are listed in Table 1.

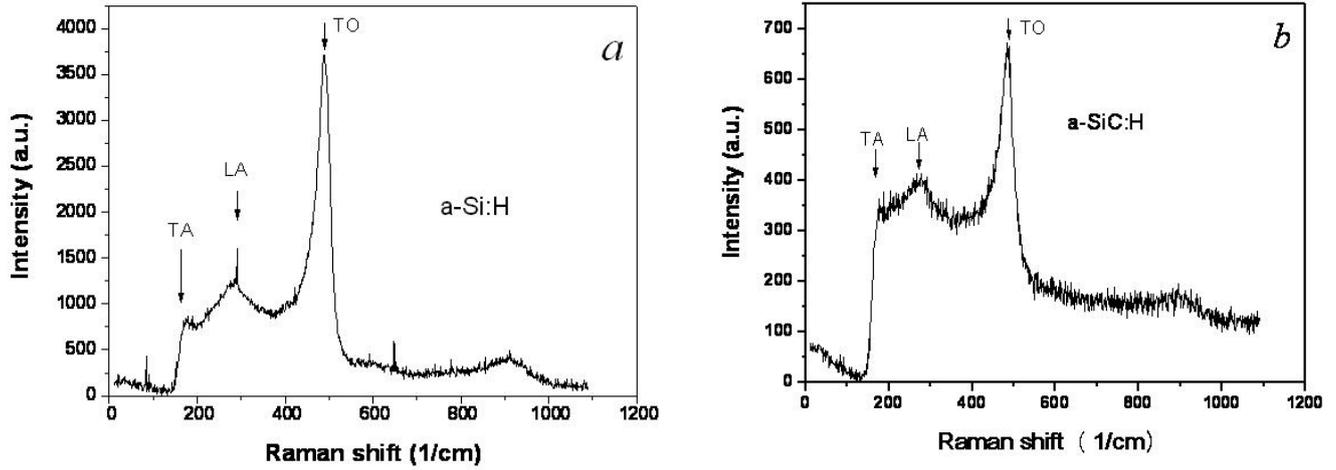


Fig.5. Raman spectra of a-Si:H (a) and a-SiC:H (b) films.

Table 1. Parameters of a-Si:H and a-SiC:H films determined from the optical transmission measurements (band gap  $E_g$ ) and Raman scattering spectra (line position  $\omega_{TO}$  and halfwidth  $\Gamma_{TO}$ , and ratios between acoustical and optical line intensities  $I_{TA}/I_{TO}$  and  $I_{LA}/I_{TO}$ , respectively).

Sample	$E_g$ , eV	$\omega_{TO}$ , (cm <sup>-1</sup> )	$\Gamma_{TO}$ , (cm <sup>-1</sup> )	$I_{TA}/I_{TO}$	$I_{LA}/I_{TO}$
a-Si:H	1.72	482	47	0.257	0.358
a-SiC:H	2.18	475	69	0.533	0.617

The absence of peak at 520 cm<sup>-1</sup> in the Raman spectra confirms that a-Si:H and a-SiC:H are completely amorphous. The peak at about 480 cm<sup>-1</sup> related to the Si-Si transverse-optical-like (TO) vibrations is sensitive to the short-range disorder of a-Si:H films. An increase in the halfwidth of TO band ( $\Gamma_{TO}$ ) and a shift of TO position ( $\omega_{TO}$ ) towards lower frequencies indicate an increase in the short-range disorder [4]. The peak at about 165 cm<sup>-1</sup> related to the Si-Si transverse-acoustical-like (TA) vibrations is indicative for the intermediate-range disorder of the films.

A decrease in the ratio of the intensity of the TA band to that of the TO band,  $I_{TA}/I_{TO}$ , manifests an increase in the intermediate-range order [5]. Since, the coordination defects (clusters) are necessary for the appearance of the longitudinal acoustical-like (LA) mode at 300 cm<sup>-1</sup> the ratio  $I_{LA}/I_{TO}$  also characterizes the average structural ordering [5].

The introduction of carbon ( $x=0.20$ ) results not only in essential increase in the optical band from 1.72 to 2.18 eV, but also it leads to an increase of the structural disorder. The increase in halfwidth  $\Gamma_{TO}$  and shift of the maximum from 482 to 475 cm<sup>-1</sup> give evidences for an increase in the structural defect density in a short-range order. The increases of ratios of  $I_{TA}/I_{TO}$  and  $I_{LA}/I_{TO}$  confirm that the amorphous network of a-SiC:H films becomes less order on short and intermediate scales in comparison with a-Si:H films.

## 4. Conclusion

Films of a-Si:H and a-SiC:H were prepared and characterized to determine the role of carbon content in the film for their surface morphology and structure. It was found that the carbon introduction in an amorphous silicon network resulted in the essential change of the morphology of film surface. The average diameter of islets (grain) in a-Si<sub>1-x</sub>C<sub>x</sub>:H films decreased from 168 nm for x =0 to 64 nm for x =0.2 and the maximum distance between the grains decreases from 259 nm to 98 nm. The height distribution function for the films was fitted by two Gaussian curves, one of which is characteristic for a-Si:H, another one is related with the carbon clusters formation. The Raman scattering spectra have verified that the amorphous network of a-SiC:H films even at low carbon content became less ordered on short and intermediate nanoscales.

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