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Effect of gas temperature on the structural and optoelectronic properties of a-Si:H thin films deposited by PECVD

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ABSTRACT

The effect of gas temperature (T_g) in the process of plasma-enhanced chemical vapor deposition (PECVD) on the structural and optoelectronic properties of the grown a-Si:H thin film has been examined using multiple characterization techniques. Gas temperature was confirmed to be an important parameter for the optimization of fabrication process and the improvement of structural and optoelectronic performances of the thin films. The structural properties of the thin films were examined using atomic force microscopy (AFM), Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy, and electronic-spin resonance (ESR). Furthermore, the spectroscopic ellipsometry (SE), the optical transmission measurement in ultraviolet–visible region and the electrical measurement were used to investigate the optical and electrical properties of the thin films. It was found that the changes in T_g can modify the surface roughness, the amorphous network order, the hydrogen bonding modes and the density of the thin films, and eventually improve the optical and electrical properties.

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) thin film prepared by plasma-enhanced chemical vapor deposition (PECVD) is a technologically important material with a wide range of practical applications. It is used in the production of solar cells [1,2], infrared detectors in night vision systems [3], and thin film transistors in flat panel display devices [4]. All these applications are based on its good electrical and optical properties as well as compatibility with semiconductor technology. However, the properties of a-Si:H thin film fabricated by PECVD are sensitive to deposition conditions, such as substrate temperature, power density, gas flow rate and process pressure. Many efforts to prepare the high-quality a-Si:H thin films with lower defect density and higher structural stability have been made. It is well known that the substrate temperature can strongly affect the diffusion of the radicals on the growing surface and hence cause these radicals more readily to locate the optimum growth sites. As a result, the substrate temperature has been one of the most studied deposition parameters. As far as the temperature parameters during PECVD process are concerned, apart from the substrate temperature, the gas temperature (T_g) fed in the PECVD reaction chamber before glow-discharge is a novel process parameter for tailoring the properties of a-Si:H thin films. In fact, the variation of T_g in PECVD system can influence the energy of plasma during glow-discharge and eventually modify the properties of the thin films [5]. According to Martins et al. [6], when the thin films are fabricated close to or within the particle formation regime in the a-Si:H thin films, the dependence of thin film properties on T_g is more remarkable than on substrate temperature. However, most of the studies so far have focused only on the impact of substrate temperature. In our previous study, we reported the effects of gas temperature on the structural evolution of phosphorus-doped a-Si:H thin films by Raman spectroscopy [7]. The results reveal that there exists a gradual ordering of the amorphous network, both in the near surface and interior region with the increase of T_g, leading to a better quality of a-Si:H thin films. But the further investigation in the effect of T_g on the structural and optoelectronic properties of a-Si:H thin films has not been done yet, which is exactly the aim of this paper.

In this study, the a-Si:H thin films are deposited by PECVD at various gas temperatures. The properties of thin films are studied using multiple characterization techniques, including atomic force microscopy (AFM), Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy, electronic-spin resonance (ESR), spectroscopic ellipsometry (SE), optical transmission measurement in ultraviolet–visible region and electrical measurement.

2. Experimental details

2.1. Sample preparation

The a-Si:H thin films were prepared by PECVD method using pure silane. Our PECVD system was equipped with a parallel-plate electrode as shown in Fig. 1. The area of both electrodes and the separation were 220 cm^2 and 2 cm, respectively. A frequency of 13.56 MHz and a power

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density of 100 mW/cm^2 were applied to the upper electrode and the lower electrode was grounded. The PECVD system was held at a given temperature for 4 h to achieve a balance in the reaction chamber. All substrates were cleaned strictly according to the following standard procedure before the deposition in order to ensure a good adhesion between the substrate and the thin film. After manual cleaning in detergent solution, the substrates were immediately subjected to ultrasonic cleaning in alcohol, acetone and deionized water for 10 min in sequence. The substrates were fixed on a sample holder which was above the plasma zone to facilitate the avoidance of undesirable larger particle contamination. The heat source of substrates came from the upper heating units (tungsten filaments) during the deposition. The heat source of gas came from the heating belts and the lower heating units (tungsten filaments). The lower heating units served as heat preservation. It is worth mentioning that the gas temperature in our case is nominal, because it is not measured directly. The total gas flow rate of SiH_4 was 40 sccm (standard cubic centimeter per minute) in all deposition. The deposition pressure was 60 Pa for all samples fabricated. The substrate temperature was fixed at 250 °C.

2.2. Characterization method

To analyze bonding formation in the a-Si:H thin films, Bruker Tensor 27 Fourier transform infrared spectroscopy (FTIR) apparatus was used at room temperature, and the moisture in the environment was controlled to a low level (relative humidity<70%). The Raman spectroscopy, carried out in a backscattering geometry using a JY-HR800 spectrometer, was used to study the changes of the amorphous silicon network order on the short and medium range scales. The incident laser power was set below 0.3 mW and the beam was defocused over a circle with a diameter of 2 μm to reduce the heating effect due to the laser irradiation and hence to improve the measurement accuracy. The ESR measurement was carried out at room temperature on a BRUKER ESP4105 spectrometer equipped with double cavities, which was operated with microwave power of 20 mW. The AFM (SPA400) images were acquired in the tapping mode using a silicon cantilever. In addition, the mass densities of thin films are measured using floatation method [8].

In the optical analysis, we have considered the dielectric function $\varepsilon(E)$ of a-Si:H thin films using SE measurement. Since SE is an indirect technique, the $\varepsilon(E)$ spectra of samples were calculated through a data inversion procedure in SE850 spectroscopic ellipsometer using the point pair $(\psi,\triangle)$, where ψ denotes the amplitude ratio of the reflectance coefficients of the polarized light parallel and perpendicular to the incidence plane, and $\triangle$ is the phase shift between the two components. In order to calculate the $\varepsilon(E)$ of a-Si:H thin films, a proper optical model should be built. In our case, optical model consists of ambient/surface roughness layer/bulk layer/substrate (K9 glass). The surface roughness layer was modeled as a mixture of 50% a-Si:H (or a-Si) bulk layer and 50% voids using the Bruggeman effective medium approximation (EMA). During the measurement, the best fits were achieved by minimizing the mean square error function. Optical transmission measurement in ultraviolet–visible region was performed using a Shimadzu UV-1700 UV–Vis spectrophotometer.

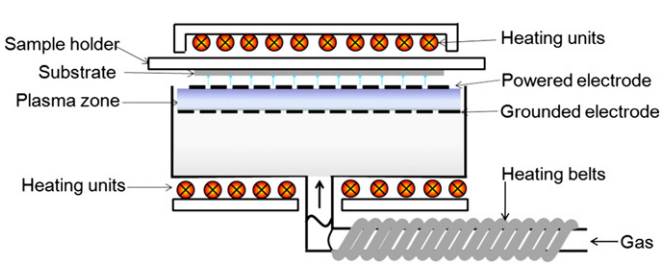

Fig. 1. Schematic configuration of the PECVD system.

The dark conductivity σ(T) of the thin film was measured using Keithley4200 semiconductor characterization system in the V/I configuration with an input impedance greater than $10^5\Omega$. The temperature dependence of dark conductivity was measured using an ESPEC ESL-02KA high-low temperature test chamber with temperature stability less than ±0.1 °C.

3. Results and discussion

3.1. Structural properties of a-Si:H thin films

Through AFM observations the information about the morphological properties of a-Si:H thin films can be extracted. In our case, the scan is performed on a 1 μm×1 μm area with the tapping mode. In Fig. 2, we present the three-dimensional AFM images of the samples deposited at T_g of room temperature (RT), 80 °C, 130 °C and 160 °C, respectively. There is an apparent trend by looking at the topographies in Fig. 2. When T_g is set at RT, the lateral size of lump is much greater than that of the other three samples. The AFM image of T_g=RT has the largest surface roughness with root mean square (rms) roughness of 3.63 nm. Sequentially, the rms value decreases from 1.84 nm to 1.50 nm as T_g increases from 80 °C to 160 °C. The growth of thin films should undergo two different stages, the island growth and the coalescence. According to Drévillon and co-workers, by the time that the coalescence occurs the surface roughness is essentially unchanged as the thin film continues to grow [9]. As a result, the difference in surface roughness is ascribed to the modification of nucleation sites due to different T_g.

ESR is one of the few experiments which give structural information about defects [10]. Fig. 3 gives the first derivative of the normalized ESR absorption spectrum of a-Si:H thin films deposited at different T_g. The electronic spin density (N_s) and the g factors are shown in the inset. It can be seen that the g values of all a-Si:H thin films are near 2.0055. Both theoretical calculation and experimental evidence reveal that the g value of 2.0055 originates from the dangling bond in amorphous network [11], instead of the fivefold-coordinated silicon atom that also is referred as the floating-bond. As shown in Fig. 3, N_s decreases from 1.1×10^{17} cm^{-3} to 3.4×10^{16} cm^{-3} as T_g increases from RT to 160 °C. It shows that the increase of T_g can reduce the dangling bonds in a-Si:H thin films to a large extent.

Raman scattering has been extensively used to estimate the evolution of network structure due to its intensity sensitive to the structural disorder in solids. As shown in the inset of Fig. 4, the Raman spectrum of a-Si:H thin film consists of several vibrational modes. The band at about 150 cm^{-1}, associated with transverse-acoustic (TA) vibrational modes, is proportional to the density of dihedral angle fluctuations, reflecting the medium-range order (MRO) of amorphous network [12]. The TO phonon band at about ω_{TO}=480 cm^{-1} is sensitive to the root-mean-square bond-angle variation $\Delta\theta$ [13]. A shift of TO band position toward higher frequencies and a decrease in the peak width (Γ_{TO}) are consistent with the increase in the short-range order (SRO). Moreover, several computational studies show that the relation between Γ_{TO} and the SRO can be quantified. On the basis on the continuous random network model, Beeman et al. have obtained a linear relation $\Gamma_{TO}=15+6\Delta\theta$, which is often used by experimentalists to determine $\Delta\theta$ from Raman measurements [14]. The presence of the longitudinal acoustic band (LA) at 300 cm^{-1} and the presence of the longitudinal optical band (LO) at 410 cm^{-1} are related to the coordination defects in thin films, and the more defects the larger values of I_{LA}/I_{TO} and I_{LO}/I_{TO} ratio [15], where I stands for the integral intensity of the corresponding vibrational mode. The peaks at about ω_{2LA}=610 cm^{-1} and ω_{2TO}=960 cm^{-1} are overtones of the main a-Si ω_{LA} and the main a-Si ω_{TO}, respectively [16]. In our case, the Raman data between 600 cm^{-1} and 1000 cm^{-1} are not analyzed due to the overlap of overtones.

The Raman spectra of a-Si:H thin films deposited at different gas temperatures are shown in Fig. 4. It can be seen that all the thin