



Enhancing optical properties of nanocrystalline silicon films with air exposure

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ABSTRACT

We report the effect of air exposure and deposition temperatures, T_d , on the optical property of nanocrystalline silicon (nc-Si). The nc-Si thin films were investigated by photoluminescence (PL), optical absorption, X-ray diffraction (XRD), Fourier-transform infrared (FTIR) absorption and Raman scattering. Experimental results show the structural change from an amorphous to a nanocrystalline phase at $T_d = 80^\circ\text{C}$. In addition, it suggests that T_d low condition leads to the increase in the density of SiH-related bonds and a decrease in the average grain size, $\langle \delta \rangle$. The oxygen absorption peak increases with the air-exposure time. The PL exhibited two peaks at around 1.75–1.78 and 2.1–2.3 eV. The PL increases and blue shifts consistently with the decrease of $\langle \delta \rangle$ and increase of oxygen content. The first peak may be related to nanocrystallites in nc-Si films and the origin of another one may be due to defect-related oxygen. Thus, by the plasma-enhanced chemical vapor deposition (PECVD) technique at low T_d , we can produce the nc-Si films with different grain sizes, causing the corresponding luminescent properties. The new method processes advantages of low deposition temperature and effective oxidation of nc-Si on inexpensive substrates, thus making it more suitable for developing low-cost array or flexible nc-Si optoelectronic devices.

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1. Introduction

Nowadays, nanocrystalline silicon (nc-Si) with grains in nanometer size has attracted more attention in optoelectronic and microelectronic devices for its superior properties [1–4]. Moreover, great efforts have been devoted to photoluminescence (PL) of Si-based materials for developing integrated optoelectronics with the standard Si VLSI technology [1]. For example, embedding the nanometer-sized silicon within an insulating host will enhance the quantum confinement effect, which spreads the band gap of Si for PL emission [5]. During the last few years, various methods were proposed to embed nanometer-sized silicon, such as implantation of Si into SiO₂ [5], Si/SiO₂ super-lattice structure [6,7], thermal-

oxidized nanocrystalline Si [8], etc. At the same time, these techniques suffer from complicated and high-temperature process, thus making it unsuitable for developing low-cost array or flexible optoelectronic nc-Si devices.

nc-Si has been synthesized by several techniques such as microwave or laser-induced decomposition of silane (SiH₄)-like precursors [9,10], pulsed-laser deposition (PLD) of Si [11], low-pressure chemical vapor deposition (LPCVD) [12], electrochemical etching of Si wafers [1,13], ion implantation of Si⁺ [14], cosputtering of Si and Si dioxide (SiO₂) [15], and plasma-enhanced chemical vapor deposition (PECVD) [16].

On the other hand, for the visible luminescence properties of nc-Si, control of the size distribution and surface condition of nc-Si with reproducibility is critical to sensitive light-emitting properties. The PL is highly dependent on the discrete size of Si nanocrystals and also changes with different surface passivation. For the

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decomposition, PLD, and LPCVD methods, both surface passivation and deposition of nc-Si thin films without agglomeration need further investigations. For the electrochemical etching method, preparation condition dependence and degradation of the PL are major concerns. For the ion implantation method, multiple implantation at different energies is required to create a thick layer of nc-Si. Compared with other fabrication methods, PECVD has been extensively utilized in the industry and is compatible with ultra-large-scale integration technology. nc-Si thin films formed by PECVD have shown strong and stable PL, robust structure, and good surface passivation. The characteristics of nc-Si films deposited by PECVD can be finely tuned through Si concentration in the films as well as through post-deposition annealing and oxidation.

Study of the influence of the different deposition parameters on the growth of the material is therefore important both for newer device applications and also for understanding the basic physics of the growth process of thin films. Several deposition parameters, such as plasma energy and density, substrate temperature, rf power, gas flow rate, deposition pressure and dilution of the source gas (silane) with other gases (argon, hydrogen, or helium) will strongly influence the structure and properties of the grown nc-Si thin films. The effect of Ar dilution on the structure of hydrogenated amorphous and microcrystalline silicon films deposited by rf glow-discharge decomposition of silane has been demonstrated [17–19]. A detailed experimental study has been reported on the effect of the dilution of silane with hydrogen on optical properties of a-Si:H prepared by plasma deposition as function of the gas–volume ratio and the substrate temperature [20]. However, most features of the a-Si:H network structure are defined at the time of growth and therefore the optical and electric properties depend on the details of the deposition process. In the present work, we report the growth and characterization of nc-Si thin films deposited by the PECVD technique. The large numbers of atomic hydrogen are necessary for passivation of dangling bonds and reconstruction of Si–Si bonds to improve film quality. Also, it has been known that the deposition of nc-Si is due to the selective etching activity of hydrogen atoms towards the amorphous phase with respect to the crystalline structure. Thus, the role of hydrogen is to promote the nucleation and the crystallization of a-Si:H at low temperature with desired grain size [21]. The use of SiF₄ has also been successfully employed to obtain more orderly materials since fluorine atoms, produced in SiF₄ plasma decomposition, are effective etchant species [22]. These facts were considered in choosing the feed gases. In the study of Lim et al. [23], the deposition temperature (T_d) was decreased until 220 °C. Thus, the grain size decreased until 20 nm. In the present contribution, the T_d was further decreased up to 60 °C with high H₂ dilution to further decrease the grain size. Moreover, a low processing temperature can enlarge the application field of nc-Si films in wider industrial production and solar cells by using inexpensive substrate. Therefore, lowering the processing temperature is becoming a challenging task for the semiconductor research community. In addition, the use of air exposure instead of

high-temperature oxidation will reduce the cost of substrates. The aim of this work is to get more insight into the effect of T_d and air exposure on the optical and structural properties of nc-Si films, and also the possibility to enhance the optical properties of nc-Si films. To our knowledge, the effect of air exposure on the optical and structural properties of nc-Si films has not been studied before.

2. Experimental method

The nc-Si films were deposited by radio frequency (rf) glow-discharge (at 13.56 MHz) decomposition of SiH₄/SiF₄ (+He)/H₂ mixtures in a hot-wall-type fused quartz reactor, 50 mm in diameter, employing the inductive coupling of rf power, which were inserted into an electric furnace. The substrates were loaded horizontally on a quartz boat with its surface parallel to the axis of the reactor. The remarkable feature of this deposition system is that the samples are exposed to the plasma (the growing surface is bombarded with ions). When PECVD nc-Si films were deposited using this deposition system, it has been reported that the resultant nc-Si films have the following two essential effects as the rf power is increased: an enhancement in the degree of preferential orientation of grains in the films and an improvement in the flatness on the film surface [24]. Such results should be caused by an effect of ion bombardments on the film surface during growth [24], and such an effect may also lower T_d for preparing high-quality nc-Si films as well as an effect due to fluorine chemistry. The deposition pressure was adjusted by throttling the cross-section in the inlet of the pump, and was measured using an absolute pressure meter. The details of the PECVD system used have been described elsewhere [25]. Just prior to the deposition of nc-Si films, the substrates were sequentially cleaned by rinsing them for 30 min in acetone and then in ethyl alcohol using an ultrasonic syringe. The samples were more cleaned by exposing them to N₂ and then to H₂ plasma at 90 W for 20 min. Then nc-Si films were deposited at the different T_d with the dynamic pressure of 0.3 Torr for every deposition. The samples were deposited on 0.3-mm-thick glass (coring 7059) substrates with an area of 10 × 20 mm² for measurements of X-ray diffraction (XRD), Raman scattering and atomic force microscopy (AFM), on 0.3-mm-thick fused quartz substrates with an area of 10 × 20 mm² for measurements of PL and optical absorption, and on 0.3-mm-thick (100) Si (N-type, high resistivity 1000–3000 Ω cm) substrates with an area of 10 × 20 mm² for measurements of Fourier-transform infrared (FTIR) spectroscopy. The rf power supply of 20 W was used. The gas flow rates were [SiH₄] = 0.6 sccm, [SiF₄] = 0.38 sccm, which was diluted with 95% helium (He: 95%, SiF₄: 5%), and [H₂] = 20 sccm. The T_d was varied from 60 to 300 °C.

The structural properties of the nc-Si films were investigated by means of XRD (SHIMADZU XD-D1) employing a diffractometer with the slit width of 0.1 mm, set at the front of the detector. The relative intensity (integrated area) of the XRD spectra from

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