

# The influence of local Si—C bonding density on the photoluminescence of Si-QDs upon thermal annealing the hydrogenated amorphous Si-rich silicon carbide thin films



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## ABSTRACT

Non-stoichiometric hydrogenated amorphous silicon carbide thin films ( $\alpha$ -SiC:H) were deposited by plasma-enhanced chemical vapor deposition. The samples were subsequently post-annealed at 750, 900, 1050, and 1200 °C, respectively. Photoluminescence (PL) was measured by fluorescence spectrometer at room temperature. Infrared absorption was carried out by Fourier transform infrared absorption. Chemical compositions were analyzed by X-ray photoelectron spectroscopy. The synthesis of silicon quantum dots (Si-QDs) was characterized by Raman scattering spectroscopy and directly by high-resolution transmission electron microscope. PL measurements revealed that there were complicatedly shifted sub-bands upon the thermal annealing temperature increase. The behaviors of these shifted sub-bands showed converse trends as that of the local Si—C bonding densities. A possible influence for the PL by the evolution of the local Si—C bonding densities and the synthesis of Si-QDs in the  $\alpha$ -SiC:H samples is supposed.

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## 1. Introduction

Ever since the enhanced visible PL was observed in porous silicon at room-temperature in 1990, great attention has been paid to nanocrystalline silicon materials with the objective to obtain potential silicon-based light emitting devices [1–3]. Nanocrystalline silicon particles embedded in amorphous silicon-oxide ( $\text{SiO}_x$ ), silicon-nitride ( $\text{SiN}_x$ ), silicon oxynitrides ( $\text{SiO}_x\text{N}_y$ ), and silicon-carbide ( $\text{SiC}_x$ ) thin films have been widely studied [4–8]. These materials have the common characteristics that nanocrystalline silicon particles play an essential role in PL and the amorphous matrix provides the means for three-dimensional quantum confinement of carriers [9,10]. The reported experimental results showed that PL of these nanocrystalline systems is influenced not only by the silicon nanostructure itself, but also by the optical-geometry and passivation states provided by the matrix [11–13]. Hao et al. had investigated the origin of PL from Si-QDs in amorphous  $\text{SiN}_x$ :H, which had been annealed in oxygen ambient. They identified that the PL was originated from the localized exciton radiative recombination via the surface Si—N or Si—O—Si bonds states [3]. Kalem et al. had synthesized silicon nanoparticles and found the PL was green-orange. They attributed the high energy band to silicon particles embedded in the polycrystalline, and the broad band to the interfacial porous oxide layer [14].

In this paper, non-stoichiometric  $\alpha$ -SiC:H thin films were deposited by plasma-enhanced chemical vapor deposition. For different measurements, the samples were subsequently post-annealed at 750, 900, 1050, and 1200 °C, respectively. The dependence of room-temperature PL on the evolution of local Si—C chemical bonding density and the synthesis of Si-QDs upon thermal annealing are investigated.

## 2. Experimental details

Non-stoichiometric  $\alpha$ -SiC:H thin samples were prepared on p-type crystalline Si (100) wafers and quartz ( $\text{SiO}_2$ ) plates simultaneously by plasma-enhanced chemical vapor deposition. The background pressure in the discharge chamber was pumped down to about  $5 \times 10^{-5}$  Pa with a mechanical pump and a turbo molecular pump. The substrate temperature, the working pressure, and the power density were maintained at 200 °C, 0.8 Torr, and 160  $\text{mW} \cdot \text{cm}^{-2}$ , respectively. The flow rates of  $\text{H}_2$ -diluted 10% silane ( $\text{SiH}_4$ ) and pure methane ( $\text{CH}_4$ ) were maintained at 50 and 10 sccm, respectively. The deposition time was 60 min. After deposition, the samples were cut into several smaller parts. The smaller parts were annealed at 750, 900, 1050, and 1200 °C, respectively, in a flowing environment of nitrogen (purity 99.999%) for 10 min and then were cooled down naturally to room temperature.

PL measurements were carried out by fluorescence spectrometer (FP-6500) in range of 300–800 nm by reflection configuration at room temperature, where the incident angle was fixed and nearly along the

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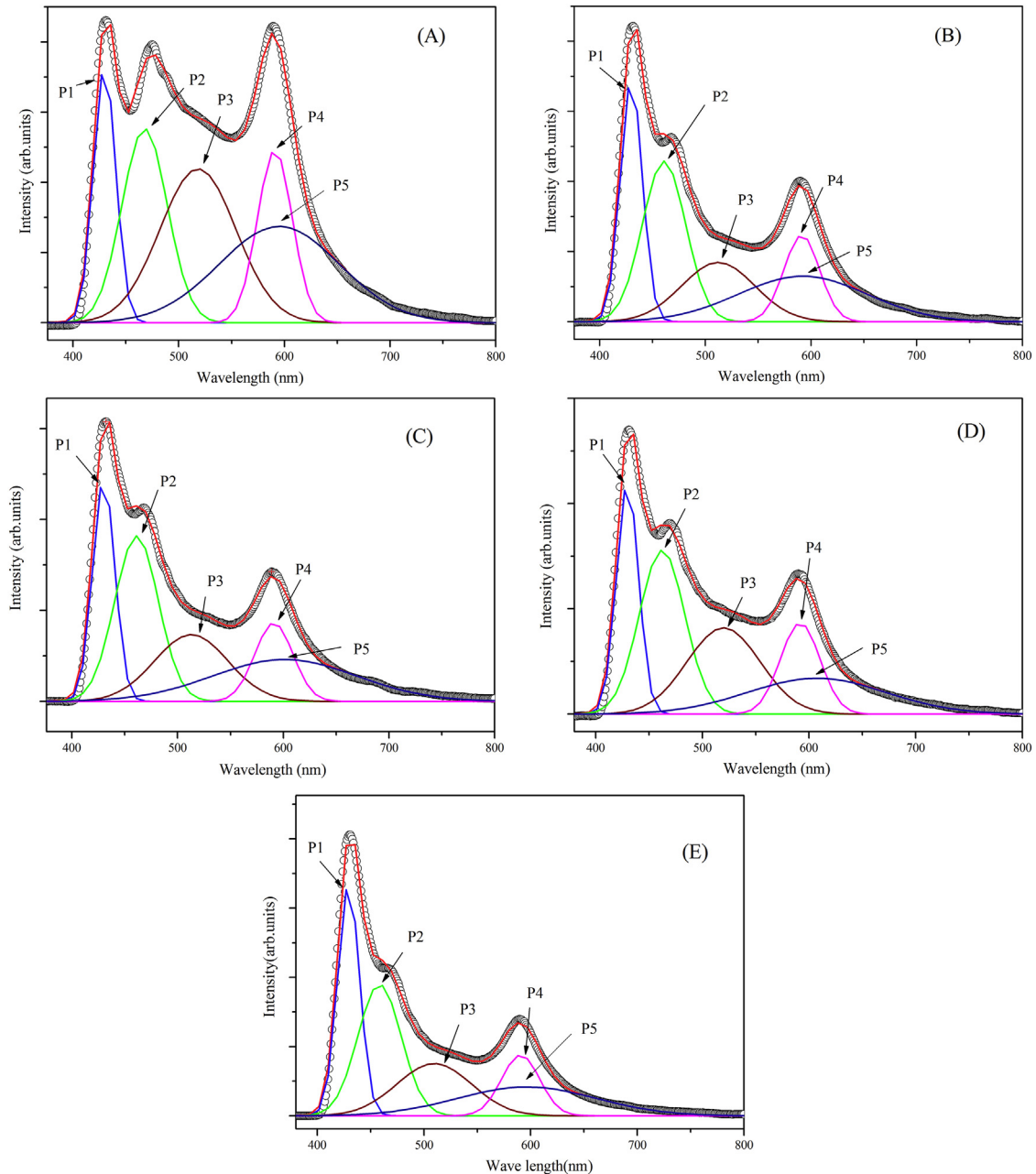
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normal direct. A 75 W Xe lamp was used as the excitation source and the excitation wavelength was fixed at 325 nm. A transmission filter (L42) was used to eliminate the influence of overtone bands centered at 650 nm. Infrared absorption measurements were carried out by Fourier transform infrared absorption spectrometer (FTIR, VERTEX 70) in range of 400–4500  $\text{cm}^{-1}$ , where the absorption of the silicon substrate was carefully corrected by a bare silicon wafer. The chemical composition analyses were carried out by X-ray photoelectron spectroscopy (XPS, VG Multilab 2000), using a monochromatic Al  $K\alpha$  (1486.5 eV) X-ray source and a hemispherical energy analyzer. Before detections, the samples were sputtered using a beam of 3 kV  $\times$  2  $\mu\text{A}$   $\text{Ar}^+$  bombardments for 180 s. The synthesis of Si-QDs was characterized by Raman scattering spectroscopy and directly by high-resolution transmission electron microscope. Raman scattering spectroscopy was performed by a micro-Raman Renishaw spectro-photometer (HORIBA Jobin Yvon LabRAM Spectrometer HR 800 UV) in range of 100–1200  $\text{cm}^{-1}$  in backscattering

configuration, whose laser excitation was 514 nm  $\text{Ar}^+$  line and the incident power was kept low to avoid crystallization and distortion. High-resolution transmission electron microscope (HRTEM, JEM-2100F) were performed at an accelerating voltage of 200 kV. The electron transparent samples were prepared by conventional mechanical polishing and  $\text{Ar}^+$  thinning technology.

### 3. Results and discussion

Fig. 1(a) to (e) shows the PL measurements of the as-grown sample and the samples annealed at 750, 900, 1050, and 1200  $^{\circ}\text{C}$  in range of 300–800 nm, respectively. It is clear that all of the samples show room-temperature PL across the whole visible light range, whose shape and intensity change slightly with the annealing temperature increase. In order to analyze the origin of the PL, each spectrum is



**Fig. 1.** (Color online) Room-temperature photoluminescence spectra of the as-grown sample and the samples annealed at 750, 900, 1050, and 1200  $^{\circ}\text{C}$ , respectively (black circles), together with the curves fitted.

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