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Effects of interfacial barrier confinement and interfacial states on the light emission of si nanocrystals

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HIGHLIGHTS

- Effect of interfacial barrier confinement is demonstrated to be determinative for the photoluminescence of Si nanocrystals.
- Effect of interfacial states is also a key factor in the photoluminescence of Si nanocrystals.
- Both interfacial effects hold for the electroluminescence of Si nanocrystals.

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ABSTRACT

The light emission of Si nanocrystal (Si-nc) depends not only on the Si-nc itself, but also on the interface between Si-nc and its matrix. However, systematic reports on the interfacial effects are rare. In this work, we investigate two interfacial effects, i.e., the effect of interfacial barrier confinement and that of interfacial states, on the photoluminescence (PL) of Si-nc in a systematic manner via designing Si-nc samples with different matrices composed of SiO_2 or/and Si_3N_4, and monitoring their PL emissions as functions of exciting photon wavelengths, combined with electron occupation probability calculations. Our results demonstrate that both interfacial effects affect strongly the PL emission of Si-nc. Interfacial states favorable and unfavorable for light emission are also examined. The conclusions drawn from the PL studies also hold for the electroluminescence (EL) of Si-nc, and the interfacial effects should be a major concern for the EL of Si-nc in addition to the carrier transfer.

1. Introduction

Si-based light emission is of critical importance for Si optoelectronics [1,2]. The composite of Si nanocrystals (Si-nc's) embedded in dielectric matrix such as SiO_2, Si_3N_4, or SiC has been regarded as a promising material for Si light sources due to its robustness in strength, stable light emission and feature of stimulated emission [1–27]. However, practical Si light sources, whether photoexcited or LED-like, are still unavailable after years of studies. One of the bottleneck problems lies in low light emission of Si-nc. To enhance the photoluminescence (PL) of Si-nc, various approaches have been proposed such as hydrogen passivation [3,8], Si-nc density modulation [22,26], and rare earth doping [8], for example. To promote the electroluminescence (EL) of Si-nc, efforts are mainly focused on designing novel channels for carrier tunneling [14,15,23,25], lowering the interfacial barrier between Si-nc and its matrix by using small band gap-width dielectrics [16,20,27], and reducing the turn-on voltage and enhancing charge transfer by using randomized morphology [9,10]. It has been known that some interfacial states are responsible for the light emission of Si-nc, while others act as non-radiative centers [3,4,7,12,17]. Our recent work shows that when the interfacial barrier confinement is weakened, the PL intensity of Si-nc could be drastically reduced, no matter how high the Si-nc density is [24]. However, systematic works about the effect of interfacial barrier confinement and that of interfacial states on the light emission of Si-nc are still rare. In this work, we investigate the two interfacial effects on the PL of Si-nc in a systematic manner. Four multilayered samples of Si-nc are proposed with matrices different in either interfacial barrier or interfacial state or both. The time-integrated PL spectra of Si-nc are monitored as functions of exciting photon wavelength (energy), and spectra before and after hydrogen passivation are compared. The conclusions drawn from the PL studies also hold for the EL of Si-nc, and the interfacial effects are a major concern for EL enhancement in addition to the carrier injection and transfer.

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2. Experimental

All the samples were prepared on degreased and supersonically cleaned substrates of p-type Si < 1 0 0 > (0.5–1.0 Ω cm) in a high-vacuum chamber with a base pressure less than 3×10^{-4} Pa. Multilayered samples of Si(1.5)/SiO_2(5), Si(1.5)/Si_3N_4(5), Si(1.5)/SiO_2(1.5)/Si_3N_4(2)/SiO_2(1.5) and Si(1.5)/Si_3N_4(1.5)/SiO_2 (2)/Si_3N_4 (1.5), termed as samples *a*, *b*, *c* and *d*, respectively, were made by alternatively evaporating Si, SiO_2 and Si_3N_4 with electron-beam heating, followed by thermal annealing in a tube furnace at 1100 °C for 1 h for a process of phase-separation, during which, Si-nc's were formed in the amorphous Si layers sandwiched by SiO_2 or Si_3N_4 layers [8,21,22,24–26]. Numbers in the parentheses mean the layer thicknesses in nanometer. For all the samples, the period number was10, with a SiO_2 or Si_3N_4 layer at the topmost. The total thickness was 65 nm. The thin film thickness was monitored by a calibrated microbalance (Sigma, SQM-160) during the deposition process. The PL measurement was performed at room temperature on a spectrophotometer (Hitachi, F-4500). For EL measurement, Al were firstly evaporated at the backside of the sample substrate, with thickness greater than 10 μm, followed by heating in pure nitrogen at 480 °C for 10 min to form an Ohmic contact. An aluminum ring electrode was then evaporated onto the front surface of sample, followed by heating in nitrogen at 200 °C for 5 min. The forwardly biased voltage and current for EL excitation were provided by a source-meter (Keithely, 2400). The EL spectra were measured also with the F-4500 spectrophotometer. Hydrogen passivation of the four samples were performed in a forming gas (H_2:N_2=5%:95%) at 600 °C for 0.5 h.

3. Results and discussion

In Fig. 1(a)–(d), one-period band diagrams of the samples *a*, *b*, *c*, and *d* are illustrated in terms of the band gap and layer thickness. The band gap widths of SiO_2 and Si_3N_4 are taken to be 9.0 and 5.1 eV, respectively, from Ref. [16]. The PL of Si-nc is due to the recombination of electron located at an interfacial state, with the hole at the maximum of the valence band of Si-nc [17]. Since the interfacial state is very close to the bottom of conduction band of Si-nc in energy [17], the band gap width of Si-nc can be estimated from its PL peak position as shown in Fig. 2, which corresponds to 1.7 eV. The relative positions of the band energies in Fig. 1 are arranged in terms of Refs. [14,15]. Fig. 2 gives the PL spectra of samples *a*–*d*. The exciting photon wavelength is 300 nm. It is seen in Fig. 2 that the PL intensity of sample *a* (or PL*a*) is ~23 times that of sample *b* (or PL*b*), ~6 times that of sample c (or PL*c*), and ~23 times that of sample *d* (or PL*d*). In Fig. 3, the ratios of PL*a*/PL*b* and PL*a*/PL*c* versus exciting photon wavelengths or photon energies are plotted. In terms of the photon energy scale, the two ratios increase slowly with the increasing photon energy at first, until at ~3.1 eV (λ~400 nm) both start to increase significantly and then at ~3.4 eV (λ~350 nm), they increase slowly again. From Fig. 1(b), it is seen that to overcome the barrier confinement of Si_3N_4, the exciting photon energy should be ≥3.4 eV ($\lambda \leq 350$ nm). Therefore, the trends of PL intensity ratio versus photon wavelength or energy are consistent with the effect of interfacial barrier confinement. In terms of the photon energy scale, for PL*a*/PL*b*, when the photon energy is < ~3.4 eV, the excited electrons are all confined within Si-nc wells of samples *a* and *b*. With the

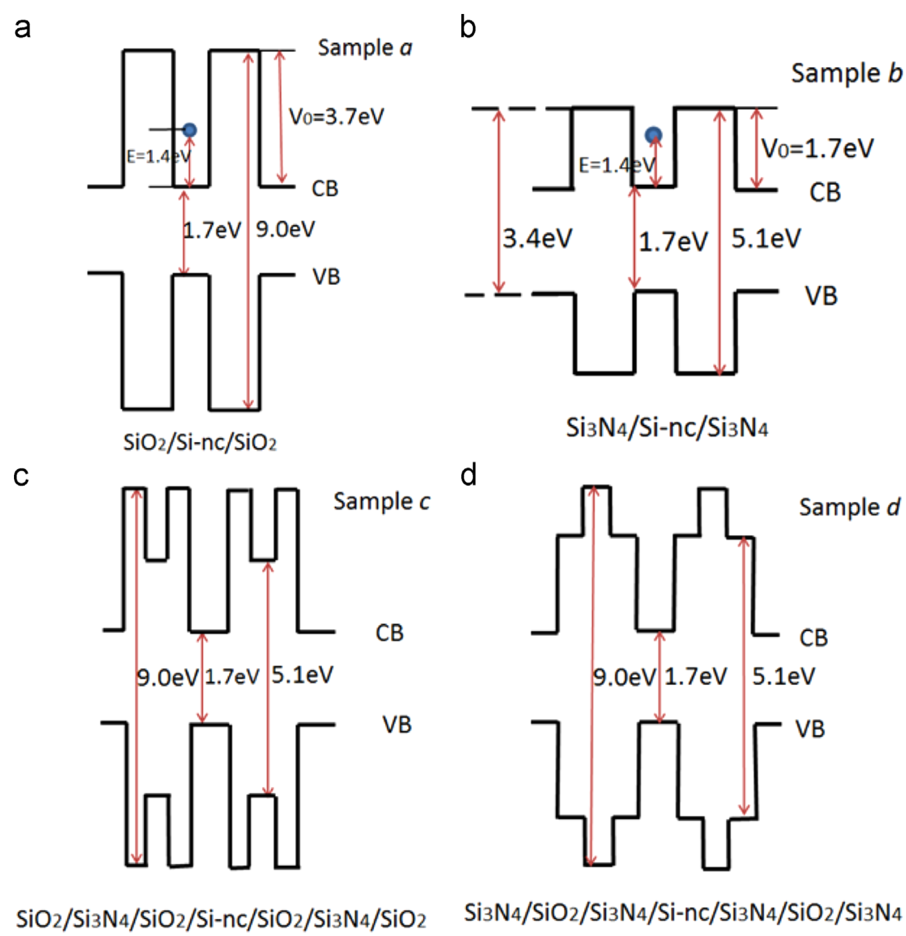

Fig. 1. Band diagrams of the samples *a* (a), *b* (b), *c*(c) and *d*(d). CB means the bottom of conduction band of Si-nc, VB means the maximum of valence band of Si-nc, V_0 is the interfacial barrier height, and *E* is the electron energy with respect to CB.