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Effect of the reflection by the silicon aggregates on the photoluminescence from silicon nitride film embedded silicon nanocrystals

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ABSTRACT

In this work, the effect of the high silicon nanocrystals density as well as the radiated waves interference on the photoluminescence (PL) spectrum of silicon nanocrystal (Si-nc) embedded in silicon nitride film is studied. The film is prepared using low pressure chemical vapor deposition (LPCVD) following by a high temperature annealing. It was found that for silicon nitride of a high silicon content sample, the interference effect based on only the thickness of the film is unable to satisfactory explain the distortion on the PL spectrum. Using Monte Carlo analysis, it was shown that the simulated and the experimental PL spectra accurately superpose when taking into account the contribution of the reflection from silicon nanocrystals as well as the distance traveled by the radiated waves.

1. Introduction

The production and the study [1–10] of photoluminescence from silicon nitride thin film containing silicon nanocrystals(Si-ncs) has acquired growing interest during the last years in order to develop low-cost, reliable, and efficient silicon-based photonic, and photovoltaic devices.

Despite the great number of research, the origin of the PL is not yet understood especially in term of stokes shift and distortion on the PL spectrum. The first proposed model trying the explanation of the PL is the quantum confinement effect model (QCE) [11,12]. Easy to perceive, it is unable to predict the position of the PL spectrum maximum, and the appearance of multiple peaks observed in the experimental spectra [11–18]. Consequently, the QCE model has been extended to include the PL from surface states as proposed by Islam et al. [13], and PL from the cap shell surrounding the nanocrystal as postulated by Daldosso et al. [14]. According to Kistner et al. [15], the PL is originated from recombination of excited carriers between the band tail states presented inside the SiN_x band gap. Recently, Rodriguez-Gomez et al. [17] found experimentally and theoretically that the increase of the thickness of SiN_x above a few hundreds of nanometers generates considerable distortions of the PL spectra of the films, such as the PL peak position and the appearance of multiple peaks, due to interference between reflected radiated waves by interfaces of the composite film. The suggestion of Rodriguez-Gomez et al. [17] permits the superposition of both the simulated and the experimental PL spectra for samples embedded only 12% of silicon nanocrystals. Unfortunately, silicon nitride of a high silicon content matrix allows also the light reflection by the silicon aggregates. Thus, the model proposed by Rodriguez-Gomez et al. [17] requires the introduction of the intrinsic properties of the thin film in order to investigate all SiN_x composition.

The deposit of amorphous SiN_x, ($x = \frac{N}{Si}$=0.12) thin film (thickness around 200 nm) was carried out in a conventional hot-wall, horizontal, LPCVD furnace by using disilane (Si_2H_6) and ammonia (NH_3) [19–21] gaseous mixture on (111) oxidized (about 120 nm of oxide) silicon wafers. The deposition temperature and the total pressure were, respectively, fixed to 465 °C and 200 mTorr. More experimental details can be found in Ref. [22]. The Si-ncs formation was assured with subsequent temperature annealing at 1050 °C for 1 h under a nitrogen (N_2) atmosphere in a conventional furnace. From Raman measurement, the silicon nanocrystal density was found 71.66% [22]. The PL was measured using a laser excitation source at 355 nm with a power density of 3:27 mW, a spot-size of 1 nm, and a resolution of 6:8 nm.

In this work, the effects of both the Si-ncs density (exceeded 70%) and the light interference in SiN_x (x=0.12) film are investigated. Based on Rodriguez-Gomez et al. model [17], it was shown that the difference between the measured and the calculated PL spectra is considerable for the present sample with high density of Si-ncs. On the other hand, the simulated spectrum has

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only a single peak due to the small sample thickness. Thus, the theoretical simulation will be performed to restore the PL spectrum using intrinsic as well as extrinsic properties of the thin film.

2. Theoretical study

In this section, we will develop the luminescence model proposed by Rodriguez-Gomez et al. [17]. Their model supposes that the light emitting by each nanocrystal inside a silicon nitride matrix regarded as uniform is subjected to multiply reflection at interfaces between the amorphous matrix and both incidence medium and substrate. Assuming that the reflected waves are coherent with respect to each other, the total light radiated by the nanocrystal is the sum of all reflections of the radiated wave in the same direction.

Results issued from the interference-effect model agree well with the experiment. However, the model can not be applicable for silicon nitride of a high silicon content, because the number of crystalline aggregates break the uniformity of the amorphous matrix as assumed by Rodriguez-Gomez et al. [17]. As shown in Fig. 1, the simulated PL spectrum of SiN_x (x=0.12) is shifted considerably from the experimental. Moreover, it has a Gaussian shape and does not exhibit interference peaks. This difference between both experimental and simulated spectra is due to the silicon aggregates that reflect partially the radiated waves. Thus, a new model taking into account the contribution of the reflection by nanocrystals seems to be necessary for the investigation of the light emitting by this samples.

The studied thin film is deposited on oxidized silicon matrix and composed of an ensemble of nanocrystals coated by a transition region characterized by the absence of reticular plane, and has a key role in the luminescence [14]. The Si-ncs are randomly distributed in an amorphous silicon nitride (SiN_x) matrix of uniform thickness d along the lateral dimensions[23]. When exciting by a plane wave of vacuum wavelength λ_{exc} (the surface scattering is neglected because the LPCVD deposit process using the disilane as precursor allows us to achieve a smoother film [23]), photocarriers are generated inside the crystallites (path a) [13], and then a fraction of these photoexcited recombine radiatively inside the Si-nc (path b) and the other part diffuses to the amorphous matrix (path c). The diffused photocarriers recombine radiatively (path d) between band tail states (Fig. 2). At the same time, photocarriers are generated at the amorphous matrix and recombined inside the Si-ncs [8]. As the matrix density is neglected in comparison with that of Si-ncs (71.66%) [22], it is more probable that the excitation occurs inside the Si-ncs.

The radius of the nanocrystals less than 2.13 nm (gap upper than 1.89 eV) allows the overlap between the excited states of the nanocrystal and the band tails of the amorphous matrix and thus

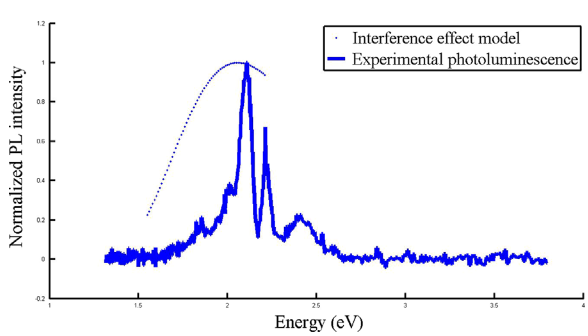

Fig. 1. Normalized simulated and experimental spectra for the SiN_x (x = 0.12). The radiated wave is subjected to multiple reflections with both the upper and the lower interfaces.

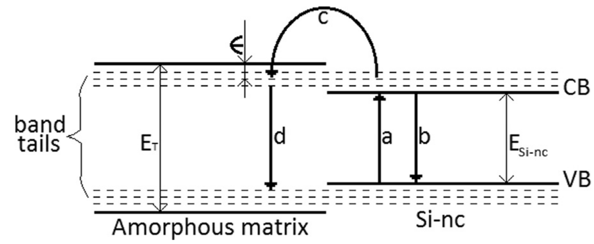

Fig. 2. Schematic of possible excitonic recombination:(a) generation of photocarriers, (b) recombination from conduction band (CB) to valence band (VB), (c) diffusion of photocarriers from Si-nc to amorphous matrix, (d) radiative recombination between band tails states.

the diffusion of the photocarriers from the Si-nc to its cap shell that is a region of the amorphous matrix [14]. Because of the probability of radiative recombination inside the Si-nc is negligible compared with that between band tails of the amorphous matrix, the majority of the excited electrons recombine radiatively neighboring the nanocrystal (in the cap shell). Thus the PL intensity $S(E_T - \epsilon)$ emitted from a single nanocrystal is proportional to the population of optically active state inside the Si-nc ($N_S \sim r^3 = \left(\frac{C}{E_{Si-nc} - E_g}\right)^{3/n}$ where E_g=1.13 eV is the gap energy of the bulk silicon, and $E_{\text{Si-nc}}$ is the gap energy of the Si-nc) [22], and both the excitable states and the emission probability of the amorphous matrix as [15]:

$$S(E_T - \epsilon) = \left(\frac{C}{E_T - \epsilon - E_g}\right)^{-3/n}\left(exp\left(-\frac{\epsilon}{2E_U}\right)\right)\left(1 - \exp\left(-\frac{\epsilon}{2E_U}\right)\right)^N \quad (1)$$

The thermalization is allowed among N+1 neighboring states, and the PL could occur from the lowest of these states with an energy (ϵ) below the optical gap (E_T=2.21 eV) [22]. E_U=0.16 eV is the Urbach energy deduced from our previous work [22]. C=3.48 eV nm^2 is the confinement factor, and n=2 is a constant [24].

The radiated waves may get multiply reflected at the interfaces with the incidence medium and the substrate resulting in a field whose amplitude oscillates inside the film [17]. Each Si-nc (the core and its cap shell) behaves as a dipole antenna, it emits a plane waves incoherently with respect to all other Si-ncs. However, all reflected waves coming from a given Si-nc are coherent with respect to each other.

The proposed model supposes that the light radiated from a given Si-nc is traveling up towards the incidence medium (We suppose that all the Si-ncs contribute to the luminescence phenomenon. However, the emitting yield depends strongly on their size [12].). It can reach the upper interface and be transmitted partially to the incidence medium or hit a Si-nc and gets reflected partially (Fig. 3). The radiated waves reflected from the upper interface or the Si-nc can also either reach the interface with

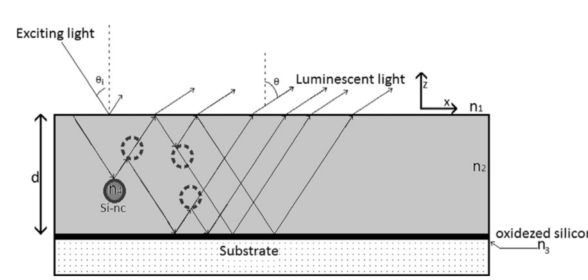

Fig. 3. Schematic of the studied film exposed to the exciting light (I_{exc}) at an oblique angle (θ_i) and emitting luminescence light back into the incidence medium. The composite film is subjected to multiple reflection of the radiated wave with the substrate, the incidence medium and the Si-ncs.