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Specific Blue Light Emission from Nanocrystalline Porous Si Treated by High-Pressure Water Vapor Annealing

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1. Introduction

Si-based light-emitting materials are highly desirable in order to (i) lower the cost of optoelectronic devices, (ii) enable VLSI compatible optical interconnects and opto-electronics and (iii) replace currently used toxic or pollutant materials.

Red-band photoluminescence (PL) of nanocrystalline porous Si (PSi) can be highly enhanced and stabilized by high-pressure water vapor annealing (HWA)[1,2]. Based on this approach, the electroluminescence emission has been much improved[3].

The practical blue shift in the PL emission has not been easily obtained from quantum confined Si systems due to localized states introduced inside the bandgap by oxygen-related defects on the Si surface.[4] Some blue PL emissions in oxidized PSi have been reported, but it is usually very weak and unstable.[5,6]

In this paper, we show that PSi treated by HWA under appropriate conditions can exhibit efficient blue and stable emission. The mechanism of the blue emission is discussed on a basis of spectroscopic surface analyses.

2. Experimental

The substrates used were (100)-oriented, B-doped p-type (4 Ωcm) silicon wafers. The PSi layers were formed by conventional anodization in an ethanoic HF solution under galvanostatic condition in the dark. For HWA treatment, PSi samples were heated in water vapor at a temperature of 260-300 °C for 2-3 h as reported in previous papers[1,2], and then cooled down to room temperature. The water vapor pressure in the container during annealing was adjusted to a value in the range of 1 to 3.9 MPa.

The PL measurements were carried out at room and low temperatures. A He-Cd laser (325 nm line) or a YAG laser (266 nm) was used as an excitation source. To study the blue light emission mechanism, surface characterization was done by Fourier transform infra-red (FTIR) analyses.

3. Results and Discussion

Photoluminescence

Figure 1 shows PL spectra of PSi treated by HWA acquired at 14 and 295 K. Two peaks are clearly observed at both temperatures. One is located in the blue spectral range (blue band) whereas the other is located in the red spectral range (red band). This behavior contrasts with that of conventional PSi PL where only a one peak red band is usually observed. The intensity of the blue emission peak is very significant even at room temperature. It becomes dominant at low temperature. This result shows that efficient blue PL is achievable by using HWA.

Each spectrum can be fitted by the sum of two Gaussians. Table 1 compares the integrated intensity (I_{PL}), the full width at half maximum (FWHM) and the peak wavelength (λ_{PEAK}) of the red and blue spectra at two different temperatures.

The relative intensity of the blue PL emission is enhanced with increasing the pressure and temperature at HWA. Under the HWA conditions to promote sufficient oxidation, the blue emission tends to be dominant even at room temperature, and then it becomes possible to get a single emission band at a peak wavelength of 450 nm without any signs of the red emission.

Basic features of blue emission

The blue PL emission observed in HWA-treated samples is characterized by the following specific properties:

- As shown in Fig. 1, the peak wavelength shows little blue shift with lowering temperature in contrast to the behavior of the red emission peak.
- Both the bandwidth and the peak wavelength of the blue emission are stable even in air, independent of the excitation power.
- The blue emission exhibits a relatively large polarization memory effect for the excitation light in comparison with the red emission. It looks that the origin of the blue PL is isotropic.
- There is a distinct difference in the decay dynamics between the blue and red emissions. Significant wavelength dispersion in the lifetime of the red emission disappears in the blue emission.

Mechanism of the blue luminescence

For the red PL band, FWHM and λ_{PEAK} depend significantly on the temperature, excitation power, and the sample preparation conditions. On the contrary, spectral features of the blue PL remain mostly temperature-independent, and do not depend on both the excitation power and the sample preparation conditions. The excitation power dependencies of red and blue emissions are summarized in Fig. 2. Also the blue PL shows characteristic behavior in the polarization memory and the lifetime. These results suggest that the blue emission originates from localized states, while the red emission from radiative recombinations of localized excitons in Si nanocrystals.

In accordance with FTIR analyses, the intensity of the

blue emission is positively correlated to the amount of oxide generated by HWA in the PSi samples. Thus, it probably originates from defects in the oxide. The oxygen deficiency center, known to emit blue PL[7], is a possible defect candidate. Details of the HWA effect on the blue emission are, however, yet to be clarified especially in relation to the decay dynamics.

4. Conclusions

Strong and stable blue emission was obtained from HWA-treated PSi. The related phenomena imply that the blue emission is attributed to defects in the oxide part of the samples. HWA is very useful to boost the emission from these defects. The present result is useful for the realization of short-wavelength Si-based optoelectronic devices.

Acknowledgements

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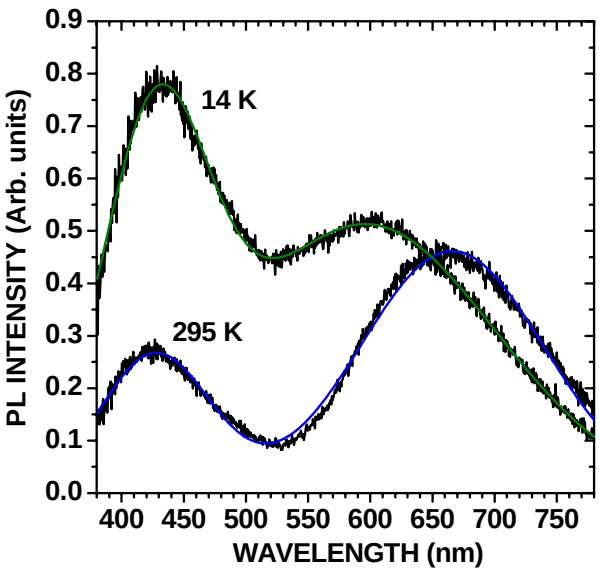

Fig. 1 PL spectra of HWA-treated PSi at 14 and 295 K. The lines are fits of PL data to the sum of two Gaussians. Excitation wavelength: 325 nm.

Table I Parameters of the two Gaussians whose sum was used to fit the spectra shown in Fig. 1. I_{PL} is the integrated intensity, FWHM is the full width at half maximum and λ_{PEAK} is the peak wavelength of each Gaussian.

PL Band	I_{PL} (Arb. unit)	FWHM (nm)	λ_{PEAK} (nm)
Red at 14 K	134	288	596
Red at 295 K	85	205	666
Blue at 14 K	68	118	427
Blue at 295 K	30	123	427

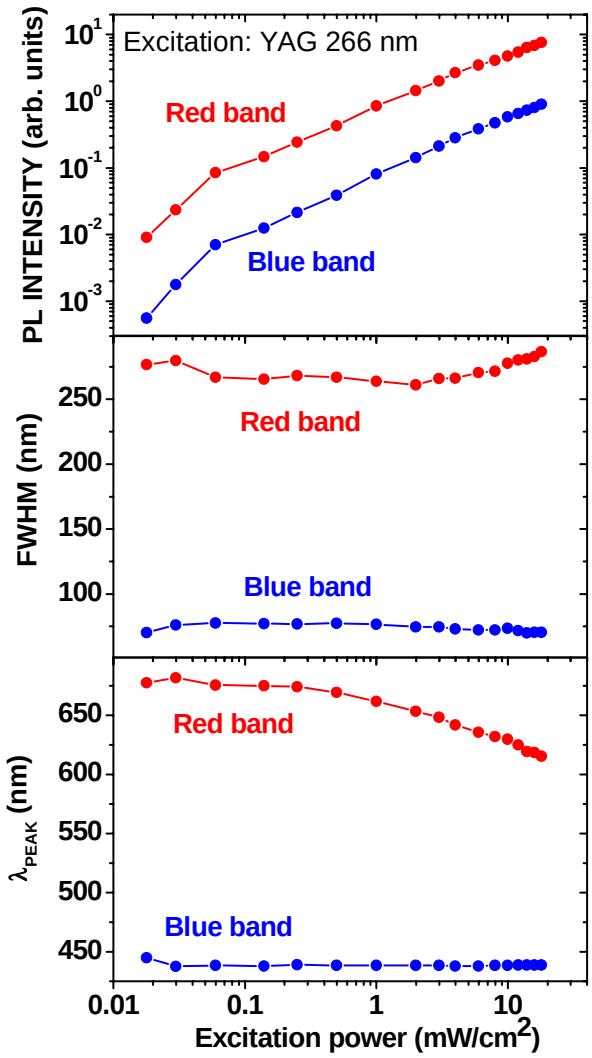

Fig. 2 The PL intensity, full width at half maximum (FWHM) and peak wavelength (λ_{PEAK}) of the red and blue emission bands at a low temperature (11 K) as a function of the excitation intensity (a 266 nm YAG laser was used in this case). Note that the peak wavelength of the blue band is totally independent of the excitation power, while the red emission peak shows a blue shift.