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Strong photoluminescence anisotropy in porous silicon layers prepared by polarized-light assisted anodization

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Abstract

Linearly-polarized infrared (1.06 μm) laser light with intensities ranging from 5.3 to 97 mW/cm^2 has been used to obtain anisotropically luminescent porous silicon (PSi) layers by photoanodic etching in a hydrofluoric acid solution. Remarkably large photoluminescence (PL) anisotropy has been observed in samples prepared with the highest illumination intensity. These samples show very low degrees of linear polarization when the PL excitation light is polarized parallel to the polarization direction of the etching light. When the excitation light is polarized perpendicular to that, we obtain usual degrees of linear polarization of several percent. This result indicates that anisotropic Si nanostructures in PSi layers can be made isotropic with high orientation selectivity by the polarized-light assisted technique. A simple two-dimensional model is presented to explain the observed prominent anisotropy.

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1. Introduction

The efficient visible photoluminescence (PL) from porous silicon (PSi) layers is currently a subject of intense research [1–4]. PSi layers are formed by electrochemically anodizing Si wafers in HF solutions. These layers contain a high density of Si nanocrystals that are thought to be responsible for the visible PL [5,6]. The Si nanocrystals are connected electronically to each other as revealed by significant electrical conductivities [7]. This structural property produces a significant electronic anisotropy around each luminescent Si nanocrystal. In fact, linearly polarized laser light preferentially excites those Si nanocrystals that have higher optical transition probabilities in the direction of $\boldsymbol{e}_{\mathbf{exc}}$, where $\boldsymbol{e}_{\mathbf{exc}}$ is a unit vector oriented parallel to the polarization direction of the excitation light. This results in a unique optical phenomenon called polarization memory [8–12], i.e. PL partially polarized in the same direction as the excitation light.

The microscopic electronic anisotropy can cause macroscopic phenomena if these anisotropic nanocrystals are oriented along the same direction. For PSi, this can be realized in two ways. One is to anodize asymmetric Si substrates such as (110)-oriented wafers [13–15]. The PSi layers produced in this way exhibits partially polarized PL with the largest degree of linear polarization being observed when $\boldsymbol{e}_{\mathbf{exc}}$ is parallel to the [001] axis of the wafer [13]. Significant birefringence is also observed [15], allowing for the fabrication of polarization-sensitive optical components. The other method is to illuminate the wafer with linearly polarized-light when it is anodically etched [13,16–19]. In this case, the maximum degree of polarization in PL can appear when $\boldsymbol{e}_{\mathbf{exc}}$ is either parallel [17–19] or perpendicular [13,16,18] to $\boldsymbol{e}_{\mathbf{etch}}$, depending on the etching and illumination conditions. Here, $\boldsymbol{e}_{\mathbf{etch}}$ denotes a unit vector along the polarization direction of the etching light. The advantage of this polarized-light assisted method is that it can induce in-plane anisotropy in any direction simply by changing the direction of $\boldsymbol{e}_{\mathbf{etch}}$ [13,18]. Also, this method can potentially be used to produce anisotropic structures in polycrystalline or amorphous layers.

To date, only moderate PL anisotropy has been obtained in PSi layers by either of the methods described above. In this study, we have employed the polarized-light assisted technique with illumination intensities up to a relatively high value. Strong PL anisotropy has been observed in the samples prepared with the highest illumination intensity.

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The remarkable anisotropy can be reproduced well with a model based on two-dimensional distribution of linear oscillators.

2. Experiment

Single-crystal Si (100) wafers of n-type, 0.05–0.5 Ω cm were anodically etched in a solution of 55% HF:C_2H_5OH = 1:3 at 20 mA/cm^2 for 30 min. During the etching, the samples were illuminated with linearly-polarized infrared (1.06 μm) light from a Nd:YAG laser. A laser beam expander was used to ensure a uniform illumination intensity over the entire sample surface. Although the anisotropy can basically be formed independent of the direction of $\boldsymbol{e}_{\mathbf{etch}}$ with respect to the crystalline axes of the wafer [13,18], there often appear small dependencies on the crystalline axes [17–19]. Therefore, all the samples were etched under such conditions that $\boldsymbol{e}_{\mathbf{etch}}$ was parallel to the [011] axis. After the photoelectrochemical etching was completed, the samples were slightly oxidized chemically in a 50-°C solution of 0.1 M H_2SO_4 for 10 min to obtain sufficient stability against laser irradiation during the PL measurements.

PL measurements were carried out at room temperature in an ambient atmosphere. The photoexcitation was provided by a laser diode emitting linearly polarized-light at 410 nm (3.02 eV). PL spectra were measured by a fiber-optic spectrometer (Ocean Optics USB2000) with a resolution of ∼4 nm. The measured PL spectra were corrected for apparatus response. To evaluate the anisotropy, we measured the intensities of the emission components polarized parallel ($I_\parallel$) and perpendicular ($I_\perp$) to $\boldsymbol{e}_{\mathbf{exc}}$. The degree of linear polarization P was then obtained by $P=(I_\parallel - I_\perp)/(I_\parallel + I_\perp)$ [8–12].

3. Results and discussion

Fig. 1 shows PL spectra of three PSi samples prepared with different illumination intensities ranging from 5.3 to 97 mW/cm^2. These spectra have similar lineshapes extending from ∼1.3 to ∼2.2 eV, with a peak being at 1.6–1.7 eV. Their peak intensities, however, are largely different. The sample prepared at 97 mW/cm^2 exhibited a peak PL intensity about 30 times higher than that of the sample made at 5.3 mW/cm^2. These features are similar to those observed in PSi layers prepared under the irradiation of visible light [20].

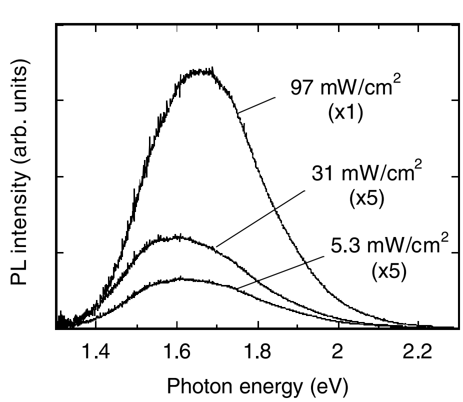

Fig. 1. Unpolarized PL spectra of three PSi samples prepared with different illumination intensities. The spectra corresponding to 31 and 5.3 mW/cm^2 are enlarged by a factor of 5.

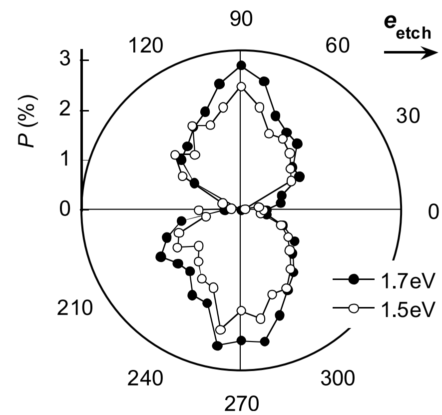

Fig. 2. The degree of linear polarization in the PL of a PSi sample prepared with the highest illumination intensity of 97 mW/cm^2. Data for two different emission energies are plotted as a function of the angle between $\boldsymbol{e}_{\mathbf{exc}}$ and $\boldsymbol{e}_{\mathbf{etch}}$.

Fig. 2 shows the degree of linear polarization in PL obtained from the most highly luminescent sample prepared at 97 mW/cm^2. In this figure, the measured values of P for two different emission photon energies are plotted as a function of the angle (θ) between $\boldsymbol{e}_{\mathbf{exc}}$ and $\boldsymbol{e}_{\mathbf{etch}}$. The experimental data show that while P gives some finite values when $\boldsymbol{e}_{\mathbf{exc}}$ is not parallel to $\boldsymbol{e}_{\mathbf{etch}}$ ($\theta \neq 0$ or 180°), it becomes very small when $\boldsymbol{e}_{\mathbf{exc}}$ is parallel to $\boldsymbol{e}_{\mathbf{etch}}$ ($\theta = 0$ or 180°). This is in clear contrast to the results of previous studies [13,16–19], where only moderate anisotropies are observed.

Fig. 3 compares P obtained for two samples prepared at 97 and 5.3 mW/cm^2. It can be seen that the effect of increasing illumination intensity only appears as decreasing values of P at around $\theta = 0$ and 180° ($\boldsymbol{e}_{\mathbf{exc}} \| \boldsymbol{e}_{\mathbf{etch}}$). No significant changes are observed in the values of P corresponding to $\boldsymbol{e}_{\mathbf{exc}} \perp \boldsymbol{e}_{\mathbf{etch}}$. The result therefore, demonstrates that the polarized-light assisted technique can considerably reduce microscopic anisotropy in PSi layers in the direction parallel to $\boldsymbol{e}_{\mathbf{etch}}$ while keeping them almost unchanged in the direction perpendicular to $\boldsymbol{e}_{\mathbf{etch}}$.

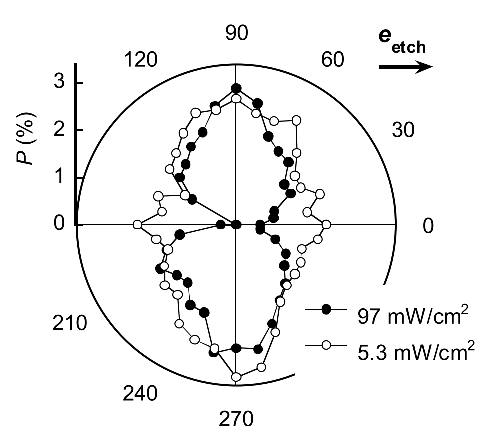

Fig. 3. Comparison of the degree of linear polarization between two PSi samples prepared with different illumination intensities. The data are for the same emission photon energy of 1.7 eV.