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RESEARCH ARTICLE

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Optical properties of low-dimensional structures formed by irradiation of laser

Abstract Some kinds of low-dimensional nanostructures can be formed by the irradiation of laser on a pure silicon sample and SiGe alloy sample. We have studied the photoluminescence (PL) of the hole-net structure of silicon and the porous structure of SiGe where the PL intensity at 706 nm and 725 nm wavelength increases obviously. The effect of intensity-enhancing in the PL peaks cannot be explained within the quantum confinement alone. We propose a mechanism on the increasing PL emission in the above structures, in which the trap state of the interface between SiO_2 and nanocrystal plays an important role.

Keywords low-dimensional nanostructures, photoluminescence, PL intensity, interface state

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1 Introduction

All types of low-dimensional silicon and germanium systems,such as porous silicon, silicon nanocrystals and germanium nanocrystals, are being actively investigated as some means of improving the light-emission properties of silicon and germanium. The physical mechanism underlying high external quantum efficiencies for photoluminescence (PL) in low-dimensional nanostructures is mainly that of the quantum confinement effect [1–10]. However, it is difficult to explain some experiment results using the quantum confinement model alone. We can obtain some hole-net structure of silicon and porous structure of SiGe after sample irradiation of laser, in which the intensity of PL peaks at 706 nm and 725 nm wavelength increases obviously. The frequency of the PL peaks is also not changeable in the different samples prepared with various conditions. The quantum confinement model is not enough for interpreting the intensity-enhancing effect, for which we propose a new three-level system associated with the crystal-oxide interface state. These findings could lead to a new way for some silicon and germanium-based laser.

2 Experiments

The samples used in the study were prepared as follows: Samples A, B and C, respectively.

(A) We took some Si-P (100) oriented wafers with a 10–20 Ω•cm as the pure silicon samples. Having been cleaned in a Summa cleaner (a mixture of choline and methanol), the samples were irradiated for 20 s under a laser beam (spot diameter: 30 μm) with 1064 nm wavelength and 30W power.

(B) The above pure silicon samples were etched by anodizing in HF(49 %): Ethanol(1:1.5) under the irradiation of a laser beam (spot diameter: 700 μm) with 532 nm wavelength and 20 mW power for 15 min. Anodizing current density was about 300 mA/cm^2.

(C) First, the undoped strained $Si_{1-x}Ge_x$ layers (2000 nm, with $x = 0.25$) were grown in a molecular beam epitaxy (MBE) system as the substrate of the sample. The samples were cleaned in a Summa cleaner for 20 min, and then were prepared by anodizing in HF(49 %): Ethanol(1:2) under the

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irradiation of a laser beam (spot diameter: 700 μm) with 532 nm wavelength and 20mW power for 5 min, 10 min, 15 min and 30 min, respectively. Anodizing current density was about 40 mA/cm^2.

Scanning electron microscopy (EPMA-1600) was used to observe the low-dimensional nanostructures of the samples. The photoluminescence spectra of the samples under the 514nm excitation were measured by using RENISHAW Raman Systems.

In Fig. 1, we can find some nanostructures of sample A from the image of SEM and some emission properties from the relative PL spectra. While a laser beam of 30 μm diameter with 1064 nm wavelength and 30 W power hits the pure silicon sample, a kind of hole-net structure of the silicon [Fig. 1 (b)] forms on the side of the hole [Fig. 1 (a)] drilled by the laser. This hole-net structure probably originates from a vibration of the plasma generated in high temperature with laser drilling. It is interesting that the inter-wall of about 10 nm thickness in the hole-net structure forms a quantum well. We think that the intensive PL peak at 706 nm [Fig. 1 (c)] should be related to the quantum well of the inter-wall. The other parts on the sample emit weaker PL or do not emit PL almost.

Figure 2 (a) shows a kind of condensed rod structure in the irradiated and anodized region. It is clear that the irradiating of a laser beam makes the porous layer crack, screw and condensate into some rod state [Fig. 2 (b)]. It is the rod that emits a more intensive PL peak [Fig. 2 (c)].

The SEM in Fig. 3 shows the low-dimensional structures of sample C. Part (a), (b), (c) and (d) are the images of the samples anodized and irradiated for 5 min, 10 min, 15 min, and 30 min, respectively. A weaker laser irradiation (spot diameter: 700 μm) on the strained $Si_{1-x}Ge_x$ film can form some dots and lines structures as shown in Fig. 3 (a) and (b). A stronger irradiation of laser (beam spot diameter: 300 μm) can dig up the $Si_{1-x}Ge_x$ film along some certain direction to form some nano-strap pieces [Fig.3 (c)]. We have found that the nano-strap pieces can emit an intensive PL with a red-shift frequency obviously [Fig.4 (c)].

Under anodizing and irradiation for 30 min, the $Si_{1-x}Ge_x$ layer is almost dug out, and a deeper porous film forms on the substrate [Fig. 3 (d)]. The intensive PL peaks at 724 nm and 725 nm wavelength can be found from the porous structure [Fig. 4 (a) and (b)].

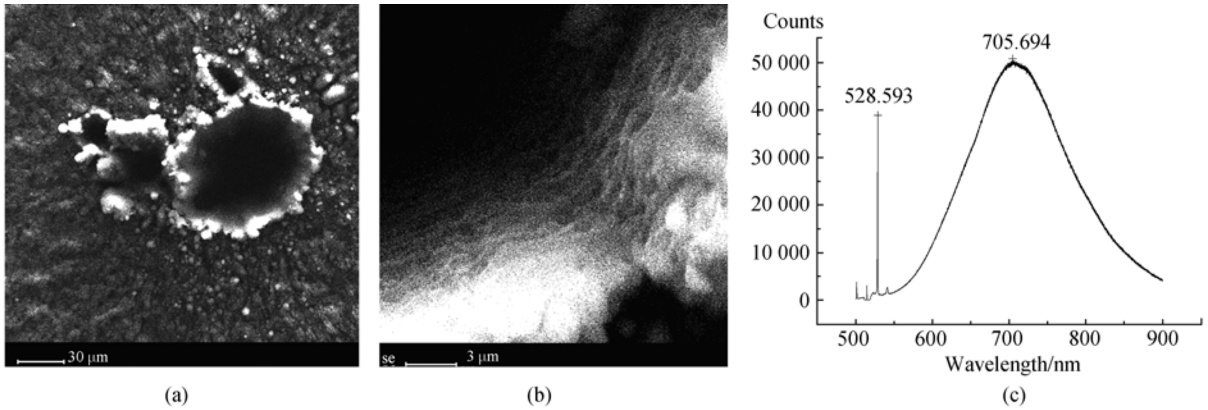

Fig. 1 The results of preparing sample A: **(a)** The hole drilled using a 30 μm spot of the laser beam with 1064 nm wavelength and 30 W power for 20 min; **(b)** The hole-net structure of silicon forming on the side of the hole drilled by the laser; **(c)** The PL peak at 706 nm wavelength related to the hole-net structure of silicon.

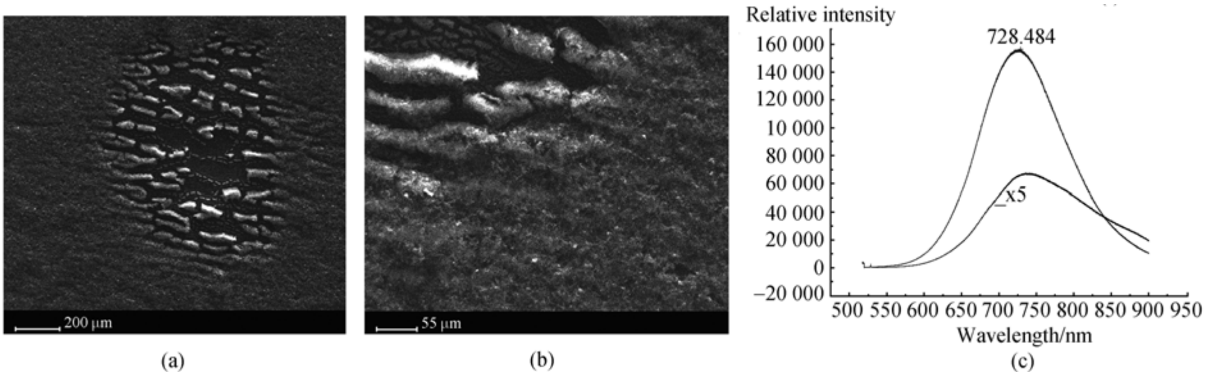

Fig. 2 The results of preparing sample B: **(a)** Anodizing the pure silicon with the irradiation of the laser beam for 15 min; **(b)** The boundary region of the irradiating spot on the sample; **(c)** Comparing the PL peak emitted from the condensed rods in the irradiating and anodizing region of the sample B with that from the porous silicon sample anodized alone, the former is higher and sharper obviously.

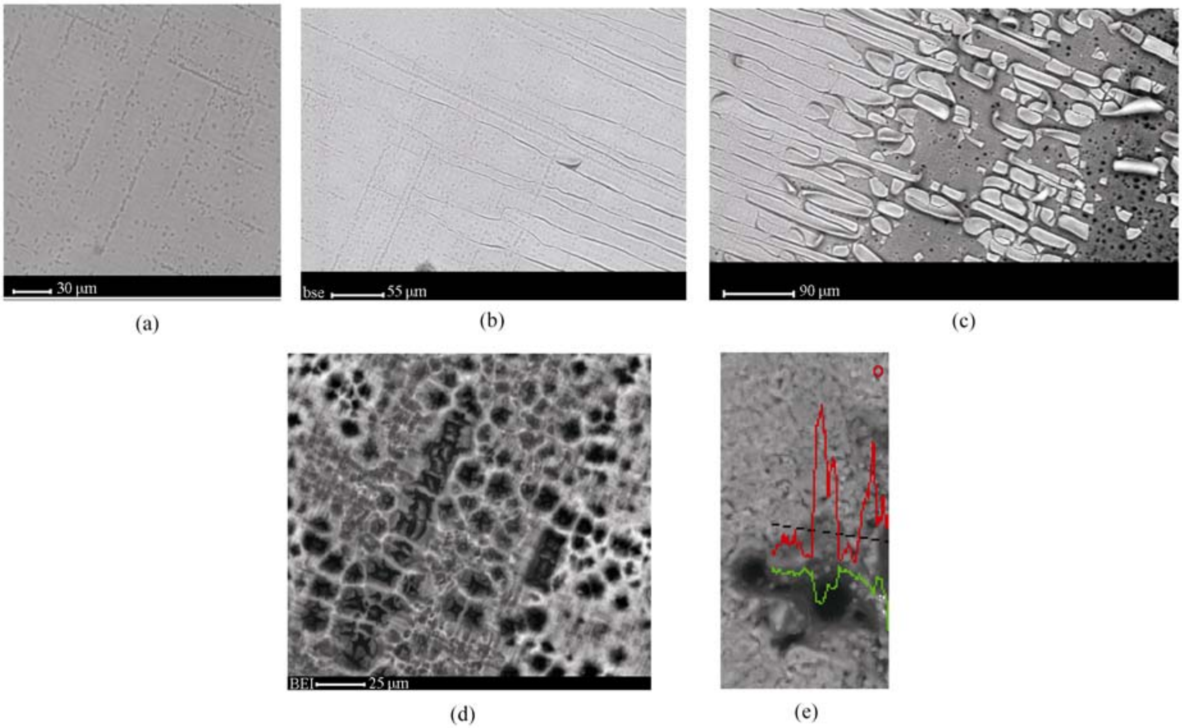

Fig. 3 SEM image of the low-dimensional structures of sample C: **(a)** Dots structure and **(b)** Lines structure on the $Si_{1-x}Ge_x$ film formed under a weaker irradiation of laser; **(c)** Nano-strap pieces structure after digging up the $Si_{1-x}Ge_x$ layer with a stronger laser; **(d)** Porous structure on the substrate under anodizing and irradiation for 30 min, when the $Si_{1-x}Ge_x$ layer is almost dug out; **(e)** Oxygen distribution diagram on the porous structure [Fig.3 (d)].

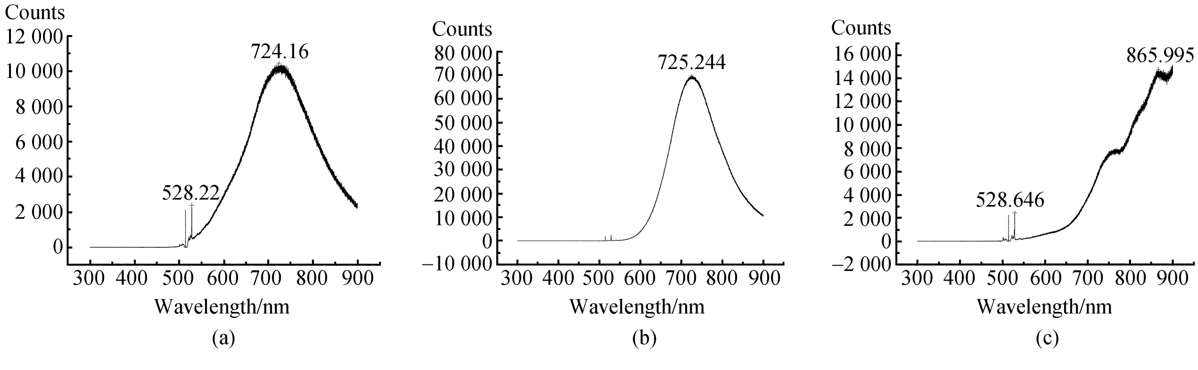

Fig. 4 PL spectra of the low-dimensional structures of sample C: **(a)** PL peak emitted from the low oxygen part of the porous structure [related to Fig. 3 (d) and (e)]; **(b)** PL peaks emitted from the high oxygen part of the porous structure [related to Fig. 3 (d) and (e)]; **(c)** PL spectra from the nano-strap pieces structure [Fig. 3 (c)].

3 Discussions

The preparation method of sample A is very easy to control in which the physical conception for forming the hole-net structure of silicon is very clear. The vibration of the plasma generated in high temperature with laser drilling produces the hole-net nanostructure of silicon on the side of the hole. The mechanism of the PL emission in the hole-net nanostructure can be explained not only with the quantum confinement effect in the quantum well of the inter-wall but also with the interface state between the silicon crystal in the hole-net nanostructure and the oxide of the surface.

As shown in Fig. 5, we propose a new three-level system associated with the crystal-oxide interfacial state to interpret the PL peak at 706 nm wavelength on sample A. An injection photo excites an electron at the top of valence band to jump up to the bottom of the conduction band which is called an absorption jumping process whose gap width increases because of the quantum confinement effect of the inter-wall in the hole-net nanostructure. It is a fast process for the electron to jump from the level E_v to the level E_c [$\Delta E = E_c - E_v$=1.12 eV+0.98 eV (quantum confinement gap)=2.1 eV (up edge of the PL band)]. The quantum confinement gap can be calculated with the formula [11−15]:

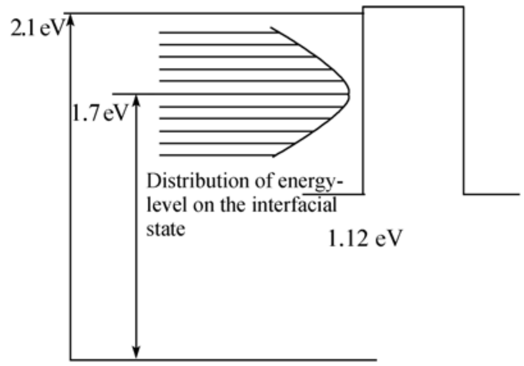

Fig. 5 A new three-level system associated with the crystal-oxide interfacial state.

$$E_{\text{confined gap}} = E_{\text{bulk gap}} + \frac{3h^2}{8} \cdot \frac{1}{L^2} \cdot \frac{2}{m_\text{c} + m_\text{v}}$$

In the formula, L is the thickness of the inter-wall in the hole-net structure. Here, m_c is the effective mass beneath the valley of the conductor-band and m_v is that on the top of the valence-band.

Then the electrons are very rapidly caught into the interface state ($\Delta E_\text{c} = 2.1\ \text{eV} - 1.7\ \text{eV} = 0.4\ \text{eV}$) distributing in the region below the conduction band. These trap states have a certain distribution around the center state of 1.7 eV energy level. We think that the states distribution is probably related to the frequency distribution of the PL emission. Electrons in the interface state have long lifetimes, and population inversion between the top of the valence band and the interface state is thus possible from which we can explain the enhancing effect of the PL peak at 706 nm wavelength on sample A.

The enhancing effect of the PL emission on the irradiation region of sample B should originate from the denser matter of the rods condensed after cracking of the porous layer.

On sample C, the strap pieces dug up from the $\text{Si}_{1\text{-}x}\text{Ge}_x$ layer by laser irradiation can emit an intensive PL peak with a red-shift frequency obviously [Fig. 4 (c)] because of the narrower bulk gap of the germanium involved in the strap pieces.

It could be an important breakthrough for finding some special low-dimensional structures of silicon and germanium to improve the emission properties.

4 Conclusions

We use the laser irradiation method to make a pure silicon sample form a hole-net structure of silicon that can emit an intensive PL peak at 706 nm related to the quantum well of the inter-wall on the side of the hole drilled. A new three-level system associated with the crystal-oxide interface state has been proposed for interpreting the PL peak at 706 nm wavelength.

It is a novel method to combine laser irradiation with etching for preparing the low-dimensional structures of silicon and germanium. By controlling the preparation conditions, we can obtain some high quality samples in which the various low-dimensional structures have some intensive PL spectra. These findings could open a route to the fabrication of a silicon and germanium laser.

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