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Low-dimensional structures formed by irradiation of laser*

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

Keywords: low-dimensional nanostructures, photoluminescence, PL intensity, interface state
PACC: 4255R, 4270Q

1. Introduction

All manners 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] But it is difficult to explain some experimental results by using the quantum confinement model alone.

We can obtain some hole-net structure of silicon and porous structure of SiGe using the irradiation of laser, in which the intensity of PL peaks at 706 nm and 725 nm wavelength increases obviously. And the frequency of the PL peaks is not changeable basically in the different samples prepared under various conditions. The quantum confinement model is not enough to interpret the intensity-enhancing effect, for which we propose a new model of three-level system associated with the crystal–oxide interface state. These findings could lead a new way to some silicon and germanium based lasers.

2. Experiment

The samples used in the study were prepared as the following procedures (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 20s under a laser beam (spot diameter: 30 μm) with 1064 nm wavelength and 30W power.

(B) The above pure silicon samples were prepared by anodizing in HF(49%): Ethanol (1:1.5) under the irradiation of a laser beam (spot diameter: 700μm) with 532 nm wavelength and 20mW power for 15min. Anodizing current density is 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 epitaxial 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 irradiation of a laser beam (spot diameter: 700μm) with 532 nm wavelength and 20mW power for 5min, 10min, 15min and 30 min, respectively. Anodizing current density is about 40mA/cm^2.

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

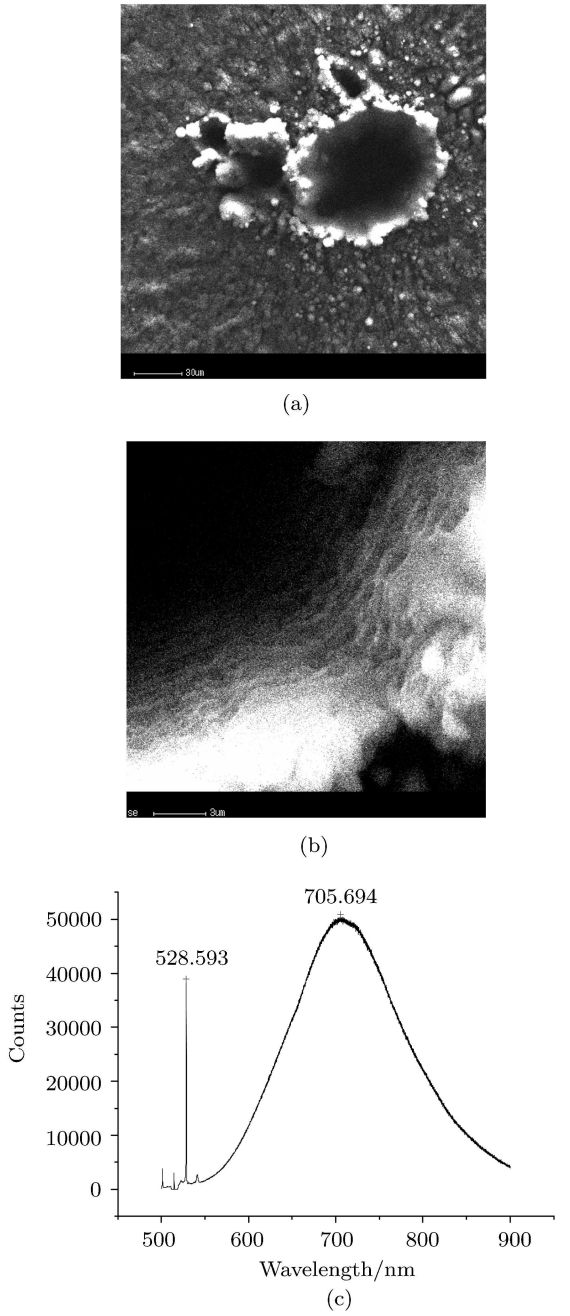


Fig.1. The results of preparing sample A: (a) the hole drilled by using a 30μm spot of the laser beam with 1064 nm wavelength and 30W power for 20 min; (b) the hole-net structure of silicon forming on side of the hole drilled by laser; (c) the PL peak at 706 nm wavelength related to the hole-net structure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

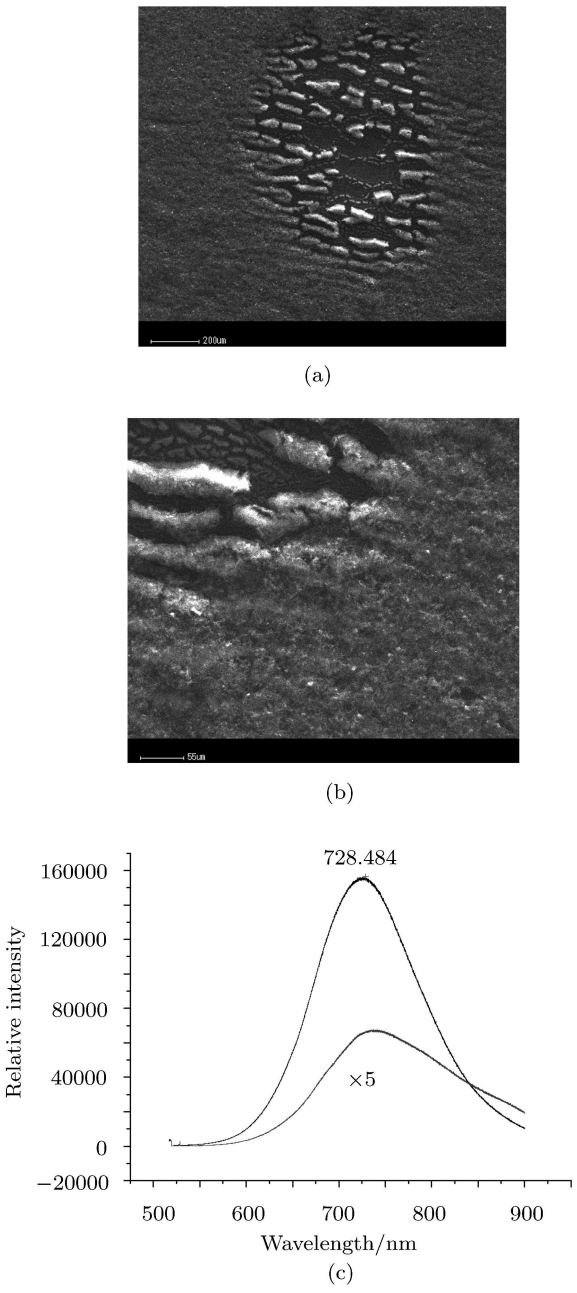


Fig.2. The results of preparing sample B: (a) anodizing the pure silicon with the irradiation of the laser beam for 15 min; (b) the boundary region of irradiating spot on the sample; (c) comparing the PL peaks emitted from the condensed rods in the irradiating and anodizing region of the sample B with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

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 30μm diameter with 1064 nm wavelength and 30W power hits on the pure silicon sample, a kind of the hole-net structure of silicon (Fig.1(b)) forms on the side of the hole (Fig.1(a)) drilled by 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 in the hole-net structure forms a quantum well whose thickness is about 10 nm. 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 almost no PL.

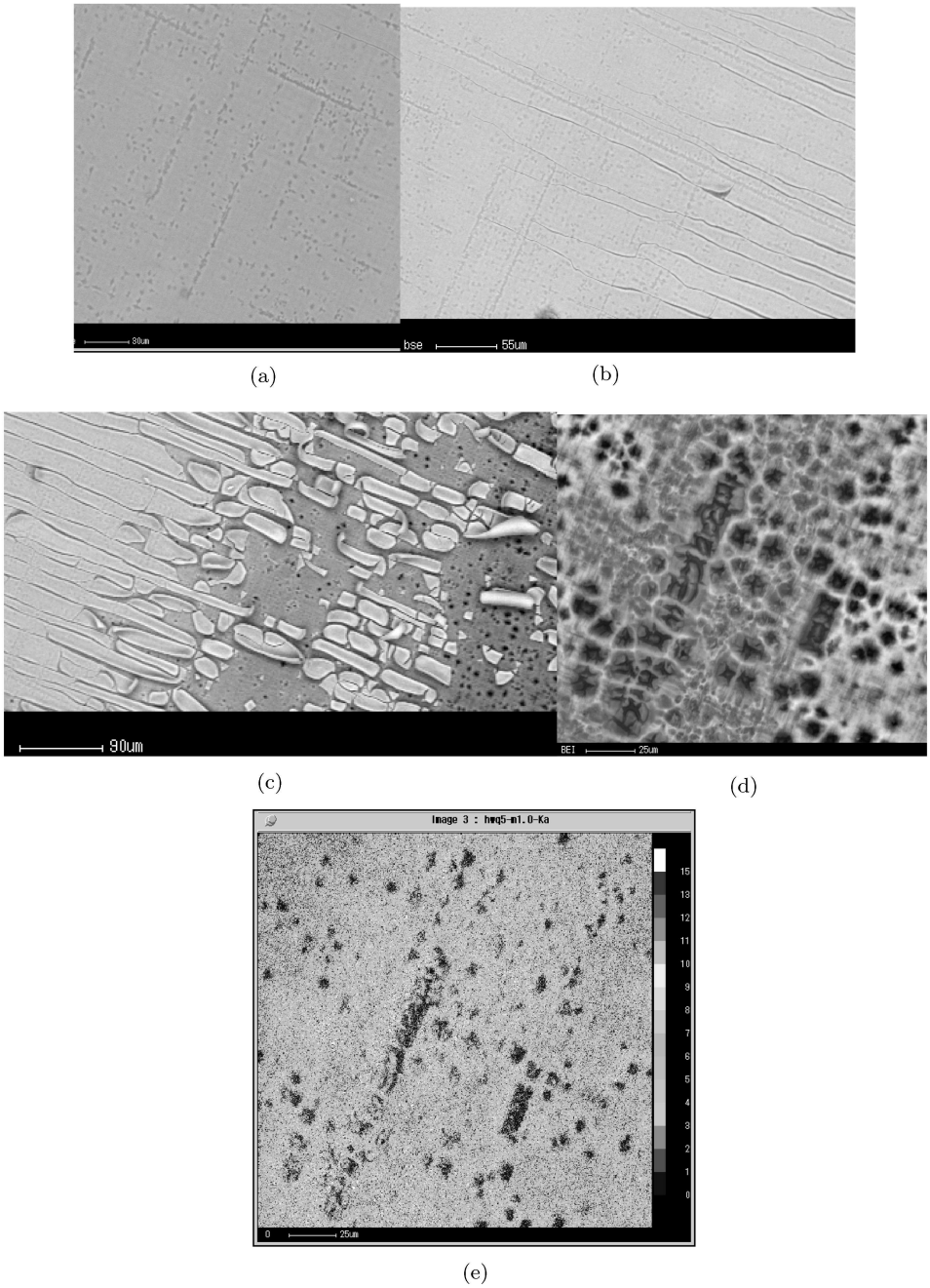


Fig.3. SEM Image of the low-dimensional structures of the C samples: (a) dot structure and (b) line structure on the $Si_{1-x}Ge_x$ film formed under a weaker irradiation of laser; (c) nano-strap pieces structure after digging up the $Si_{1-x}Ge_x$ layer with a stronger laser; (d) porous structure on the substrate under anodizing and irradiating for 30min, when the $Si_{1-x}Ge_x$ layer is almost dug out; (e) oxygen distribution diagram on the porous structure (Fig.3(d)).

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

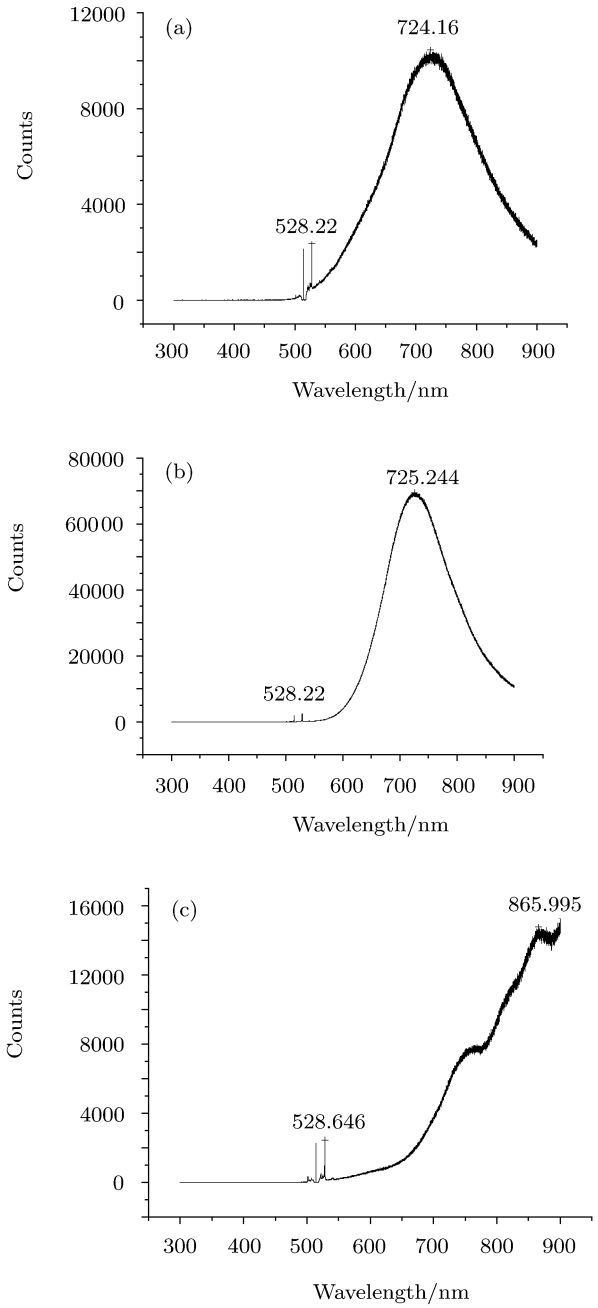


Fig.4. PL spectra of the low-dimensional structures of the C samples. (a) PL peak emitted from the less oxygen part of the porous structure (related to Figs.3(d) and 3(e)); (b) PL peaks emitted from the more oxygen part of the porous structure(related to Figs.3(d) and 3(e)); (c) PL spectra from the nano-strap pieces structure (Fig.3(c)).

Figure 3 shows the low-dimensional structures of the C samples using SEM. The part (a), (b), (c) and (d) in Fig.3 are the SEM images of the samples anodized and irradiated for 5min, 10min, 15min and 30min respectively. A weaker laser irradiation (beam spot diameter: 700μm) on the strained $Si_{1-x}Ge_x$ film can form some dots and lines structures as shown in Figs.3(a) and 3(b). A stronger irradiation of laser (beam spot diameter: 300μm) can dig up the $Si_{1-x}Ge_x$ film along 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 irradiating for 30min, 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 (Figs.4(a) and 4(b)).

3. Discussion

The preparing method of sample A is very easy to control and 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 side of the hole. The mechanism of the PL emission in hole-net nanostructure of silicon can be explained not only by the quantum confinement effect in the quantum well of the inter-wall but also by the interface state between the silicon crystal in the hole-net nanostructure and the oxide of the surface.

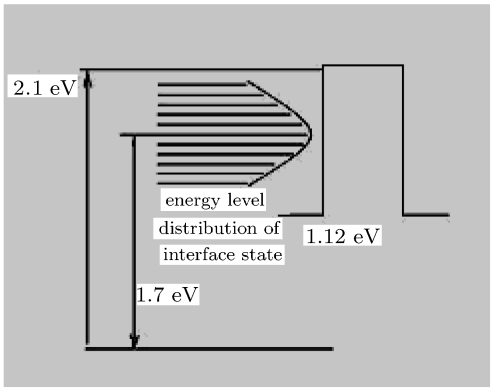


Fig.5. A new model of three-level system associated with the crystal–oxide interface state.

As shown in Fig.5, we propose a new model of three-level system associated with the crystal–oxide interface state for interpreting the PL peak at 706 nm

wavelength of sample A. An injection photon excites an electron in the top of valence band to jump up into 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 of silicon. It is a fast process for electron to jump from the level $E_{\rm v}$ to the level $E_{\rm c}(\Delta E = E_{\rm c} - E_{\rm v} = 1.12\text{eV} + 0.98$ eV (quantum confinement gap) = 2.1 eV (up edge of the PL band)). The quantum confinement gap can be calculated with the formula[11−13]

$$E_{\text{confined gap}} = E_{\text{bulk gap}} + (3h^2/8)(1/L^2)(2/(m_{\rm c}+m_{\rm v})),$$

where L is the thickness of the inter-wall in the hole-net structure of silicon, $m_{\rm c}$ is the effective mass beneath the valley of the conductor-band and $m_{\rm v}$ is the effective mass on the top of the valence-band.

Then the electrons are very rapidly caught into the interface state ($\Delta E_{\rm c} = 2.1$ eV – 1.7 eV = 0.4 eV) distributing in the region below the conduction band. These trap states have a certain distribution round the centre state of 1.7eV 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 the sample B should be originated from the denser matter of the rods condensed after cracking of the porous layer.

On the sample C, the strap pieces dug up from the $Si_{1-x}Ge_x$ layer by laser irradiating can emit an intensive PL peak with a red-shift frequency obviously (Fig.4(c)) for reasons of the less bulk gap of the germanium involved in the strap pieces.

May be it is the most important thing that some special low-dimensional structures of silicon and germanium could be found to improve the emission properties.

4. Conclusion

We use the laser irradiation method to make the pure silicon sample form the hole-net structure of silicon which can emit an intensive PL peak at 706 nm related to the quantum well of the inter-wall on side of the hole drilled. A new model of 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 irradiating with etching for preparing the low-dimensional structures of silicon and germanium. Through controlling preparing conditions, we can obtain some high quality samples in which the various low-dimensional structures have the intensive PL emission. These findings will open a route to the fabrication of a silicon and germanium laser.

References

[1] Tetelin C S, Wallart X and Nys J P 1998 *J. Appl. Phys.* **83** 2842

[2] Hellberg P E, Zhang S L and dHeurle F M 1998 *J. Appl. Phys.* **82** 5773

[3] Madsen J M, Cui Z J and Christos G Takoudis 2000 *J. Appl. Phys.* **87** 2046

[4] Qin G G, Liu X S and Ma S Y 1997 *Phys. Rev.* B **55** 12876

[5] Wolkin M V, Jorne J and Fauchet P M 1999 *Phys. Rev. Lett.* **82** 197

[6] Pavesi L, Dal Negro L and Mazzoleni C 2000 *Nature* **408** 440

[7] Huang W Q and Cai S H 2002 *Chin. Phys. Lett.* **19** 1657

[8] Li S S, Chang K and Xia J B 2005 *Phys. Rev.* B **71** 155301

[9] Li S S and Xia J B 2005 *Appl. Phys. Lett.* **87** 043 102

[10] Li S S, Xia J B and Hirose Kenji 2005 *J. Appl. Phys.* **98** 083704

[11] Huang W Q and Liu S R 2004 *Chin. Phys.* **13** 1163

[12] Huang W Q and Liu S R 2005 *Mod. Phys. Lett.* B **19** 1767

[13] Huang W Q and Liu S R 2006 *Chin. Phys.* **15** 390