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Activation of silicon quantum dots and coupling between the active centre and the defect state of the photonic crystal in a nanolaser

Activation of silicon quantum dots and coupling between the active centre and the defect state of the photonic crystal in a nanolaser*

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A new nanolaser concept using silicon quantum dots (QDs) is proposed. The conduction band opened by the quantum confinement effect gives the pumping levels. Localized states in the gap due to some surface bonds on Si QDs can be formed for the activation of emission. An inversion of population can be generated between the localized states and the valence band in a QD fabricated by using a nanosecond pulse laser. Coupling between the active centres formed by localized states and the defect states of the two-dimensional (2D) photonic crystal can be used to select the model in the nanolaser.

Keywords: nanolaser, Si quantum dots, localized states, photonic crystal

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1. Introduction

Light emission and amplification in silicon have focused on the use of silicon engineered materials such as nanocrystals. Experimental evidence of high intensity emission on silicon quantum dots (QDs) has aroused broad interest in the last decade.[1−8] Various experiments confirmed the following behaviours of Si QDs, observed at room temperature in different atmospheres: (i) wider and weaker photoluminescence (PL) emission appears in the wavelength range from the near infrared to the ultraviolet in pure hydrogen or in vacuum, and a blue-shifted PL peak is observed with nanocrystal size decreasing; (ii) after exposure to oxygen, which may not provide good passivation on a Si QD, stronger emission occurs in a narrower wavelength range.

In the present paper, we provide a new concept of Si QD nanolaser, which is prepared by using a second pulse laser. The state in the conduction band opened due to the QC effect forms a pumping level from which the electron is trapped into the localized state of surface bonds in the band gap. An inverse population can occur between the localized state in the gap and the top state of the valence band. The active centres formed by localized states of surface bonds such as Si=O and Si–O–Si bonds are very important. After exposure to oxygen or air atmosphere, which may not provide good passivation in small crystallites, localized electronic states can be formed in the gap to generate active centres where the stimulated emission can be generated. In general, various experiments have shown the wide PL intensity in the wavelength range of 500 nm–800 nm, which includes some stimulated emission and amplification spontaneous emission in Si QDs prepared in oxygen, nitrogen or air.[9,10] In our work, the goal is to make a narrow range Si QD sizes and obtain a sharp distribution of the localized states to form active centres to improve stimulated emission. A good way to do this is by preparing the Si QD using a nanosecond pulse laser in oxygen or air. Suitable annealing with the irradiation of a pulse laser is very important for obtaining active nanolaser materials.

In a general laser, a pair of flat mirrors is used to form a feedback cavity so that a laser mode can be selected and an intensive beam can be obtained.

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However, a nanolaser is built by using a Purcell cavity and a two-dimensional (2D) photonic crystal, without using a mirror.[11,12] We use a pulse laser to make a Purcell cavity and a 2D photonic crystal. The band of the 2D photonic crystal is opened because of the confinement of photonic states in which the gap of the photonic band becomes larger than the one in classical optics. Here the effect of enhancing and selecting the mode can be generated from the regime of cavity quantum electrodynamics and the coupling between the active centres and the defect states of photonic crystal.

It is noticed that the localized electronic states in the band gap come from the defects formed by some surface bonds on the Si QD, and similarly the localized photonic states in the photonic band gap come from 2D photonic crystal defects. This kind of the coupling has a selecting effect for the emission of a single quantum dot remarked by its size and its localized state, which is useful for quantum communication, the quantum information process, and quantum computing schemes in the future.

2. Experiment and results

We prepared the Si QD by using a pulse laser. The quantum dots were fabricated by using plasma produced from the interaction between silicon and a nanosecond laser pulse.[8,9] After irradiation, many Si QDs were prepared by using plasma and distributed on the wall of the cavity hit by the laser beam. Dangling bonds are formed on the surface of the Si QD. Passivation of the dangling bonds on the surface could be obtained by using different gas atoms at different atmospheres. Some kinds of surface bonds could not provide good passivation in small crystallites so some defect states remained there. Therefore there were more surface dangling bonds and defect states before annealing. After annealing, the range of QD sizes became narrower and the defect states decreased.

We designed and fabricated a kind of nanolaser in which oxidized silicon quantum dots can serve as an active laser material, and a 2D photonic crystal can be used as a feedback device to select the longitudinal laser mode. We used a pulse laser to fabricate the 2D photonic crystal. Figure 1(b) shows the structure of the nanolaser in which a 2D photonic crystal with an array of cavities is built on the silicon wafer and the active laser material made from Si QDs is on the wall of each cavity, as shown in Fig. 1(a). Under TEM, we can observe the QD structures embedded in silicon oxide (the concentration of QDs is about $10^{12}/\mathrm{cm}^2$) as shown in Fig. 1(a), which was fabricated by using a nanosecond pulse laser in oxygen and annealed at 1000 °C for 20 min.

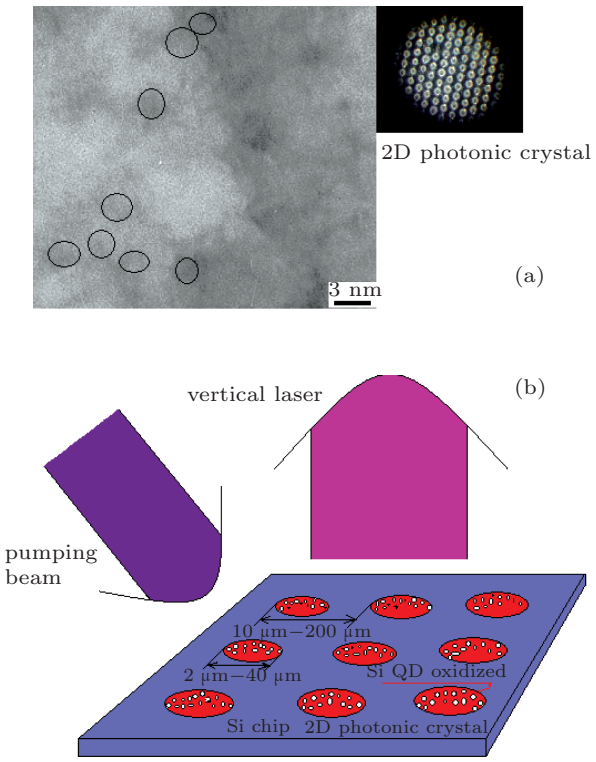

Fig. 1. (colour online) (a) TEM image of quantum dot structure embedded in silicon oxide and an image of a 2D photonic crystal fabricated by using a nanosecond pulse laser; (b) Nanolaser structure in all-silicon including the main components: a 2D photonic crystal with an array of cavities and active materials (an active QD set in every cavity).

As shown in Fig. 2(a), curve B describes the QC effect of Si QDs, curves C and E relate to the localized states in the gap due to the Si=O and Si–O–Si bonds, respectively, and curve D is due to the Si–N bond on the surface of the QDs. In Fig. 2(b), curve B describes the confinement effect of photonic QDs and curve C relates to the defect states in 2D photonic crystal where the coupling for the localized electronic states occurs.

Room-temperature PL measurement of samples was performed by exciting with the 514-nm line of a Ar^+ laser which has a perfect filter system. The PL signal was analysed using Renishaw micro-Raman systems. Figure 3 shows the peaks of the stimulated emission at 1.78 eV and 2.03 eV originating from the active centres due to Si=O and Si–O–Si bonds on the QD surface. A clear threshold transition from sub-threshold to super-linear evolution was measured with pump power increasing from 0.2 $\mathrm{mW}/\mu\mathrm{m}^2$ to 20 $\mathrm{mW}/\mu\mathrm{m}^2$ in the stimulated emission on a silicon QD prepared by using a nanosecond pulse laser in oxygen, and its optical gain was measured by using variable stripe length (VSL) method.

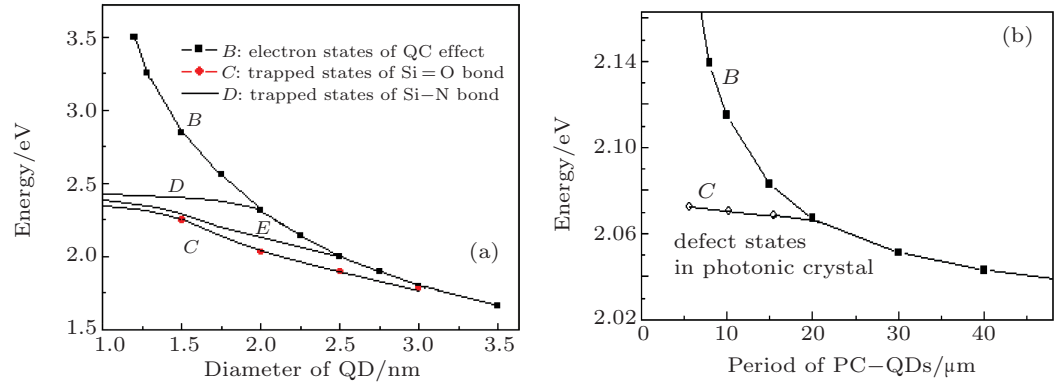

Fig. 2. (colour online) (a) Electronic states varying with size of Si QD, where curve B represents the relation between the energy of band gap opened and the size of QD, curve C the evolution of the localized level due to Si=O on surface, curve E the evolution of the localized level due to Si–O–Si on surface, curve D the evolution of the localized level due to Si–N on surface; (b) photonic states varying with period size of photonic crystal, where curve B denotes the relation between the energy state of perfect photonic crystal and its size, curve C is the evolution of the photonic crystal defect states.

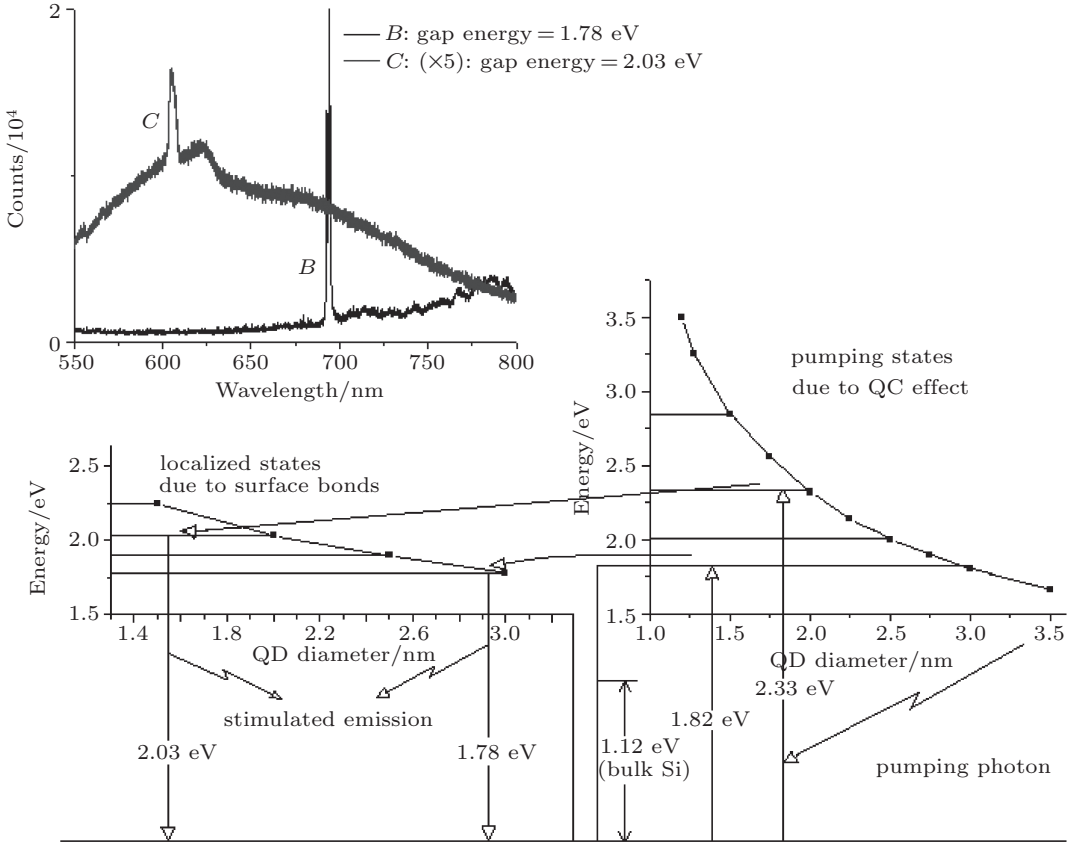

Fig. 3. The physical nanolaser process, where the electrons are first pumped into the states of the conduction band opened by the QC effect (on the right side), and then into smaller QDs, the electrons can tunnel into the localized states on the QD surface where the population inversion forms (on the left side) in which the peaks of stimulated emission are observed at about 600 nm and 700 nm.

3. Discussion

The physical nanolaser process is shown in Fig. 3. First, the electrons are pumped to the states of the conduction band opened by the QC effect as shown in the right panel of Fig. 3, and then in smaller QDs ($d < 3$ nm) the electrons can tunnel into the localized states in the band gap, forming the active centres due to Si=O or Si–O–Si on the surface of a QD where the population inversion can be formed as shown in the left panel of Fig. 3. In Fig. 2(a), it is clear that Si–H bonds have a good passivation on a QD surface

so that the change of the energy states depending on the size of QD could be consistent with the QC effect (B curve) which provides a pumping level. But some localized states due to Si=O, Si–O–Si or Si–N bonds on the surface will enter into the band gaps of small crystallites (C, E, and D curves) which provide some active nanolaser centres. Figure 2(a) also shows that the Si–N bond on the surface forms a localized state in the gaps on smaller Si QDs ($d < 2$ nm). These localized states each have a lower level than the bottom states (pumping level) of the conduction band in smaller QDs, such as the pumping level of 1.82 eV in the QD of 3 nm is higher than the localized state of 1.78 eV and the pumping level of 2.33 eV in the QD of 2 nm is higher than the localized state of 2.03 eV. The stimulated emission peaks at about 700 nm or 600 nm for the 514-nm excitation at 300 K can be observed obviously in Fig. 3 where the QD sizes concentrate to a narrower range near 3 nm or 2 nm respectively after suitable annealing treatment.

We have chosen some models in order to simulate the experimental process for Si QD structures prepared in different atmospheres. The electronic behaviour is investigated in the work by an *ab initio* non-relativistic quantum mechanical analysis. The density functional theory (DFT) calculation was carried out by using the local density approximation (LDA) and non-local gradient-corrected exchange–correlation functional (GGA) for the self-consistent total energy calculation. It is considered that both LDA and GGA underestimate the band gap for semiconductors and insulators. Figure 4 shows the Si QD structure and its density of states (DOS) for smaller nanocrystals in which we can make a comparison of DOS between the Si=O and Si–O–Si surface bonds. The two kinds of surface bonds can produce the localized states in gap as shown in Figs. 4(c) and 4(d). Figure 5 shows that the energy of Si=O bond is deeper than that of Si–O–Si bond, so the Si=O bond on the surface is more stable.

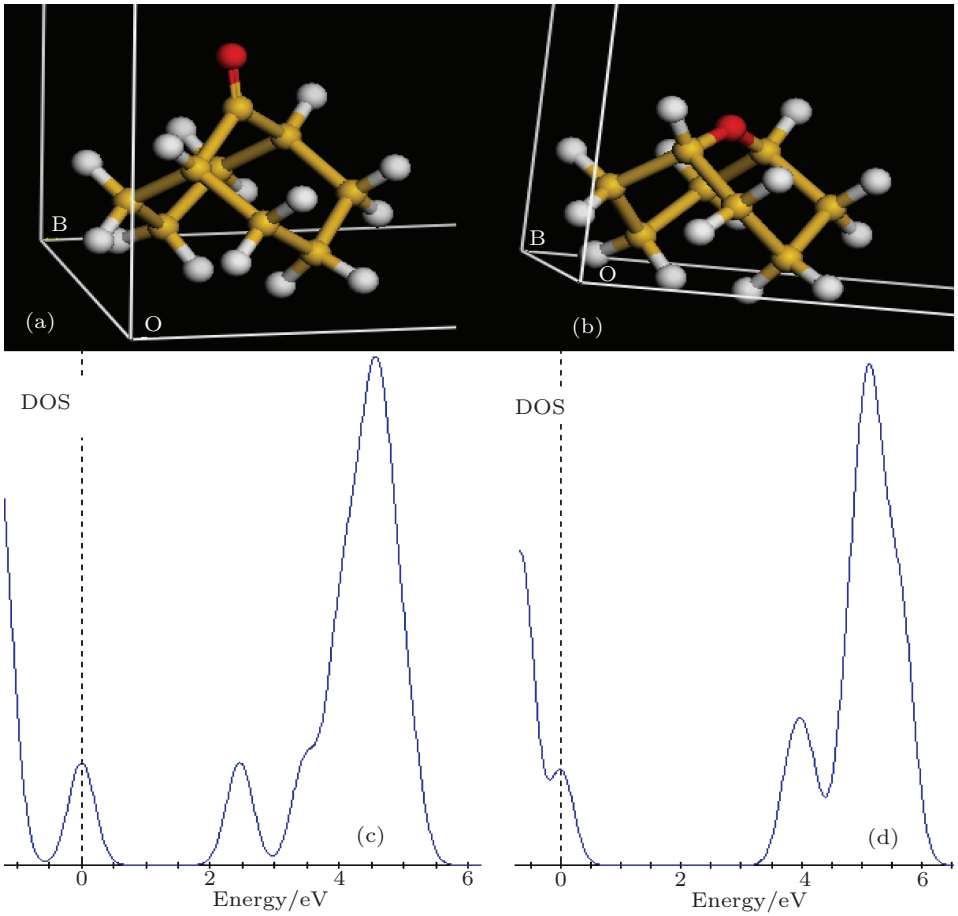

Fig. 4. (colour online) Silicon QD structures and their DOSs for smaller nanocrystals, in which we can make comparisons between (a) Si=O bond and (b) Si–O–Si bond on surface, and between (c) the DOS of the Si=O bond and (d) the DOS of the Si–O–Si bond.

In a two-dimensional photonic crystal with a Purcell cavity array, various modes can be selected and its emission can be enhanced. As is well known, the gap of a photonic band related to a larger period is very narrow so it is difficult to observe. However, because of the confinement effect in a photonic QD, a photonic band gap with a bigger period can be opened. We can explain the confinement effect of photonic QDs by using the following formula:

$$\Delta E = hc(1/n(1/(a-b)^2 + 1/b^2)^{1/2}),$$

where a is the period and b is the radius of the photonic QD. This is a negative QD made from a Purcell cavity. The $1/(a-b)^2$ in the formula is the confinement term.

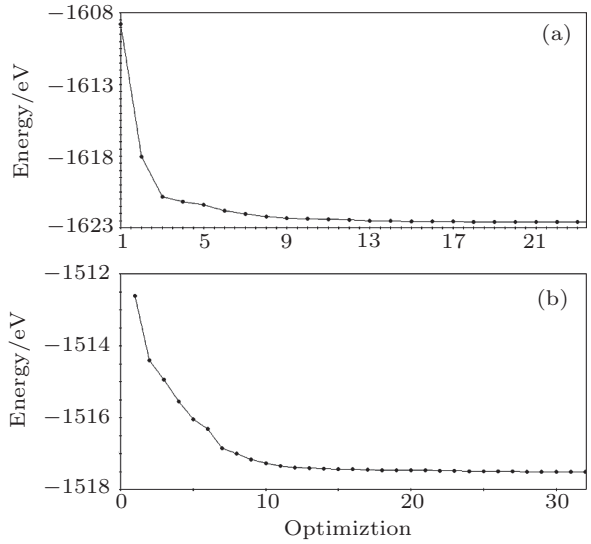

Fig. 5. Evolutions of the energy of (a) Si=O bond and (b) Si–O–Si bond on surface.

Generally speaking, the calculation indicates that a large blue-shift of the beam takes place with the decrease of period of the perfect 2D photonic crystal, which is in agreement with the behaviour of the B curve in Fig. 2(b). However, it is interesting that less blue-shift of the beam occurs under the coupling of the 2D photonic crystal with defects when the period of tge 2D photonic crystal decreases, which can be shown in curve C of Fig. 2(b). Comparing the localized electronic states in the band gap due to not having good passivation on the QD surface with the localized photonic states in the photonic band gap due to photonic crystal defects, we can understand their coupling deeply where the defect states of photonic crystal have the same energy levels as the localized electronic states have. Here, we can mark an Si QD with its size and the passivation bond of the surface as the single QD. Then the emission of the single QD can be selected by coupling the localized photonic states to it through adjusting the photonic crystal defects.

4. Conclusion

Overall, we discussed the mechanism of a nanolaser including several main components: quantum dots on all-silicon, active centres due to surface bonds, and 2D photonic crystal with an Purcell microcavity array. We fabricated the main structures of the nanolaser by using a nanosecond pulse laser. Evidence of the coupling between the localized states of surface bonds and the defect states of the 2D photonic crystal are clearly observed in samples. On the other hand, this kind of the coupling has a switch effect on the emission of an active centre in a single QD remarked by its size and its localized state, which is useful for quantum communication, quantum information processes, and quantum computing schemes in the future.

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