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Applied Surface Science 216 (2003) 419–423

applied
surface sciencewww.elsevier.com/locate/apsusc

Precise control of size and density of self-assembled Ge dot on Si(1 0 0) by carbon-induced strain-engineering

Y. Wakayama^{a,*}, L.V. Sokolov^b, N. Zakharov^c, P. Werner^c, U. Gösele^c^a*Nanomaterials Laboratory, National Institute for Materials Science, 2268-1 Shimo-shidami, Moriyama-ku, Nagoya 463-0003, Japan*^b*Institute of Semiconductor Physics, Russian Academy of Science, Lavrentiynva 13, 630090 Novosibirsk, Russia*^c*Max-Planck Institute of Microstructure Physics, Weinberg 2, Halle 06120, Germany*

Abstract

In order to produce dome-shaped Ge dots with small size and high density, C submonolayers (C-SMLs) were incorporated at the interface between Ge wetting layers and Ge dots. The C atoms are considered to induce a local strain field by forming Ge–C bonding. Such strain field enhanced dome formation even at low temperature (<500 °C). Optimization of experimental conditions enabled precise control of the Ge dome size in the range of 30–40 nm with the density of 10^{10} cm^{-2} . The Ge domes thus prepared exhibited intensive photoluminescence (PL) compared to those prepared by a conventional self-assembling technique.

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Keywords: Si; Ge; Quantum dot; Strain-engineering; Photoluminescence

1. Introduction

Fabrication of nanostructures in the Si–Ge–C system has been investigated to integrate quantum functional devices into the Si-based electronics. Recently, many efforts have been made to develop light emitters [1–5], photodetectors [6,7] and a resonant tunneling diode [8]. Well-controlled fine structures on nanometer scale are strictly required to develop the device performance. However, unsolved issues still remain to improve the quality of nanoscale structures. One of such issues is to control structures of Ge islands grown on a Si substrate. Three-dimensional (3D) growth is driven by elastic energy caused by the lattice mismatch between Ge and Si. Then, different structures, which are hut, pyramid

and dome, are simultaneously grown as shown in Fig. 1(a). Such a complicated feature makes it difficult to control structures and properties of the Ge dots precisely. The main purpose of this study is to establish a control technique of nanostructures of self-assembled Ge dot. In particular, we focused our study on the dome-shaped Ge dot because they showed excellent homogeneous size distribution. Here, a small amount of C atoms incorporated into Ge play a key role. We found that a C submonolayer (C-SML) deposited at the interface between a Ge wetting layer and 3D islands caused dome formation even at a low temperature by inducing a lattice strain [9,10]. For further improvement of size controllability, experimental conditions including the temperature and C-SML thickness were optimized. As a result, very small Ge domes were successfully fabricated in the diameter range of 30–40 nm at 5 nm^2 each, which showed intensive photoluminescence (PL) around 0.90–0.95 eV.

* Corresponding author. Tel.: +81-52-736-6011;

fax: +81-52-736-6012.

E-mail address: wakayama.yutaka@nims.go.jp (Y. Wakayama).

Fig. 1. (a) AFM image of typical Ge dots on Si(1 0 0). Different structures, hut, pyramid and dome are grown simultaneously. (b) Illustration of each structure. Substrate temperature is main factor to determine dot structure. (c) Cross-sectional TEM image of a Ge dome.

2. Experiments

All samples were prepared by an ultrahigh vacuum molecular beam epitaxy (UHV-MBE) machine. All of the samples were prepared as follows. First, Ge of 4.2 ML thickness was deposited at 500 °C. This forms a two-dimensionally-flat layer, which is called a wetting layer (WL). Subsequently, a small amount of C atoms, of which thickness is less than one monolayer, were deposited on the WL at 500 °C. Finally, a Ge top layer of 2.1 ML thickness was deposited for 3D dot formation. The substrate temperature only for the top layer was changed in the range of 380–530 °C to control the dot size, while those for the Ge-WL and C-SML were

kept at 500 °C. Preparation procedure of the samples was described more detail in elsewhere [9,10]. Structures of the samples were examined by atomic force microscopy (AFM) and transmission electron microscopy (TEM). To evaluate an optical property, PL spectra were measured at 4 K. Ar⁺ laser was used as an excitation source. The PL spectra were obtained by a liquid nitrogen-cooled InGaAs detector using a standard lock-in technique.

3. Results and discussion

Fig. 1(a) shows an AFM image of typical Ge dots (without C-SML) grown on the Si(1 0 0) substrate. So-called hut, pyramid and dome structures coexist simultaneously. The main parameter to determine these structures is a substrate temperature during deposition as illustrated in Fig. 1(b). High temperature around 600 °C is required to form dome structure, meanwhile hut structure is dominant at relatively low temperature below 500 °C. In these structures, dome has an advantage for a quantum dot due to high size uniformity. Large vertical size is also another merit of the dome. This is because that an increase in confinement energy of holes in a valence band can be expected. Typical TEM image of the dome is shown in Fig. 1(c). However, high temperature results in a large size over 100 nm. We need, therefore, to solve a contradict issue; to grow domes at low temperature.

In general, the dome structure prepared by conventional process has large size (>80 nm) as can be seen in Fig. 2(a). Here, the total thickness of Ge was 6.3 ML. The substrate temperature of 580 °C was necessary, in

Fig. 2. AFM images of the Ge dots with dome structure. Scan area is 1 μm². (a) Domes produced by conventional self-assemble process. (b) Domes produced by a C submonolayer. Downsizing and increase in density were achieved.

order to obtain the size uniformity instead of small size and high density. In contrast, C-SMLs were found to promote the dome formation [9,10] even at lower temperature. Here, the samples were prepared by a three-step process: (i) Ge-WL on Si, (ii) C-SML on Ge-WL and (iii) Ge dot on C-SML. The thicknesses of each layer are 2.1 ML (Ge-WL), 0.03 ML (C-SML) and 4.2 ML (Ge dot), respectively, i.e. total thickness of Ge was identical to that of Fig. 2(a). All layers were deposited at 500 °C. Fig. 2(b) shows the Ge dots thus prepared. Obviously, both a decrease in diameter (35 nm) and an increase in density ($\sim 1 \times 10^{10} \text{ cm}^{-2}$) were achieved. Then, the homogeneous size distribution was maintained. We consider that a local strain field was induced by the Ge–C bonding at the WL/dot interface. Such strain field acts to reduce the interface area, enhancing dome formation even at lower temperature. In the meantime, temperature is main parameter to determine dot size as mentioned above. Namely, decrease in temperature led to reduction in the dot size. Hence, it can be concluded that combination of low temperature deposition and C-SML is an effective technique for small dome formation with the high density.

With a view to control the dot size more precisely, the substrate temperature in the third process, T_{dot} , was modified. AFM images and their size distribution were shown in Fig. 3. All samples consist of three layers: Ge-WL, C-SML and Ge dot. Ge-WL and C-SML were deposited at 500 °C, while T_{dot} was changed to (a) 530 °C and (c) 470 °C, respectively. The AFM image of $T_{\text{dot}} = 500$ °C is also shown in Fig. 3(b) for comparison. As can be seen here, slight change in T_{dot} allows precise size control in the range of 30–40 nm at 5 nm each.

The C-SML thickness and T_{dot} were changed in wide range to obtain a phase diagram of Ge dot structure. Various structures were observed as shown in Fig. 4. Dome structure was formed only in limited conditions. That is, the thickness of C-SML should be increased with decreasing T_{dot} . An irregular structure was observed in addition to small domes at the low temperature range around 400 °C where indicated by a dashed circle in Fig. 4. Here, the irregular structure has no specific facets or orientation. Cross-sectional TEM observation in Fig. 4(b) revealed that ultra small domes were formed, of which diameter is as small as 15 nm, in this temperature range. Kinetic limitation due to low

Fig. 3. AFM images of the Ge dots produced by C-SMLs and their size distribution: (a) $T_{\text{dot}} = 530$ °C, C-SML, 0.03 ML; (b) $T_{\text{dot}} = 500$ °C, C-SML, 0.03 ML; (c) $T_{\text{dot}} = 470$ °C, C-SML, 0.04 ML. Size of Ge domes was well controlled by adjusting temperature of Ge dot deposition, T_{dot} , and thickness of C-SML. The average diameters, d_a , were about 40, 35 and 30 nm, respectively.

T_{dot} caused such small, but high-density and irregular dot growth. A high density in the order of $1 \times 10^{11} \text{ cm}^{-2}$ and an average dot size of 20 nm were achieved in return for the size uniformity as shown in Fig. 5.

When the thickness of C-SMLs were excessive, so-called super domes were formed as shown in Fig. 4(c). Super domes have large size, which are produced through an elastic relaxation by introducing stacking faults and other defects. Large amount of C atoms may prevent coherent Ge growth. Otherwise, high mobility of Ge atoms owing to high T_{dot} is another possible

Fig. 4. Phase diagram of Ge dot structure. Various kinds of structures, which are (a) irregular structure, (b) ultra small dome, (c) super dome, (d) hut/dome and (e) small dome, are grown depending on T_{dot} and thickness of C-SML. In the dashed circle, high density was attained at the expense of size uniformity. Well-defined small domes were obtained in the gray area.

reason for the super dome formation. Hut and dome coexist, on the other hand, when the coverage of C-SML is insufficient as can be seen in Fig. 4(d). Well-defined dome structure was obtained only in the

range of $T_{\text{dot}} = 450\text{--}550\text{ }^{\circ}\text{C}$ and C-SML = 0.03–0.04 ML, where the gray area in Fig. 4.

The photoluminescence spectra were measured for evaluating optical property of these Ge dots. For this purpose, Si cap layers with 40 nm thickness were deposited on the Ge dots at 500 $^{\circ}\text{C}$. The PL spectra were obtained from Ge dots shown in Fig. 2, that is, the conventional domes grown at 580 $^{\circ}\text{C}$ and the small domes grown at 500 $^{\circ}\text{C}$ with the C-SML. The PL spectrum from the conventional Ge domes is shown in Fig. 6(a). A strong peak from the Si substrate and cap layer can be seen at 1.10 eV. A weak peak around 1.03 eV is attributed to a carrier recombination in the Ge-WL. However, this spectrum indicates no clear PL signal from 3D dots. In Fig. 6(b), the PL spectrum obtained from small domes is shown. Broad peak around 0.90–0.95 eV is originating from the 3D dots. The broad peak from 3D dot basically consists of two components those are no-phonon (NP) and transverse

Fig. 5. Average size (■) and density (●) of Ge domes as a function of the temperature of Ge dot deposition.

Fig. 6. Photoluminescence spectra from Ge domes. (a) Ge domes ($d_a = 80$ nm) produced by conventional self-assembling process. (b) Ge domes ($d_a = 35$ nm) produced by C-SML.

optical (TO) phonon-assisted transition, although the peak in Fig. 6(b) can not be resolved in detail. The intensity of both spectra was normalized by the peaks at 1.10 eV from Si for accurate comparison. As clearly seen, the small dots with C-SML indicate intensive light emission; the integral intensity is more than 10 times intensive compared to the conventional one. A single layer of the dome-shaped Ge dot has been reported to show only a weak PL signal and multiple stacked Ge dot layers are necessary to get higher PL intensity [11,12]. Whereas, it is worth noting that the drastic increase in the PL intensity was realized even in the single dot layer by the C-SML effect. We consider that the small size contributes carrier localization, enhancing recombination probability. Additionally, high density of 3D dots is also responsible to intensive light emission.

4. Conclusion

We have investigated a new technique for the self-assembled Ge dot growth. Then, the incorporation of

C-SMLs at the Ge-WL/Ge dot interface was a key technique. This technique is based on an idea of strain-engineering, where lattice strain induced by the C atoms drastically affect on the dot morphology. In our study, the C-SMLs were found to bring about dome formation even at 500 °C, attaining downsizing of the domes. Furthermore, precise adjustment of the experimental conditions enabled a fine-tuning of dome size particularly in the range of 30–40 nm. The Ge dots thus prepared demonstrated the intensive light emission around 0.90–0.95 eV (1.31–1.38 μm). This result suggests a potentiality of optical functional devices in the Si–Ge–C system.

Acknowledgements

The authors would like to express sincere thanks to Mr. K. Kurihara of Tokyo University and Mr. T. Tateno of Tokai University for a technical support of the photoluminescence measurement.

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