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Modification of the growth mode of Ge on Si by buried Ge islands

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Photoluminescence experiments on double Ge layers were performed to give deep insights on the growth mode of Ge on Si in the presence of buried 4.5 monolayers of Ge islands. The critical coverage of the island formation and the wetting layer thickness were confirmed to be reduced in the second Ge layer. In addition, a drastic increase of the island density as well as a shape transition were observed by atomic force microscopy. These modifications of the growth mode are explained in terms of the surface strain induced by the buried Ge islands and the reduction of the nucleation barrier due to the alloying. © *2000 American Institute of Physics.* [S0003-6951(00)03725-6]

Self-assembled Ge islands on Si(100) have been intensively investigated as the basis of future electronic and optical devices.[1–6] The island is known to develop during epitaxial growth in the Stranski–Krastanow growth mode; where beyond a critical thickness, a nucleation of islands on the two-dimensional wetting layer becomes energetically favorable since the gain of strain energy due to the partial strain relaxation overcompensates the increased surface energy. Owing to the simple *in situ* growth approach without lithography, the self-assembly is now regarded as an attractive and promising route to the fabrication of the small islands which might act as quantum dots. However, to establish a method to achieve sufficiently uniform island sizes with regular spatial distribution still remains a critical issue. This should be solved since a well defined size with little dispersion is generally required for any practical applications.

As a possible way to obtain the size uniformity, Tersoff *et al.* have pointed out that the growth of multilayer arrays of coherently strained islands is useful.[7] Based on a simple model of the surface strain, the arrangement of islands was predicted to become more uniform with stacking the islands regardless the initial condition, and atomic force microscope (AFM) images of SiGe/Si superlattices were given as experimental evidence.

In this letter, we report on the growth mode of Ge on Si(100) with buried Ge islands to give deep insight on the initial stage of stacking process. By using photoluminescence (PL) spectroscopy, we demonstrate that the critical coverage of the island formation and the wetting layer thickness are reduced in the second Ge layer. These changes are shown to be accompanied by morphological changes such as a drastic increase of the island density and a shape transition.

The samples were grown by gas-source molecular beam epitaxy (MBE) (Daido-Hoxan VCE S2020) using disilane (Si_2H_6) and germane (GeH_4) as source gases at fixed growth temperature of 700 °C. The substrates were *p*-type Si(100) with resistivity of 10–20 Ω cm. The samples contain double Ge layers separated by a 144 Å Si spacer layer. The Ge coverage of the first layer was fixed to be 4.5 monolayers (ML), which is larger than the critical coverage of the island formation of 3.7 ML.[1] Therefore, three-dimensional Ge islands with leaving 3.0 ML of the wetting layer are expected in the first layer. The coverage of the second Ge layer was systematically changed from 0.6 to 9.0 ML. As a sensitive tool to investigate the growth mode, PL spectroscopy was performed. In Ge/Si system, PL spectroscopy has been already demonstrated to be a powerful tool to monitor the growth mode changeover to the island formation at high growth temperatures[1,2] as well as the different island phases such as domes, pyramids, and hut clusters grown at low temperatures.[8] The samples for PL measurements were capped with 300 nm of Si. PL spectra were recorded using a standard lock-in technique with a liquid-nitrogen-cooled Ge photodetector (North-Coast EO-817L). The samples were mounted on a cold finger of a closed-cycle cryostat. An argon ion laser was used as an excitation source. For some samples, AFM measurements were performed *ex situ* by interrupting the epitaxial growth at heterointerfaces.

Figure 1 shows the PL results of a series of samples where the Ge coverage of the second layer was systemati-

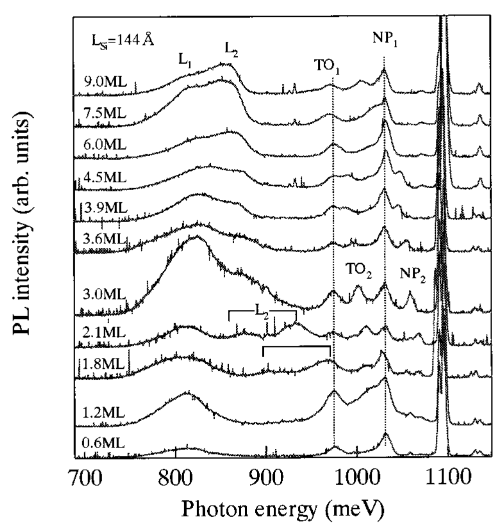

FIG. 1. PL spectra of a series of samples where the Ge coverage of the second layer was systematically changed while that of the first layer and the Si spacer thickness were fixed at 4.5 ML and 144 Å, respectively.

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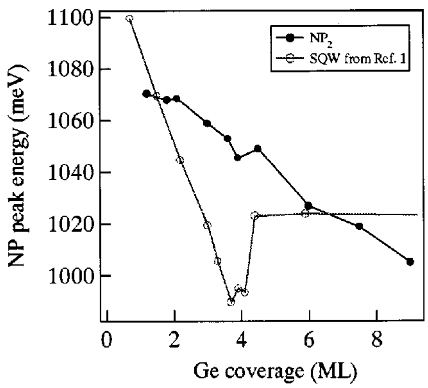

FIG. 2. PL peak energies from the wetting layer (solid circles), NP_2, as a function of the Ge coverage of the second layer. For comparison, the NP energies of a single Ge layer (open circles) in Si are also plotted as a function of the Ge coverage.

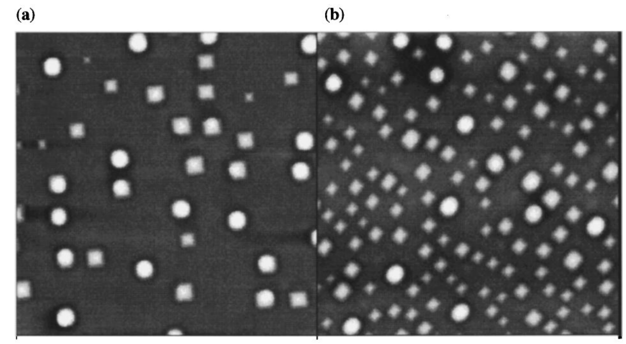

FIG. 3. 2 μm$\times$2 μm AFM images of (a) the first Ge layer and (b) the second Ge layer separated by a 144 Å Si layer. It is noted that the Ge coverage is 4.5 ML for the both samples.

cally changed measured at around 25 K. The PL lines at around 1130 and 1095 meV originate from the Si substrates. At lower energies of the Si-related peaks, several additional lines can be identified. Based on our previous publication where we investigated PL from Ge embedded in Si,[1] we assigned these peaks as no phonon (NP) and transverse optical (TO) phonon lines from the wetting layer and PL from the Ge islands (L). PL from the wetting layer is clearly seen to consist of two (NP,TO) pairs. The pair at lower energies labeled NP_1 and TO_1 is considered to come from the first Ge layer since they show no peak shift with changing Ge coverage of the second layer. This confirms that the electronic coupling between the wetting layers of double Ge is negligible, showing that PL spectral feature related with the wetting layer of the second Ge layer reflects the modification of the growth mode. PL from the Ge islands (L) are also divided into two components; L_1 and L_2. A striking feature is that L_2, which seems to be related with the second Ge layer, can be clearly identified at 1.8 ML. This amount is much smaller than the critical coverage of the island formation of a single Ge layer, showing that the critical coverage of the island formation is drastically decreased in the presence of the buried Ge islands.

In Fig. 2, the PL peak energies from the second wetting layer, NP_2, are plotted against the Ge coverage of the second layer. For comparison, the NP energies of a single Ge layer in Si are also plotted as a function of the Ge coverage. The energy dip at 3.7 ML and following blueshift was previously interpreted as evidence for the island formation.[1,2] That is, after the nucleation of the islands, Ge atoms at the topmost layer are considered to be incorporated to the islands, resulting in the reduction of the wetting layer thickness. This mechanism is not directly applied to the stacked Ge islands since NP_2 continuously shifts to lower energies after the appearance of L_2 at 1.8 ML. This suggests that a part of additional Ge atoms contribute to the increase of the wetting layer thickness even after the nucleation of the islands takes place.

At an intermediate Ge coverage, NP_2 lies at higher energies than NP of the single Ge layer, showing that the wetting layer thickness of the second layer is reduced. This result is consistent with the recent report of Schmidt and Eberl[9] where they clarified the reduction of the wetting layer thickness in a five-fold stack of 6.5 ML of Ge. On the other hand, this energy relationship is seen to be broken at lower and higher Ge coverages. It should be noted that NP_2 does not reach the band gap of unstrained Si even if the Ge coverage is extrapolated to 0. This would be explained by the reduction of the band gap of the Si spacer layer due to the strain. In other words, this supports that the modification of the growth mode of the second Ge layer is brought by the strain induced by the buried Ge islands.

As shown in Fig. 1, PL from the islands, L_1 and L_2, can be clearly resolved even if the Ge coverage of each layer is same. AFM observations revealed that the PL feature from the islands is closely related to morphological changes. Figure 3 shows 2 μm$\times$2 μm AFM images of (a) the first Ge layer and (b) the second Ge layer separated by a 144 Å Si layer. It is seen that the density of the islands in the second layer is drastically increased and about four times larger than that in the first layer. As a result, the averaged volume of the island in the second layer is much smaller than that in the first layer. This would be the reason for the blueshift of L_2 compared to L_1 and the dominance of ''pyramids'' which are known to be stable phases for smaller volume.[10]

It is noted that the total volume of the islands in the second layer estimated from Fig. 3 showed only 8% increase compared with that in the first layer, which could be compensated with the decrease of the wetting layer thickness. Therefore, the growth rate of Ge seems to be not greatly affected by the presence of the buried islands. The small amount of excess Ge atoms for the islands formation is unlike to bring the observed drastic increase of the island density. The observed growth mode could be explained in terms of the surface strain induced by the buried Ge islands. It is well known that the Si lattice on top of the islands becomes an energetically preferable site for the successive Ge growth due to smaller lattice mismatch, resulting in the vertical ordering of the islands.[11] In fact, the vertical ordering of ''domes'' were observed in our samples by cross-sectional transmission electron microscopy. The additional appearance of pyramids would be due to the interaction of the strain induced by neighboring islands, leading to the formation of the preferable sites not only on top of the islands but also between the islands.[7] Another possibility is the reduction of the nucleation barrier in the second layer since Ge atoms segregating to the growth front is predicted to drastically reduce it due to the contribution of the free energy of entropy mixing.[12] The interface mixing due to the Ge surface segregation is less probable since gas-source MBE was used for the sample growth where is suppressed by the hydrogen.[13] However, an alloying during the capping process[14] must be considered since the averaged height of domes is around 200 Å, which is even larger than that of the spacer thickness of 144 Å. In fact, a preliminary experiment showed that to cap Ge islands with a thin Si layer brings enhanced nucleation.[15] Further study to clarify the mechanism of the growth mode modification is in progress.

In summary, we investigated the growth mode of Ge on Si(100) with buried Ge islands by using PL spectroscopy and AFM. The critical coverage of the island formation and the wetting layer thickness were found to be reduced in the presence of the buried Ge islands. The morphological changes such as a drastic increase of the island density and a shape transition were also observed.

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