Temperature effect on the formation of uniform self-assembled Ge dots

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The effect of the growth temperature on the formation of uniform self-assembled Ge dots on Si (001) substrates was studied. The ratio of pyramid dots to dome dots varies with the growth temperature from 500 to 700 °C. Temperature of 600 °C was optimum to form uniform self-assembled Ge dots, and is attributed to the enhanced diffusion kinetics. Highly uniform Ge dots with height deviation of ±3% were obtained at this growth temperature. Discontinuity in characteristic length was found in an Arrhenius plot between 600 and 625 °C, and it is due to intermixing of Si with Ge which occurred at high temperature. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1616978]

Self-assembled growth of coherent and strained Ge dots on Si (001) has attracted extensive interest in their fundamental properties and potential applications in nanoelectronics and optoelectronics. Previous studies showed that self-assembled Ge dots on Si (001) had a bimodal size distribution, i.e., square-based pyramids with four {105} facets and polygon-based domes with multifacets.[1] For pragmatic applications, one essential issue is how to realize uniform self-assembled dots with monomodal distribution (the dots have similar morphology and a narrow size distribution).

Recently, we have observed the formation of a *mono*modal distribution of Ge dots on Si stripe mesas formed via selective epitaxial growth on patterned substrates.[2] A similar observation of the monomodal distribution of Ge dots on high-index facets has been also reported.[3] Monomodal dome dots were observed when Kamins *et al.* studied the Ge thickness effect on self-assembled dots.[4] Ross *et al.* reported a narrow size distribution of Ge dots on planar Si (001) substrates.[5] These results suggest that it may be possible to realize uniform Ge dots on planar Si (001) substrates.

In this letter, we systematically study the effect of growth temperature on the formation of uniform self-assembled Ge dots grown on Si (001) substrates. Atomic force microscopic (AFM) results show that the bimodal distribution changes with the growth temperature and an optimum growth temperature is about 600 °C for the formation of monomodal Ge dots. Both an Arrhenius plot of the characteristic length and the number ratio of bimodal dots show a transition at 600 °C, suggesting a change of mechanism from enhanced diffusion to the intermixing of Si with Ge at higher temperature.

The substrates were commercially available p-Si (001) wafers. Growth was carried out in a gas source molecular beam epitaxial (GSMBE) system with a Si_2H_6 gas source and a Ge Knudsen cell source. Samples were first chemically cleaned and then dipped in a diluted HF solution to form a hydrogen-terminated surface before being loaded into the vacuum chamber. After *in situ* thermal cleaning at 900 °C, a Si buffer layer (~100 nm) was grown at 660 °C with a growth rate of about 0.1 nm/s. The temperature was measured with a calibrated pyrometer and a thermal coupler back contacted to the sample. After Si growth, Ge was deposited at various growth temperatures from 500 to 700 °C with a deposition rate of about 0.01 nm/s. All samples have a nominal Ge layer thickness of 1.6 nm. The samples were then *ex situ* characterized with an AFM in contact mode, which is preferred for characterizing small quantum dots. The typical scanning direction was parallel to an ⟨110⟩ direction of the Si (001) substrates. The scan frequency was between 0.5 and 1.0 Hz.

Figure 1 shows AFM images of the self-assembled Ge dots at various growth temperatures from 500 to 700 °C. In Fig. 1(a) with growth temperature of 500 °C, one can clearly see that there are two kinds of Ge dots, pyramids and domes. This is the typical bimodal distribution frequently observed. As the growth temperature increases, the number ratio of pyramids to domes decreases as plotted in Fig. 2 and finally approaches a very low value at 600 °C, leading to the formation of a monomodal morphology distribution. As the temperature further increases to 650 and 700 °C [Figs. 1(d) and 1(e), respectively], the pyramids reappear and are larger in size, and there are also some very small dots, whose height is in the range of 2–3 nm. The very small dots were thought to be precursors of the larger dots in Ref. 6. The difference is that the precursors are only observed at higher temperature in our case. The appearance of the small precursors is related to the change of strain distribution on the surface because of Ge/Si intermixing, which will be discussed in the following. Figure 1(f) shows a three-dimensional view of the sample grown at 600 °C. The uniform self-assembled dots are dome shaped with base diameter and height of about 70 and 15 nm, respectively. The areal density of the dots is about 3

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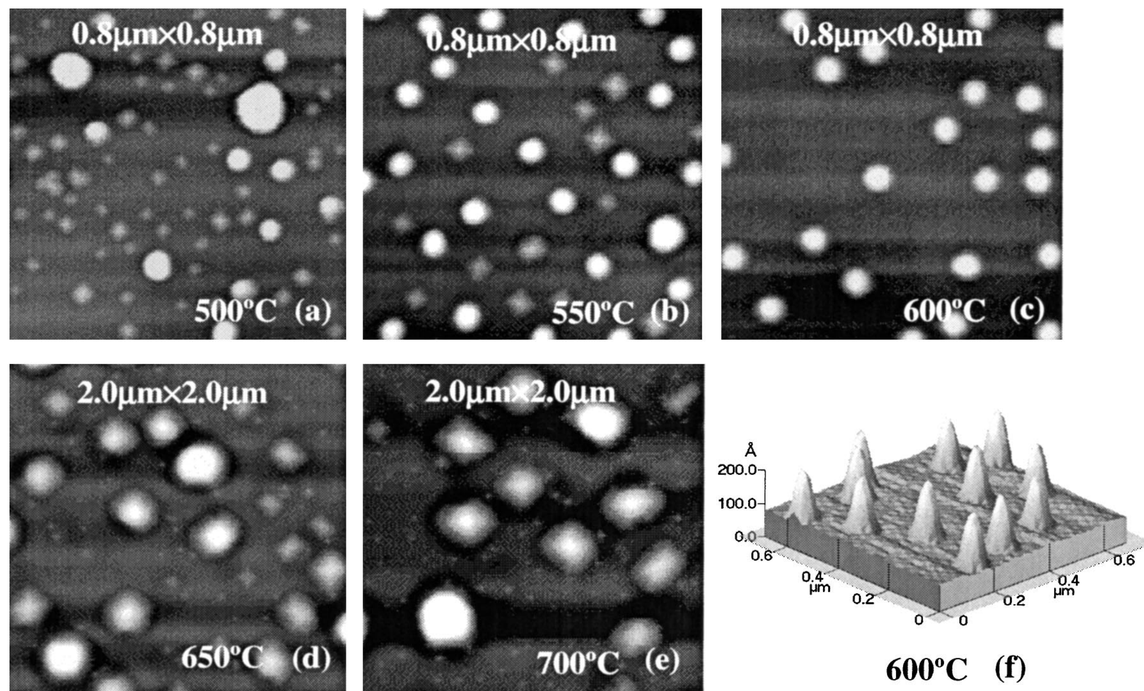

FIG. 1. Two-dimensional AFM images of Ge dots on Si (001) at various growth temperatures: T_s=(a) 500, (b) 550, (c) 600, (d) 650, and (e) 700 °C. (f) Three-dimensional AFM image of the sample grown at 600 °C. Uniform dots are evident.

$\times 10^9$ cm^{-2} and the deviation in height of the dots is about $\pm 3\%$. This result has demonstrated the possibility of forming uniform self-assembled Ge dots on Si (001) by controlling the substrate temperature.

The formation of uniform dots at 600 °C arises from enhanced diffusion. One experimental way to prove the Ge diffusion process is to study the dot morphology evolution at different postgrowth holding times, that is, the time interval between the finish of Ge growth and the drop in substrate temperature. Figure 3 gives the dot density as a function of the holding time. The growth temperature was kept at 575 °C for this set of samples. This is akin to an *in situ* annealing process. The results indicate that the change in density slows down with an increase in holding time, consistent with Kamins *et al.*'s annealing study.[1] If Ostwald ripening is dominant in the process, the size of the dots would linearly increase over time, then the dot density will also roughly linearly decrease. If the equilibrium process is dominant, an equilibrium distribution is anticipated. We believe that, after stopping growth, the density decreases over time as a result of surface diffusion, and after about 5 min, the dot density does not show any significant change over time, suggesting that the diffusion has reached an equilibrium state. The time constant is about 2.3 min with the best fit of exponential decay.

In order to understand the mechanisms of the temperature effect on the dot morphology evolution, we measured the areal dot density including the pyramids and domes on the samples grown at different temperatures and analyzed the characteristic length L_c, which is defined as the inverse of the square root of the areal density ($1/\sqrt{\text{areal_density}}$). It should be noted that many previous studies on dot morphology were based on size measurements, such as the height, base size, and volume of each dot. However, the accuracy of the dot base is questionable because of the AFM tip effect and nontrivial estimation of the dot volume for the real morphology of the dots. Figure 4 shows an Arrhenius plot of the characteristic length L_c. For a typical Arrhenius relation, the data sit around a line, whose slope represents the correspond-

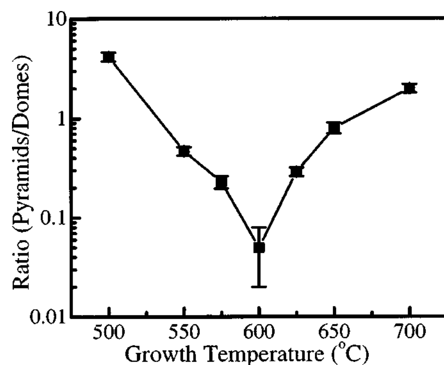

FIG. 2. Ratio of the number of pyramidal dots to that of the dome dots as a function of the growth temperature from 500 to 700 °C. The optimum temperature is about 600 °C for the formation of monomodal distributed dots.

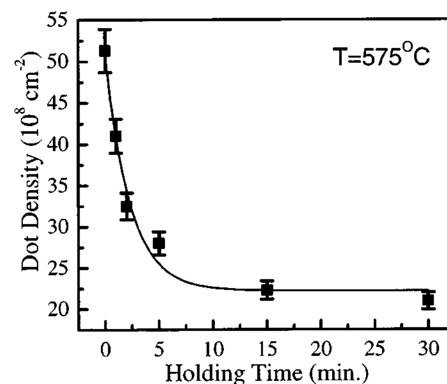

FIG. 3. Areal dot density vs the holding time at growth temperature of 575 °C. The solid line is a simple exponential decay fit showing that the time constant is 2.3 min.

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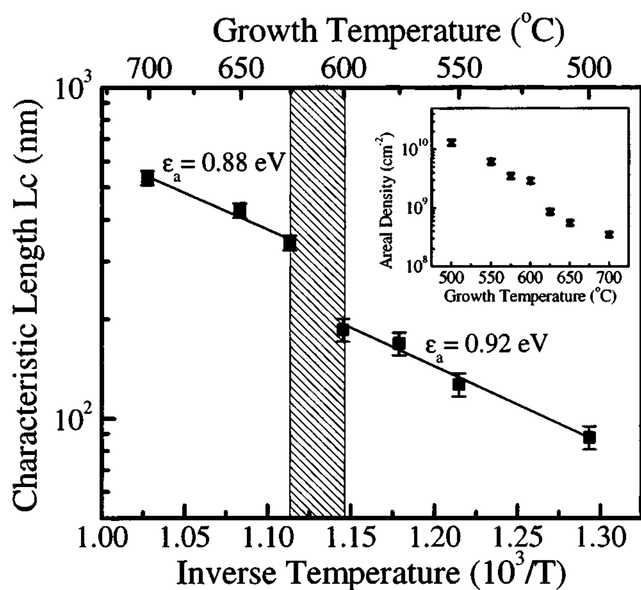

FIG. 4. Arrhenius plot of the characteristic length (L_c) vs the growth temperature. Discontinuity of the length takes place between 600 and 625 °C. The inset shows the areal dot density as a function of the temperature.

ing activation energy. Two line segments, however, were found to fit the experimental data with a discontinuity of L_c at a temperature between 600 and 625 °C well. The offset suggests lattice relaxation due to the interdiffusion at high temperature. Two different slopes give two slightly different activation energies of 0.88 and 0.91 eV, respectively. This may be due to the fact that the Ge atoms diffuse on wetting layers with different Ge mole fractions before and after significant Si and Ge interdiffusion.

It is generally believed that the dot morphology depends on whether the process is thermodynamically or kinetically constrained.[7] In real-time low-energy electron microscopy studies, Ross *et al.*[5] observed that larger dots grow while smaller dots shrink and disappear with an increase in Ge deposition, instead of all the dots growing, consistent with the Ostwald ripening phenomenon, which is a coarsening process of a particle size driven by the Gibbs–Thomson effect. Hence, no stable size distribution would be reached. Theoretical justification based on a kinetic mechanism was also made to support their observations. On the other hand, based on thermodynamic analysis, Shchukin *et al.*[8] proposed that for some systems there are minima in the free energy of self-assembled dot ensembles, resulting in a stable size distribution at a particular dot size instead of suffering Ostwald ripening. A convincing experimental observation is the scanning tunneling microscopy measurements of the dot size distribution,[9] in agreement with the Shchukin model, showing the size with a stable Boltzmann distribution. Kamins *et al.*[1] conducted annealing experiments on a constant material to distinguish the two mechanisms, that is, Ostwald ripening and equilibrium distribution. They found that dot evolution slowed with an increase in annealing time at 550 and 600 °C and it reached an equilibrium distribution. The realization of uniform Ge at 600 °C supports Shchukin's model, a final stable size distribution instead of Ostwald ripening.

The line offset with two slopes in Fig. 4, however, suggests that diffusion is not the only driving force for determining dot density at higher temperatures (>600 °C). The bimodal distribution at low temperature is due to the limit of diffusion. As the temperature increases, Ge adatoms have sufficiently long diffusion length (about 3 μm at 600 °C)[10] and find preferential sites, and it is much longer than the characteristic length (0.2 μm). However, if the temperature T_s is higher than 625 °C, intermixing between the Si and Ge interface results in modification of the interface strain relaxation and redistribution.[11,12] Moreover, the strain at the edges of Ge dots becomes stronger as the dot size gets bigger.[13] The consequence of thermoinduced and strain-induced interdiffusion is that the wetting layer is no longer a pure Ge layer, but a nonuniform SiGe-like layer on the surface, leading to a nonuniform strain distribution. The lower strain from the lower Ge concentration of a SiGe structure allows the formation of larger pyramids, i.e., corruption of the monomodal distribution and reoccurrence of bimodal dots [shown in Figs. 2(d) and 2(e)]. Both the line offset and the change in activation energy in Fig. 4 have proved that there was a change of mechanism from enhanced diffusion to intermixing of Si with Ge in the temperature range investigated.

In summary, the temperature effect on the formation of self-assembled Ge dots on Si (001) substrates was investigated. It was found that growth temperature of 600 °C is an optimum temperature for the formation of uniform Ge dots due to enhanced diffusion. Highly uniform Ge dots with height deviation of about ±3% formed at temperature of 600 °C. At lower temperatures, diffusion tends to limit the formation of uniform dots while at high temperatures, the intermixing between Si and Ge reduces strain and allows the formation of larger size dots. Finally, it is worth pointing out that the optimum temperature of 600 °C was obtained for our particular experimental conditions. This number may vary depending on the growth rate, vacuum conditions, source type (gas or solid source), whether or not there is all hydrogen ambient, postgrowth holding time, and other growth conditions.

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