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Ion-Beam Assisted Surface Islanding During Ge MBE on Si

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Effects of low-energy Ge^+ ion irradiation on the transition from two-dimensional (2D) to three-dimensional (3D) growth during Ge/Si(111) heteroepitaxy were studied by *in situ* reflection high-energy electron diffraction (RHEED) and *ex situ* scanning tunnelling microscopy (STM). The continuous and pulsed ion beams were used. The data received by these methods directly indicate that ion irradiation leads to facilitation of 2D - 3D transition. The STM investigations have shown that the density of 3D islands is higher and size distribution is more narrow at ion-assisted growth in comparison with conventional epitaxy. The results of Monte-Carlo simulation have shown that two mechanisms of ion beam action can be responsible for facilitation of 2D - 3D transition. There are: 1) generation of adatoms by ion impacts which leads to transfer of material from underlying layers to upper layer and 2) enhancement of surface diffusion which may be caused by ion-stimulated reconstruction of the surface. Both mechanisms promote 2D -3D transition.

1. Introduction

Heterojunctions and nanostructures, formed by SiGe heteroepitaxy, have attracted considerable interest in recent years because of their potential applications in high-speed electronic, infrared detection and promising devices based on quantum effects [1,2]. Mechanical stresses in the growing layer caused the morphological changes of the surface. As a result, the flat surface has grown until critical thickness of wetting layer is reached and then this process is interrupted by nucleation of 3D islands on the top of the epilayer (Stranski-Krastanov growth mode). The SiGe nanostructures containing islands are commonly studied at present as a system with quantum dots.

A conventional manner to control island formation (size, form, density) is to alter the growth conditions by changing substrate temperature and molecular flow. Tunability in zero-dimensional semiconductor technology thus offers obvious advantages in extending the range of possibilities for devices. The new facility to tune island dimensions and their surface densities is expected to be provided by the use of ion beam with energy exceeding of energy in the molecular beam, but less energy of defects generation in the bulk of wetting layer (and substrate). Energy of particles in the molecular beam is defined by temperature of the beam source. Usually its value does not exceed 0.1 eV. Using hyperthermal species (with energy ~100eV) during epitaxy result in dramatically changes in growth kinetics and final physical properties of solid films [3-5]. It promotes crystal growth at extremely low temperature and improvement of surface smoothness.

In this study the morphology of Ge surface of pseudomorphic layer under irradiation by low energy (~200 eV) Ge^+ ions during molecular beam epitaxy (MBE) of Ge on Si(111) was investigated. Two different approaches were used. In the first, the continuous low-energy ion beam irradiation was carried out through epitaxy. In the second one, we affected the growing surface by the pulsed low-energy ion beam at selected times during epitaxial growth. This approach was found to be a powerful tool to study the mechanism of ion beam induced surface morphology modification during Si(111) homoepitaxy [5].

2. Experimental technique

The experimental setup includes a growth chamber with background pressure less then 10^{-10} Torr equipped with BN crucible evaporation cell. The silicon wafers were Si(111) within 0.15° according to X-ray diffraction data. Density of Ge flux varied within 10^{12}–10^{15} $cm^{-2}s^{-1}$ by changing crucible temperature. Above the crucible cell the system for ionisation of Ge flux and accelerating of Ge^+ ions was located. The ionised part of molecular beam depended on design of the MBE source and had a value of 0.1% or 0.5%. The pulsed accelerating voltage unit allowed to form pulses of ion current with duration from 0.1 to 1 s. The energy of Ge^+ ions varied within 50–270 eV. The molecular and ion beams hit on the substrate at 54.5° off-normal direction.

To study the surface morphology *in situ*, we used the RHEED with observation of specular intensity oscillations during two-dimensional layer-by-layer growth [6]. The experiments were made in two irradiation modes: with continuous ion beam and with pulsed ion beam irradiation during Ge MBE. Ion pulsed action was made at different stages of growth according to the different degree of filling of the surface layer.

Scanning tunnelling microscopy is capable of imaging surface on the atomic scale. All STM images were taken *ex situ* at room temperature in the constantcurrent mode with a tunnelling current.

3. Experimental results

During Ge/Si(111) heteroepitaxy from molecular beam we have observed the RHEED intensity oscillations. The period of RHEED oscillations was equal to the deposition time of 1 biatomic layer (1 BL=1.56 10^{15} atoms/cm^2). After deposition a few BL the response of the RHEED oscillations is rapidly damping of both the amplitude and the average RHEED intensity level. This is connected with increasing of surface roughness due to transition from layer-by-layer 2D growth to 3D growth mode of Ge islands at the critical thickness of the pseudomorphic film. The results of our experiments with observed 2D-3D transition are in good agreement with those obtained previously [6,7]. The experiments were performed at different substrate temperatures, deposition rates and ion fluxes.

Recently we have published the temperature and flux density dependencies of RHEED intensity during Ge/Si(111) heteroepitaxy with low-energy ion irradiation [8]. We used previous results to find the optimal regime of ion action to reach more pronounced effect.

We selected the following experimental parameters: the temperature of Ge/Si heteroepitaxy - 350°C, deposition rates - 0.12 BL/s, the ion flux density - 5.5 10^{11} $cm^{-2}s^{-1}$ (the ionized part of molecular beam was 0.5%), energy of Ge^+ ions - 200eV.

The irradiation with continuous ion beam during Ge/Si(111) heteroepitaxy resulted in reducing a number of RHEED oscillations (Fig. 1, curve 2) as compared with the conventional heteroepitaxy (Fig. 1, curve 1). This corresponds to facilitation of 2D-3D transition or decreasing of critical thickness of Ge wetting layer in Stranski-Krastanov growth mode.

Ion pulsed action was found to increase RHEED specular beam intensity if the pulsed ion beam was turned on at a fractional surface coverage more than half (Fig. 1, curve 3). The intensity enhancement corresponds to the improvement of surface smoothness during molecular beam growth apparently due to increasing of adatom mobility on ion-beam reconstructed surface.

For detailed investigations of observed effects we concentrated on the 3 types of structures obtained by: 1) usual epitaxy from molecular beam in layer-by-layer growth mode, 2) epitaxy with continuous ion beam irradiation, 3) epitaxy with pulsed ion beam. The amount of deposited Ge was identical for three types of structures and was equal to 3.5 BL. The STM study has shown that type 1 structures contained only two-dimensional Ge islands (Fig. 2 a), type 2 and 3 structures contained three-dimensional Ge islands (Fig. 2 b, c), formed on the surface of pseudomorphic Ge(111) layer and having the shape of truncated pyramids with {113} facets. The density of 3D islands on surface of type 2 structures was about three times higher than density of type 3 structures.

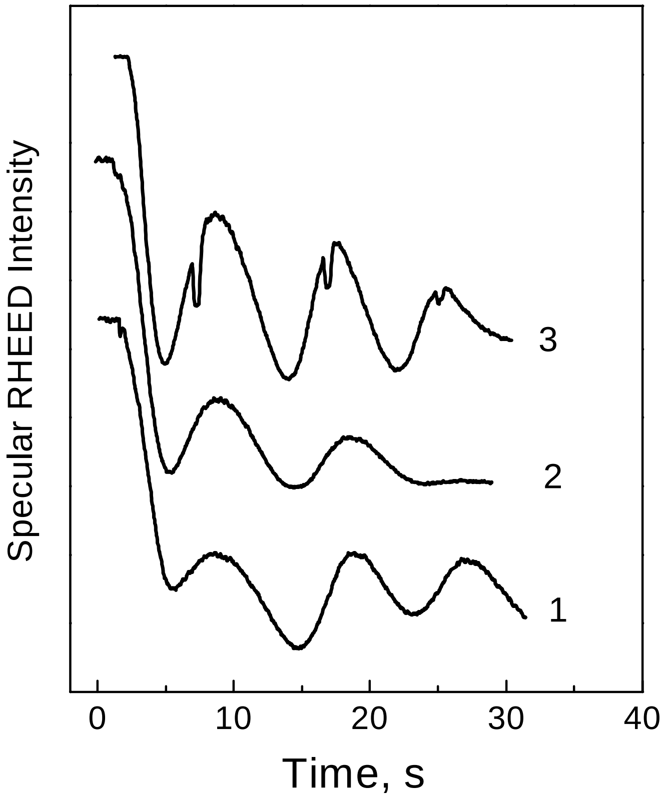
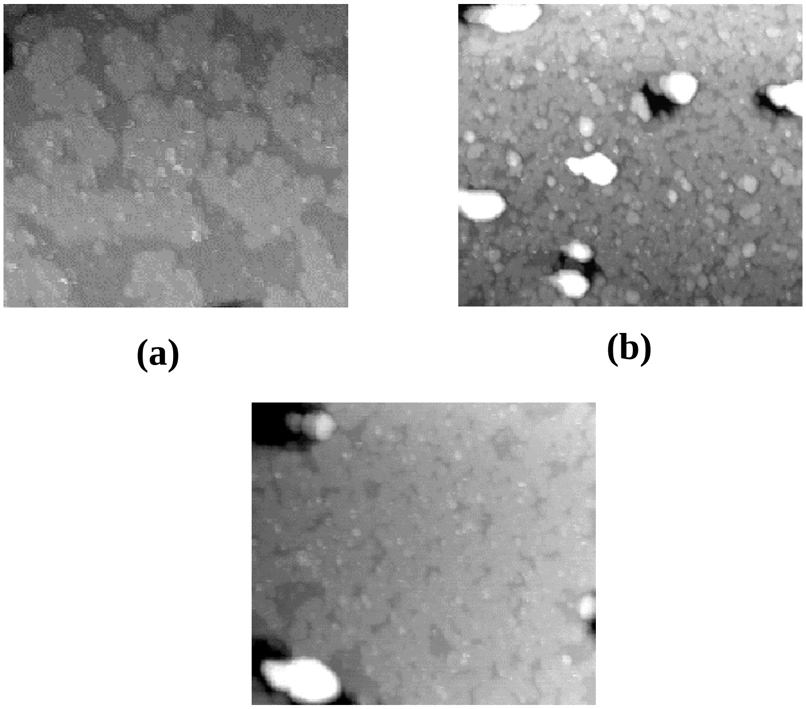


Figure 1. Specular RHEED intensity vs time recorded during Ge/Si(111) heteroepitaxy in the three regimes: 1) without ion irradiation; 2) with continuous ion beam irradiation, 3) with pulsed ion beam actions. Substrate temperature - 350^0 C; deposition rate - 0.12 BL/ s; the ion flux density - 5.5 10^{11} cm^{-2} s^{-1} ; the energy of Ge ions - 200 eV; pulse duration - 0.5s. The arrows indicate the times of pulsed ion beam actions.

Near the 3D islands one can observed the vacancy depressions on the surface, which disappear at the latest stages of growth. The data received by STM method directly indicate that irradiation by continuous and pulsed ion beam stimulate the transition to 3D growth.

The STM investigations at latest growth stages (after deposition of 5 BL) shown that the density of 3D islands is higher in the case ion-assisted growth (Fig. 3 b) than without ion beam (Fig. 3 c). The size distribution become more narrow (Fig.3 b, d).



(a)

(b)

(c)

Figure 2. STM images of three surface structures obtained after deposition of 3.5 BL of Ge on Si(111) substrate by: a) usual epitaxy in layer-by-layer growth mode; b) epitaxy with continuous ion beam irradiation; c) epitaxy with pulsed ion beam actions. The size of STM images is 300nm x 300 nm. Parameters of molecular and ion beams and substrate temperature are the same as on Fig 1.

4. Modelling.

In order to clarify the influence of the ion irradiation on the 2D-3D transition, we have simulated this process by Monte–Carlo method. We include in the model the main elementary processes, which may provide contribution to the observed phenomena. At the first step we have modelled the pure heteroepitaxy Ge/Si(111) without ion irradiation. It is commonly accepted that the lattice-mismatched strain is the origin of the 2D-3D transition [9,10]. The strain accumulated in the surface layer changes the diffusion across the surface. The strain effects stronger the surface step edge atoms, because these atoms are more weakly bonded to the surface. In the framework of this task we calculated the strain distribution near the 2D island arising during heteroepitaxy. For present calculations we used the Keating potential of elastic atomic interaction [11].

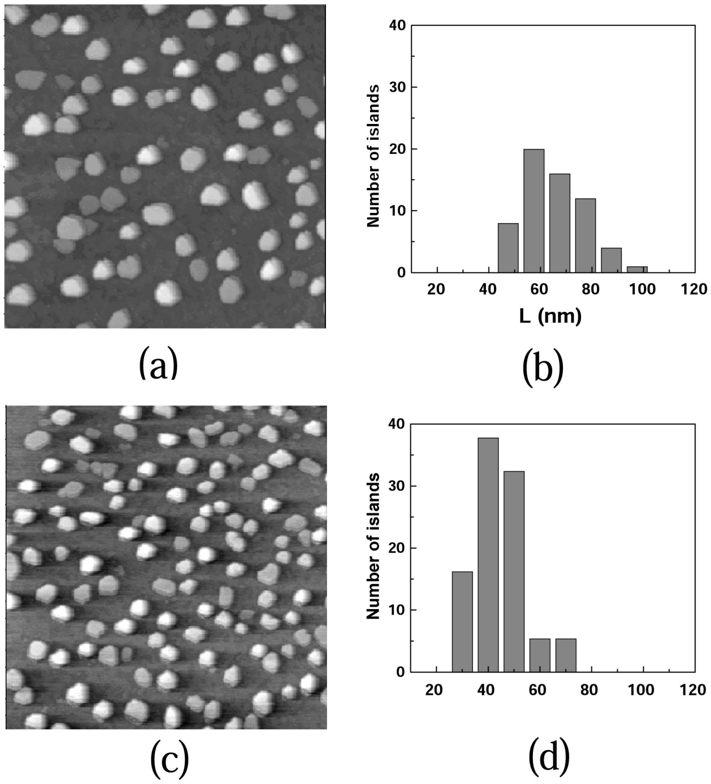


Figure 3. STM images and corresponding size distributions of 3D islands after deposition of 5 BL of Ge at a) usual epitaxy; b) epitaxy with pulsed ion beam actions. The size of STM images is 1000nm x 1000nm. Parameters of molecular and ion beams and substrate temperature are the same as on Fig 1.

The results of these calculations showed that the maximum of the strain is located near the island edge (see Fig. 4). The strain energy was found to dependent on the island size. When size is increased, the strain energy (E_{strain}) at the islands edge rises (Fig. 5). We used these results in simulation of surface diffusion. We adopted commonly accepted concept that the diffusion activation energy depends on the bonding environment and elastic energy associated with the strain is $E=E_{bond} -- E_{strain}$, where $E_{bond}=n_1 E_1 + n_2 E_2$, ($E_1$ is nearest-neighbour binding energy, E_2 is next nearest-neighbour binding energy, n_1 is the number of nearest neighbours, n_2 is the number of next nearest neighbours). The following parameters were used in modelling: $E_1 = 1.2$ eV, $E_2 = 0.1$ eV. We taken into account E_{strain} only for atoms on the island edge. So, the probability of atom detachment from the edge of islands is enhanced, atoms become mobile and can hop to the next level, that can lead to forming of the 3D islands. The simulation of growth within above assumptions results in the 2D-3D transition when the critical thickness is achieved. The main features of the simulation model presented in detail elsewhere [6,9,10].

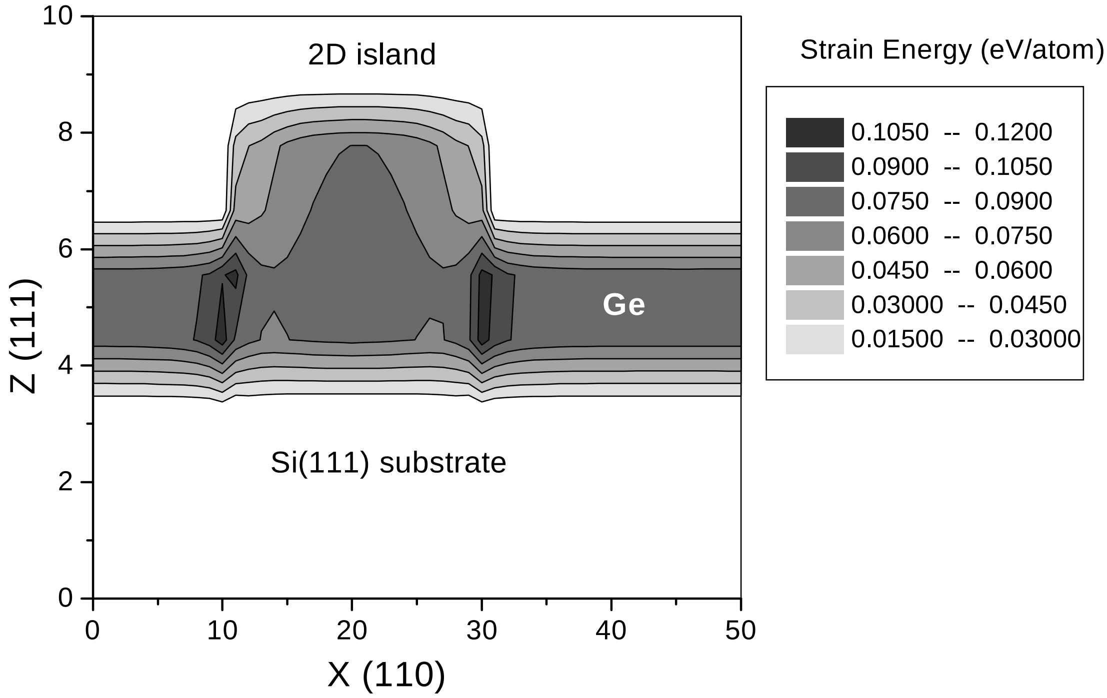


Figure 4. The strain energy distribution in plane for Ge/Si(111) heterostructure near 2D triangle-shaped island. This plane crossed the centres of island sides. The distances taken in the number of atomic layers.

At the second step we include in the model the low energy ion beam irradiation. We assume that the influence of the ion beam consists of following processes:

a) the sputtering of the material;
b) the generation of additional adatoms and surface vacancy clusters;
c) ion-assisted enhancement of adatom diffusion.

According to molecular dynamics simulations of low-energy interaction with Si(111) surface [12], ion impact produces one surface vacancy cluster and additional adatoms at a few interatomic distance from this cluster. We assume that these results remain true qualitatively for Ge(111). Only the quantitative characteristics are changed. For simulations the following parameters are taken: the size of vacancy cluster is 10, the number of exited adatoms is 9 and one atom is sputtered [13].

We distinguish the following two mechanisms of ion influence which can be responsible to observed phenomena of ion-assisted facilitation of 2D-3D transition. 1) Generation of adatoms leads to transfer of material from underlying layers to upper adatom layer, in other words, atoms release from bulk to layer of mobile adatoms. 2) Enhancement of surface diffusion may be caused by ion-stimulated reconstruction of surface. This reconstruction occurs due to release of the energy of accelerating particles. For our simulations we used the surface diffusion coefficient in 10 times greater than one for case without ion-irradiation according to recent experimental measurements [14].

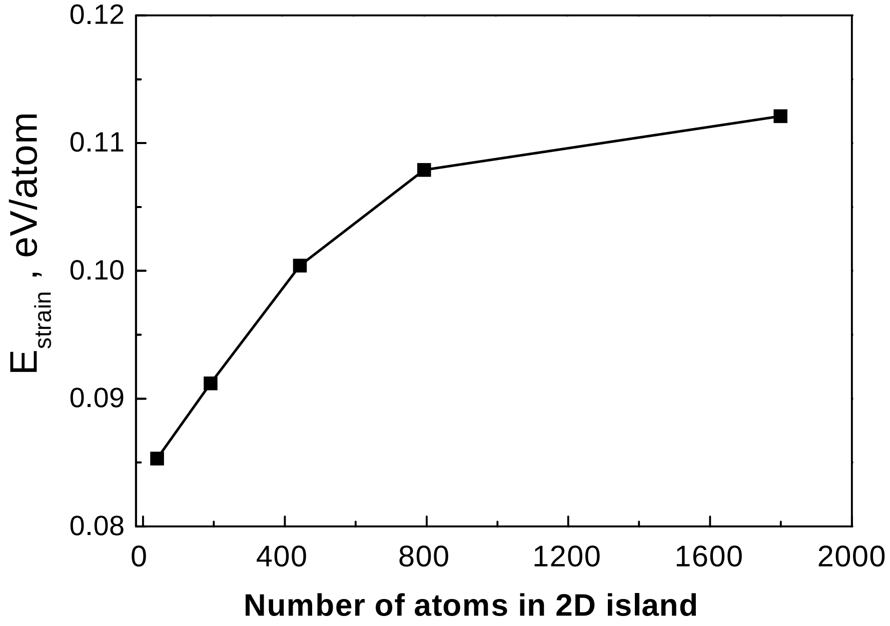


Figure 5. The strain energy dependence on the island size taken at the island step edge.

As a parameter characterised the surface morphology, we took surface step density (S), which is the analogue to experimental RHEED intensity profiles [15]:

$$S = \frac{1}{4M} \sum_{i=1}^{\sqrt{M}} \sum_{j=1}^{\sqrt{M}} \left[\left| h_{i,j} - h_{i+1,j} \right| + \left| h_{i,j} - h_{i,j+1} \right| \right],$$

where *M* is number of surface lattice sites, $h_{i,j}$ is the surface height at the *(i,j)* point. This quantity is proportional to the number of atoms along the perimeter of islands and surface vacancy clusters. Also we monitored the surface morphology by tracing of the images of the simulated surface at selected times. MC modelling was performed at the same temperatures, molecular and ion beam fluxes as in experiments.

The simulations have shown that the growth can occur in two regimes: 2D layer-by-layer growth, when the oscillations of S are observed, and 3D growth, when oscillations disappear (Fig.6). The 2D - 3D transition was also confirmed by images of the simulated surface. When we simulated growth with the ion beam, we obtained the facilitation of 2D - 3D transition.

For the case when the main mechanism responsible for facilitation is material transfer from underlying layers to upper layer due to generation of adatoms, the simulations have shown, that the 2D - 3D transition occurs earlier (Fig. 6 b) than in the case of the usual heteroepitaxy (Fig. 6 a). The number of oscillations reduced down to 2. The density of 3D islands is higher, than the one in the case usual epitaxy with the same deposited material (3.4 BL, that corresponded to the onset of the 2D - 3D transition).

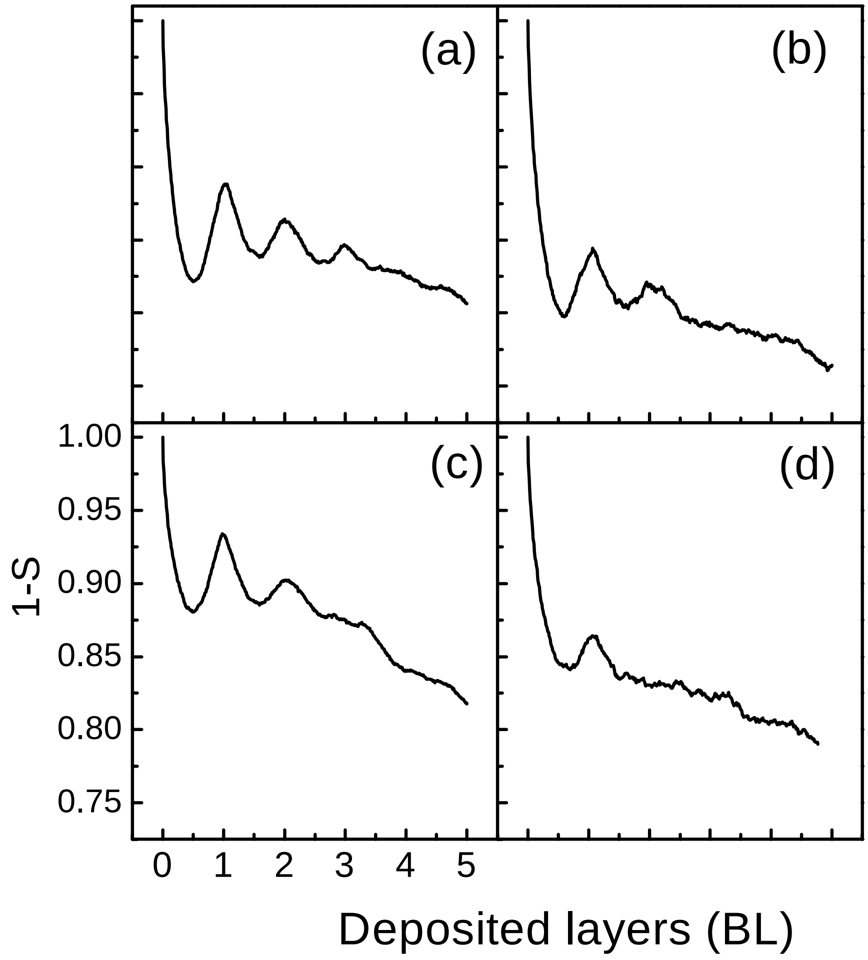


Figure 6. Simulated step density evolution during Ge/Si(111) heteroepitaxy without ion irradiation-(a) and with ion irradiation-(b),(c),(d). The three latest cases corresponds to different mechanisms of ion beam influence: (b) adatom generation by ion impacts leading to transfer of material to higher atomic layers; (c) ion stimulated surface diffusion; (d) including both mechanisms.

For the case when the main mechanism responsible for facilitation is the enhancement of surface diffusion caused by ion-stimulated reconstruction of surface, we obtained that the transition occurred at the same critical thickness as in the first case (Fig. 6 c). But the size and density of islands are different. The average size of islands becomes larger and higher, and density is decreased. In this case the surface smoothness is higher in comparison with the first case, when we only took into account the generation of adatoms by ion beam.

The ion-induced facilitation of 2D - 3D transition by second mechanism is clear. Enhancement of surface diffusion leads to an increase in average size of 2D islands. As consequence, the strain energy become higher, and the edge atoms are promoted to the higher level. This leads to nucleation of 3D islands at the earlier stage of growth. The same effect can be achieved by raising of the substrate temperature.

The facilitation of 2D - 3D transition by first mechanism is not so obviously. It can be expected that the ion-beam action will lead to reduction of the average size of islands and, as a consequence, to a decrease in the strain energy at the island edge inhibiting the nucleation of 3D islands. Exactly this phenomena was observed in experiment at epitaxy $Ge_{0.5}Si_{0.5}$ at more higher ion fluxes [4] than in our experiment. This is explained by destruction of 3D islands caused by the ion impacts. But at our ion beam fluxes the possibility of direct hit into a 3D island is low. And ion action provided the reverse effect. The ion impacts produced additional adatoms from the surface which can pile up on the top of the existing 2D islands and nucleate 3D islands. So, both mechanisms promote transition to 3D growth and the simulations including both mechanisms simultaneously have shown more fast transition (Fig. 6 d), the critical thickness decreasing down to 1 BL.

The results of MC modelling showed that generation of adatoms and surface vacancy clusters by ion impacts and ion-enhanced surface diffusion lead to facilitation of the 2D - 3D transition during Stranski-Krastanov growth. Thus, the observed experimental results can be explained in terms of these two mechanisms.

5. Summary

We have studied the initial stages of low temperature Ge/Si(111) heteroepitaxy in two modes: in the presence of continuous irradiation and pulsed action by low energy Ge ions. We have found that the ion beam irradiation leads to facilitation of the 2D - 3D transition, enhancement of 3D island density and narrowing of size distribution. In order to clarify the influence of the ion irradiation on the 2D - 3D transition, we have simulated this process by Monte–Carlo method. The results of MC modelling showed that two mechanisms of ion influence can be responsible for the observed phenomena of ion-assist facilitation of the 2D-3D transition. There are: 1) generation of adatoms, which leads to transfer of material from underlying layers to upper adatom layer and 2) enhancement of surface diffusion which may be caused by ion-stimulated reconstruction of the surface. Both mechanisms promote transition to 3D growth.

Our experimental results demonstrated that the low energy ion beam irradiation during GeSi heteroepitaxy give the possibility to control the size and density of islands, and moreover, it provides the narrower island size distribution compared with conventional epitaxy. This is important for potential applications in nanostructure technology.

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References

[1] O.P. Pchelyakov, Yu.B. Bolkhovityanov, A.V. Dvurechenskii et al, Thin Solid Films, **367** (2000) 75.

[2] A.I. Yakimov, V.A. Markov, A.V.Dvurechenskii, O.P. Pchelyakov, Phil. Mag. B, **2** (1992) 701.

[3] J.W. Rabalais, A.H. Al-Bayati, K.J. Boyd et al, Phys. Rev. B, **53** (1996) 10781.

[4] S.W. Park, J.Y. Shim, H. K. Baik, J. Appl. Phys., **78** (1995) 5993.

[5] A.V. Dvurechenskii, V.A. Zinovyev, V.A. Markov, V.A. Kudryavtsev, Surf. Sci., **425** (1999) 185.

[6] O.P. Pchelyakov, V.A. Markov, A.I. Nikiforov, L.V. Sokolov, Thin Solid Films, **306** (1997) 299.

[7] B. Voigtlander and A. Zinner, Appl. Phys. Lett., **63** (1993) 3055.

[8] A.V. Dvurechenskii, V.A. Zinovyev, V.A. Kudryavtsev, J.V. Smagina, JETP Letters, **72** (2000) 131.

[9] K.E. Khor and S. Das Sarma, Phys. Rev. B., **62** (2000) 16657.

[10] D.V.Brunev, I.G.Neizvestny, N.L.Shwartz, and Z.Sh.Yanovitskaja, Izv. Akad. Nauk Fiz., **65** (2001) 196.

[11] P.N. Keating, Phys. Rev., **145** (1966) 637.

[12] V.A. Zinovyev, L.N. Aleksandrov, V.A. Dvurechenskii, K.-H. Heinig, D. Stock, Thin Solid Films, **241** (1994) 167.

[13] J. A. Floro, B.K. Kellerman, E. Chason et. al, J.Appl.Phys., **77** (1995) 2351.

[14] R. Ditchfield and E.G. Seebauer, Phys. Rev.B., **63** (2001) 125317.

[15] D.D. Vvedensky, S. Clarke, Surf.Sci., **373** (1990) 225.