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Ge/Si quantum dot nanostructures grown with low-energy ion beam-assisted epitaxy

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Abstract

Scanning tunneling microscopy (STM) and reflection high-energy electron diffraction (RHEED) experiments were performed to study growth modes induced by hyperthermal Ge^+ ion action during molecular beam epitaxy (MBE) of Ge on Si(100). The continuous and pulsed ion beams were used. These studies have shown that ion beam bombardment during heteroepitaxy leads to decrease in critical film thickness for transition from two-dimensional (2D) to three-dimensional (3D) growth modes, enhancement of 3D island density, and narrowing of island size distribution, as compared with conventional MBE experiments. Moreover, it was found that ion beam assists the transition from *hut*- to *dome*-shaped Ge islands on Si(100). The crystal perfection of Ge/Si structures with Ge islands embedded in Si was analyzed by Rutherford backscattering/channeling technique (RBS) and transmission electron microscopy (TEM). The studies of Si/Ge/Si(100) structures indicated defect-free Ge nanopaticles and Si layers for the initial stage of heteroepitaxy (five monolayers of Ge) in pulsed ion beam action growth mode at 350 °C. Continuous ion beam irradiation was found to induce dislocations around Ge clusters. The results of kinetic Monte Carlo (KMC) simulation have shown that two mechanisms of ion beam action can be responsible for stimulation of 2D–3D transition: (1) surface defect generation by ion impacts, and (2) enhancement of surface diffusion.

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1. Introduction

Self-assembled Ge islands on Si(100) have been intensively investigated as the basis of future electronic and optical devices [1,2]. The self-assembled (ordering) effects imply the appearance of islands with preferred characteristics: sizes, shapes, spacing between nanoclusters, and mutual arrangement. The ordering processes are accompanied by the minimization of free energy of the system. At present, particular attention is being given to the size distribution of islands because this parameter of a system of quantum dots is of crucial importance in practical applications. It is commonly accepted that the energy gain caused by the strain relaxation in island apexes is the key factor in the transition from a two-dimensional (2D) to three-dimensional (3D) island growth. The 3D islands are formed due to the morphological instability of strained films in systems with a large (more than 2%) lattice mismatch between a film and substrate, among which Ge/Si (4%) and InAs/GaAs (7%) are most familiar.

The conventional way to control island formation (size, shape, and density) is variation of growth conditions by the alteration of substrate temperature and molecular flux. However, establishing a method to achieve sufficiently uniform island sizes with regular spatial distribution still remains a critical issue. This should be solved since well-defined sizes with little dispersion are generally required for any practical applications. The new facility to tune

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island dimensions and their surface densities is expected to be the use of ion beam with energy exceeding the energy in the molecular beam, but less than the energy of defects generation in the bulk of the growing layer (and substrate). The results of our recent study indicate that irradiation with low-energy Ge^+ ions during Ge/Si(111) heteroepitaxy stimulates the nucleation of 3D Ge islands and reduces the critical thickness at which the 2D–3D transition occurs [3,4].

In this work, we present the results of investigation of size ordering and shape transition in an ensemble of Ge nanoislands formed by Ge/Si(100) heteroepitaxy under low-energy ion beam irradiation and crystal perfection of Ge/Si structures with quantum dots embedded in Si. In order to clarify the effect of the ion irradiation on 3D island nucleation, we have carried out the simulation of ion-assisted growth of Ge films on Si by kinetic Monte Carlo (KMC) method.

2. Experimental

The experiments were carried out in an ultrahigh-vacuum chamber of molecular beam epitaxy (MBE) setup equipped with electron beam evaporator for Si and effusion cell (boron nitride crucible) for Ge. A system of ionization and acceleration of Ge^+ ions provided the degree of ionization of Ge molecular beam from 0.1% to 0.5%. A pulsed accelerating voltage supply unit generated ion current pulses with duration of 0.5–1 s and ion energy of 50–200 eV. The angle of incidence of the molecular and ion beams on the substrate was 54° to surface normal. The analytical section of the chamber included a reflection high-energy (20 keV) electron diffraction unit.

Heteroepitaxy was carried out at substrate temperature varied in the range of 300–500 °C. The rate of Ge deposition varied from 0.05 to 0.1 monolayer (ML) per second. We investigated three types of Ge/Si(100) heterostructures grown by (1) conventional MBE of Ge on Si; (2) MBE with single-pulsed Ge^+ ion beam action for each Ge monolayer completed at layer-by-layer growth mode; and (3) MBE under continuous irradiation by Ge^+ ion beam. Evolution of surface morphology was studied in situ by reflection high-energy electron diffraction (RHEED) and ex situ by scanning tunneling microscopy (STM). Crystal perfection of Ge/Si structures was analyzed by Rutherford backscattering/channeling technique (RBS) and transmission electron microscopy (TEM). For that, a 150-nm-thick cap layer of Si was grown at 500 °C by conventional MBE (with no irradiation) over the Ge layer.

3. Results and discussion

Examples of STM patterns observed for structures with 5 ML of Ge grown on Si(100) at 350 °C are shown in Fig. 1. An average size of islands obtained by conventional MBE was 22 ± 3.5 nm and their dispersion (size inhomogeneity) was 16%. In experiments with pulsed irradiation of Ge^+ ion beam, the average island size was 6.5 ± 0.7 nm and dispersion was 10%. In the case of continuous irradiation with Ge^+ ion beam, the size of islands diminished (18 ± 5.4 nm), too, but their dispersion increased (30%) in comparison with those at conventional MBE. The surface density of Ge nanoislands for the structures of the second type was 6.8×10^{11} cm^{-2}, which is approximately seven times higher than that for the structures of the first type (~10^{11} cm^{-2}). The density of Ge nanoislands in experiments under continuous ion irradiation with Ge^+ was 2×10^{11} cm^{-2}. A decrease in the full width at half maximum of the size distribution function is evidence for the size ordering in an ensemble of Ge nanoclusters.

We found that this ordering process is caused by pulsed ion beam actions at each Ge monolayer completed in layer-by-layer growth mode. RHEED was used for in situ control of morphology and stressed state of Ge/Si(100) surface. In addition, the starting point of *hut* and *dome* cluster formation was observed due to specific reflexes produced in RHEED images by {105} and {113} facets [2,5].

Fig. 2 shows the evolution of Ge lattice constant during the conventional MBE and MBE with pulsed Ge^+ ion beam actions. The arrows in this figure separate the stages of 2D growth: the growth of *hut*- and *dome*-shaped islands, respectively. One can see that the ion beam action results in the earlier 2D–3D transition as well as the formation of dome clusters. The effect was found to be dependent on the energy of ions. Under irradiation by 200-eV ions, the *hut* clusters are formed 1 ML earlier as compared to case of 100 eV, while *dome* clusters are created 2 ML earlier.

It is generally believed that the nucleation of 3D islands occurs at the imperfections of the 2D layer (heterogeneous nucleation mechanism). Hence, preliminary creation of nucleation sites such as vacancy depressions by ion impacts is an efficient way to control island density [6]. Indeed, our STM results demonstrate that ion bombardment stimulates nucleation of 3D islands. However, the narrowing in island size distribution is observed only in pulsed ion beam action growth mode (Fig. 1b). The continuous ion irradiation leads to reversed effect (Fig. 1c). The size ordering of the Ge islands during heteroepitaxy with pulsed Ge^+ ion beam irradiation is, most likely, caused by the following factors: (1) a synchronization of island nucleation by pulsed ion beam actions, and (2) ion-induced enhancement of surface diffusion. The latter facilitates adatom exchange between islands.

The continuous ion beam irradiation offers random Ge islands nucleation, that is why it fails with Ge nanoparticle size ordering.

RHEED measurements of the lattice constant during Ge/Si(100) heteroepitaxy with 200-eV pulsed ion beam actions showed that Ge films after formation of *dome* clusters were more strained (~2%) than those in the case of conventional heteroepitaxy. This means that shape tran-

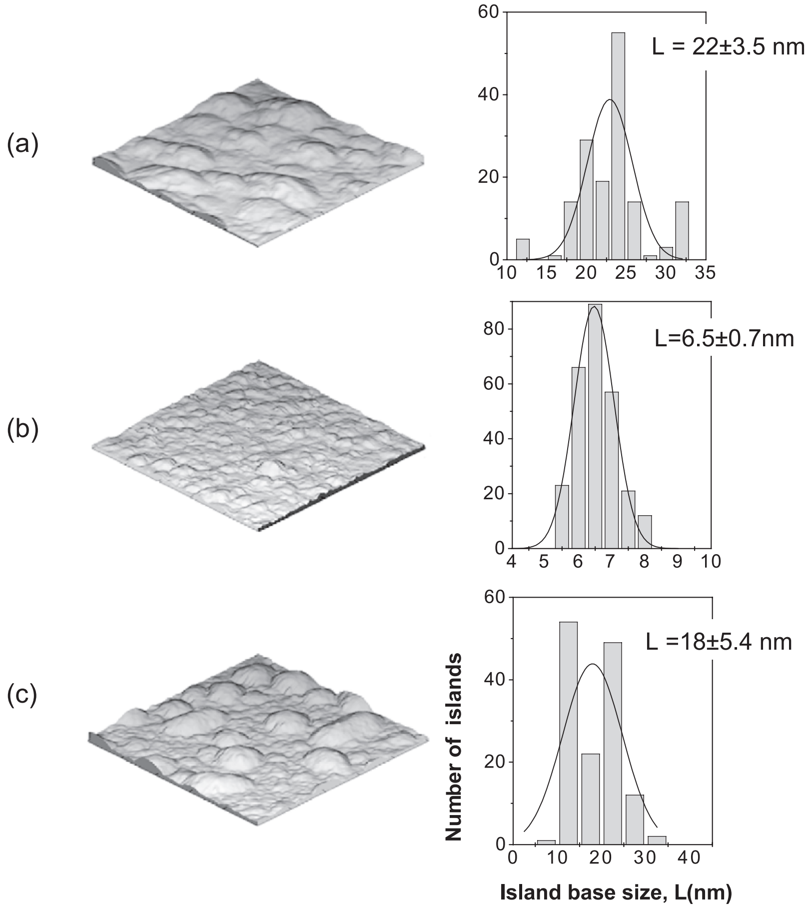

Fig. 1. STM images of 100×100 nm surface area and size distribution of 3D islands for three types of Ge/Si(100) heterostructures after Ge deposition of 5 ML: (a) conventional MBE; (b) MBE with pulsed irradiation of 100-eV Ge^+ ions; (c) MBE with continuous ion irradiation. The rate of Ge deposition is 0.1 ML/s. Substrate temperature, 350 °C.

sition from *hut* to *dome* clusters is driven by stress production induced by ion beam bombardment. These additional stress can be attributed to interstitial complex generated near the surface by energetic particles (so-called ion-peening effect [7]).

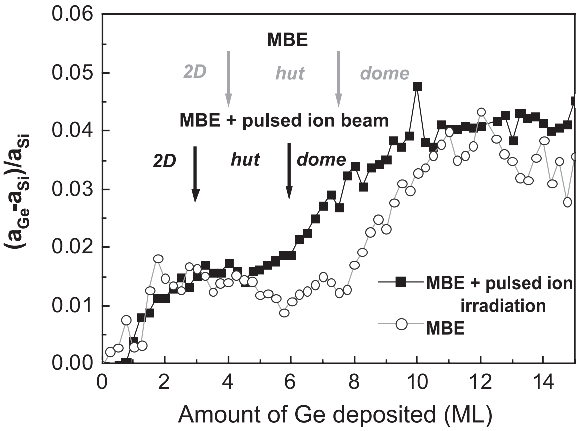

Fig. 2. Variation of Ge lattice constant during conventional MBE of Ge on Si(100) and MBE with 100-eV Ge^+ pulsed ion beam actions. The arrows indicate the appearance of *hut* and *dome* clusters registered by RHEED.

The treatment of RBS spectra permitted to calculate the backscattering yield for self-ordering Ge islands embedded in Si and for Si layers. The backscattering yield for perfect crystal of Si(100) is about 3%. The backscattering yield from Ge layers turns out to be sensitive to growth conditions (Fig. 3). The perfect structure of 2.5% backscattering yield was found in a mode of a pulsed irradiation for a range of 1–5 ML of Ge deposited at a temperature of 350 °C. For a lower temperature of 300 °C, the yield exceeded 5% in similar structures. This increase in backscattering has been observed also for even thicker Ge layers at higher temperatures of growth (400–500 °C). The enlarged yield was found also in the structures formed with continuous beam irradiation at 300–350 °C temperatures. The increase in backscattering yield can be attributed to altering of elastic deformation inside the Ge islands due to ion-assisted change of their size, shape, and density, and/or to generation and accumulation of point defects in the bulk region of Ge. The yield from the Si matrix slightly depends on the growth conditions and corresponds to a perfect Si structure.

TEM studies indicated defect-free Ge dots and Si layers for the initial stage of heteroepitaxy (5 ML of Ge) in pulsed

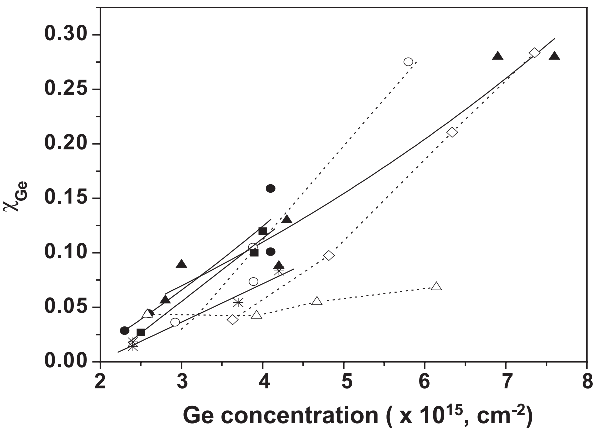

Fig. 3. Backscattering yield from Ge embedded into Si layers as dependent on Ge layer thickness. Conventional MBE: (△) 300 °C, (◇) 400 °C, (○) 500 °C; MBE with continuous ion beam: (■) 300 °C, (●)350 °C; MBE with pulsed ion beam: (▲) 300 °C, (*) 350 °C.

action growth mode at 350 °C. Continuous beam irradiation was found to induce dislocations around Ge dots.

4. Modeling

The process of 2D–3D transition under ion irradiation was simulated by the Monte Carlo method. The main elementary processes included in the model were atom deposition, diffusion, and ion impacts. At the first step, we have simulated the pure Ge/Si heteroepitaxy without ion irradiation. The diffusion activation energy was assumed to depend on the bonding environment and elastic energy associated with the strain: $E=E_{\mathrm{bond}}-E_{\mathrm{strain}}$, where $E_{\mathrm{bond}}=n_1E_1+n_2E_2$ (E_1 is the nearest-neighbour binding energy, E_2 is the next nearest-neighbour binding energy, n_1 is the number of nearest neighbours, n_2 is the number of the next nearest neighbours); E_{strain} is the strain energy per atom, calculated using the Keating potential [8]. It follows from those calculations that the strain energy is maximal near the island edge and depends on the island size [9]. We took E_{strain} into account only for atoms on the island edge. The simulation of growth within the above assumptions results in the 2D–3D transition as soon as the critical thickness of Ge layer is achieved. The main features of the simulation model are presented in detail elsewhere [9,10].

At the second step, we included the low-energy ion beam irradiation in the model. The ion beam was assumed to be responsible for the following processes: (a) sputtering of the material; (b) generation of additional adatoms and surface vacancy clusters; and (c) ion-assisted enhancement of surface diffusion.

According to molecular dynamics simulation of low-energy interaction with Si surface [11,12], qualitatively true for Ge, an ion impact in conditions similar to those in our experiment produces a cluster of 10 vacancies, 9 excited adatoms, and 1 sputtered atom.

In our simulations, we used the magnitude of surface diffusion coefficient 10 times higher than that for the case without ion irradiation, which agrees with recent experimental measurements [13,14]. The simulation has shown that growth can occur in two regimes: 2D layer-by-layer growth, where the oscillations of surface roughness are observed, and 3D growth, where oscillations disappear (Fig. 4). The 2D–3D transition is confirmed also by images of the simulated surface. For the case when the main ion-assisted process is the generation of additional adatoms and surface vacancy clusters, the 2D–3D transition occurs earlier (Fig. 4b) than in the case of the usual heteroepitaxy (Fig. 4a). The number of oscillations is reduced to two. The density of 3D islands is higher than that in the case of usual epitaxy taken at same amount of Ge deposited (3.4 ML, corresponding to the onset of 2D–3D transition).

For the case when the main ion-assisted process is the enhancement of surface diffusion, we found that the transition occurred at the same critical thickness as in the first case (Fig. 4c). But the size and density of islands are different. The islands become larger and higher, and density decreases. Also the surface roughness is lower in comparison with the case when only the surface defect generation by ion beam was taken into account. The ion-assisted enhancement of surface diffusion leads to increase in the average size of the 2D island. As a consequence, the strain energy becomes higher, which promotes the hops of atoms from an edge to the upper layer. This leads to nucleation of 3D islands at the earlier stage of growth. The facilitation of 2D–3D transition by defect generation mechanism is interpreted as the result of ion impact producing excited

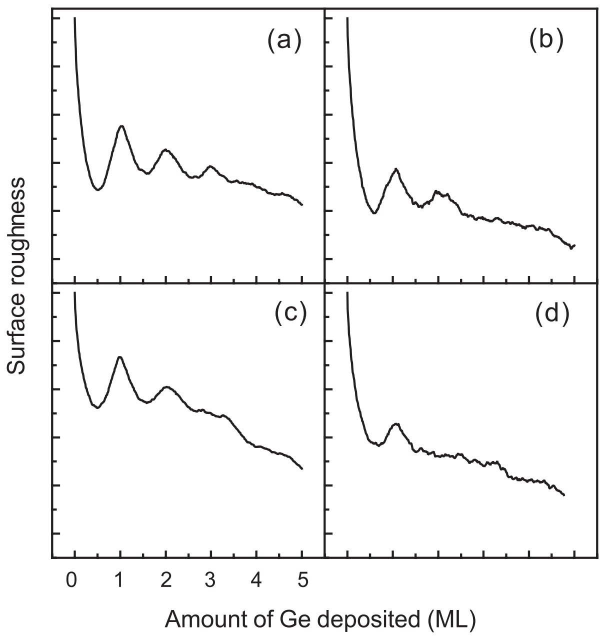

Fig. 4. The calculated surface roughness of Ge/Si structures obtained by KMC simulation as dependent on the amount of Ge deposited. The model included: (a) no ion beam effect; (b) generation of adatoms by ion beam; (c) enhancement of surface diffusion; (d) both (b) and (c).

adatoms, which can pile up on the top of existing 2D islands and nucleate 3D islands [15]. So, both mechanisms promote transition to 3D growth. The simulations including both mechanisms simultaneously have shown stronger effects on 2D–3D transition (Fig. 4d). In this case, critical thickness is decreased up to 1 ML.

5. Summary

Our experimental results demonstrated that Ge/Si(100) heteroepitaxy with pulsed low-energy ion beam action enables to create defect-free 3D Ge islands with small sizes and high density. Moreover, it provides a narrower size distribution of islands in comparison with conventional MBE. This is promising for potential applications in the technology of nanostructures. The results of KMC modeling have shown that both generation of adatoms by ion beam and enhancement of surface diffusion promote transition from 2D to 3D growth mode.

Acknowledgments

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