JETP Letters, Vol. 74, No. 5, 2001, pp. 267–269. Translated from Pis'ma v Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, Vol. 74, No. 5, 2001, pp. 296–299.

Self-Organization of an Ensemble of Ge Nanoclusters upon Pulsed Irradiation with Low-Energy Ions during Heteroepitaxy on Si

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Received July 30, 2001

Size distribution of Ge islands formed in the course of Ge heteroepitaxy on Si(111) was studied by scanning tunneling microscopy in experiments of two types: (i) conventional molecular beam epitaxy (MBE) and (ii) pulsed (0.5 s) irradiation with Ge ions of energy ≃200 eV at instants of time corresponding to a filling degree >0.5 for each monolayer. Experiments were performed at a temperature of 350°C. The pulsed ion-beam irradiation during heteroepitaxy leads to a decrease in the average size of Ge islands, an increase in their concentration, and a decrease in the root-mean-square deviation from the mean, as compared to the analogous values in conventional MBE experiments. © *2001 MAIK "Nauka/Interperiodica".*

PACS numbers: 68.65.Hb; 61.80.Jh; 68.37.Ef; 81.15.Hi

Self-organization of semiconducting nanostructures in the course of heteroepitaxy underlies the most promising method of forming ensembles of quantum dots [1–6]. At present, it is commonly accepted that the energy gain caused by the strain relief in 3D islands through the elastic relaxation in protrusion vertices is the key factor in the transition from a two-dimensional layered (2D) to three-dimensional island (3D) heteroepitaxial growth of pseudomorphic films. The islands are ordinarily formed due to the morphological instability of strained films in systems with a large (more than 2%) lattice mismatch between a film and a substrate, among which Ge/Si (4%) and InAs/GaAs (7%) are most familiar.

The self-organization (ordering) effects imply the appearance of islands with preferred characteristics: sizes, shapes, spacing between nanoclusters, and their 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.

Among the possible ways of improving island homogeneity in sizes, the following are most significant: (a) deviation from the singular surface plane of substrate (see [7] and references therein) and (b) nucleation synchronization [8].

It is generally believed that the nucleation of 3D islands occurs at the imperfections of the 2D layer (heterogeneous nucleation mechanism). Hence, the preliminary creation of the cluster nucleation sites is an efficient way for the island ordering control.

Nucleation synchronization is based on the idea of affecting the adatom supersaturation through a short-term increase in the density of molecular beam or short-term lowering of the substrate temperature. A cyclic variation of supersaturation in the course of growing each atomic layer underlies the optimized method of forming quantum-dimensional structures.

It has recently been found that irradiation with low-energy Ge ions during Ge heteroepitaxy on silicon stimulates the nucleation of Ge islands and reduces the critical thickness at which the 2D $\longrightarrow$ 3D transition occurs in a pseudomorphic Ge film [9].

The purpose of this work was to study the process of formation of an ensemble of Ge islands and their self-organization upon short-term irradiation of a pseudomorphic Ge film with its own ions during the heteroepitaxy on Si(111) from molecular beams. The (111) face provides the highest stability against the morphological instability that leads to the formation of 3D islands at the surface of a pseudomorphic film during the conventional epitaxy. This was precisely the reason why the surface with (111) orientation was chosen for studying ion irradiation effects.

Experiments were performed in an ultrahigh-vacuum chamber of an MBE setup equipped with an electron-beam evaporator for Si and an effusion cell (boron nitride crucible) for Ge. A system for the ionization and acceleration of germanium ions was designed and fabricated by us and placed over the crucible. The Ge molecular beam was ionized by the transverse electron

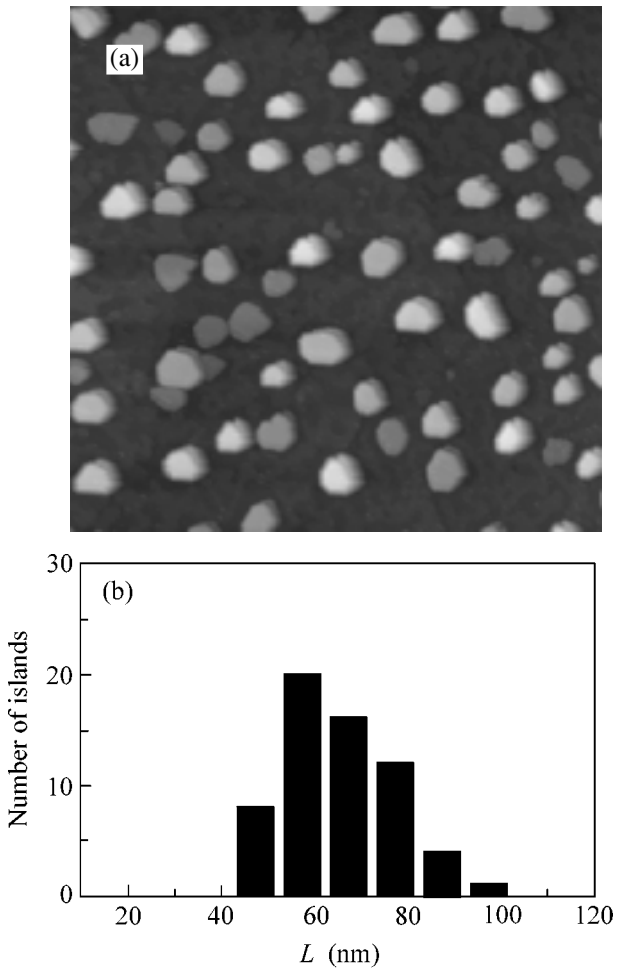

Fig. 1. (a) STM image of a 1000×1000 nm surface area, as obtained *ex situ* after Ge heteroepitaxy on Si(111) at a temperature of 350°C (five bilayers are deposited) and (b) size distribution of Ge islands.

beam. The ionization device allowed the degree of ionization of the Ge molecular beam to be varied from 0.1 to 0.5%. A pulsed accelerating voltage supply unit generated ion-current pulses with a duration of 0.5–1 s and an ion energy of 50–200 eV. The angle of incidence of the molecular and ion beams on the substrate was 54.5°. The analytical section of the chamber included a reflection high-energy (20 keV) electron diffractometer. The high-energy electron diffraction (HEED) pattern was detected by a digital video camera during the growth of germanium film, whereupon the data were fed into a computer and processed using a specially developed program package.

The growth velocity of the Ge film was ~1 bilayer (BL) in 10 s (one BL = 1.56×10^{15} at./cm^2), and the substrate temperature was varied in the range 200–400°C. Si(111) wafers with a misorientation angle less than 14′ were used as substrates. Prior to growing the Ge film, the wafers were subjected to high-temperature annealing followed by the growth of a buffer Si layer. The experiments on Ge heteroepitaxy on Si(111) were of two types: (i) conventional molecular beam epitaxy and (ii) molecular beam epitaxy with pulsed (0.5 s) irradiation by Ge ions of energy 200 eV at instants of time corresponding to a filling degree of ≥0.8 for each bilayer. The total amount of the deposited Ge was the same in both types of experiments.

The surface morphology was studied *ex situ* using scanning tunneling microscopy (STM), which allowed the real-space erect image of surface relief to be obtained with an atomic resolution. Measurements were carried out at room temperature in the dc tunneling current regime (bias 2 V, current 0.085–0.7 nA, feedback 3.76–5.31%). The STM images were processed using special programs for determining the size distribution of islands and their concentration.

The experimental results presented in Figs. 1 and 2 were obtained for a Ge flux density of 2.1×10^{14} at./cm^2 s, an ion-current density of 5.5×10^{11} at./cm^2 s, and a substrate temperature of 350°C. The amount of deposited Ge was equal to 5 BLs.

In the experiments of the first type, the concentration of Ge islands was found to be 7.3×10^9 cm^{-2}, their average size was $L = 63 \pm 9$ nm, and the size inhomogeneity was 14% (Fig. 1). In the second-type experiments, the island concentration was found to increase approximately twofold (1.2×10^{10} cm^{-2}), while the average size and size inhomogeneity diminished ($L = 43 \pm 4$ nm and 9%, respectively; Fig. 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, the ordering process being the result of the irradiation of a pseudomorphic Ge film with low-energy ions.

Of interest was to carry out some estimates characterizing the process of ion-stimulated nucleation and the growth of Ge islands. It should be noted, first of all, that the integrated Ge ion flux (total irradiation dose) was $\Phi = 0.5 \times 5 \times 5.5 \times 10^{11}$ at./cm^2 s $\simeq 1.4 \times 10^{12}$ at./cm^2. Every impact of the accelerated Ge ions gives rise to ~10 adatoms as a result of knocking out atoms from their regular sites in one or two nearest-to-surface monolayers of the pseudomorphic Ge film. At the sites of knocked-out atoms, a vacancy cluster (of monoatomic thickness) is formed at the surface of the growing layer [10, 11]. The Φ value exceeds the experimentally obtained concentration of Ge islands by almost two orders of magnitude. Consequently, under the experimental conditions chosen, most of the vacancy formations created at the surface of the Ge layer by ion irradiation are smoothed out (annealed) due to the interaction with adatoms coming from the molecular beam or generated by the irradiation. Only a small fraction of vacancy clusters, likely, become the sites of Ge-island nucleation. It is also not improbable that the Stranski–Krastanov mechanism of island formation makes a certain contribution in the second-type experiments with a chosen amount of deposited Ge.

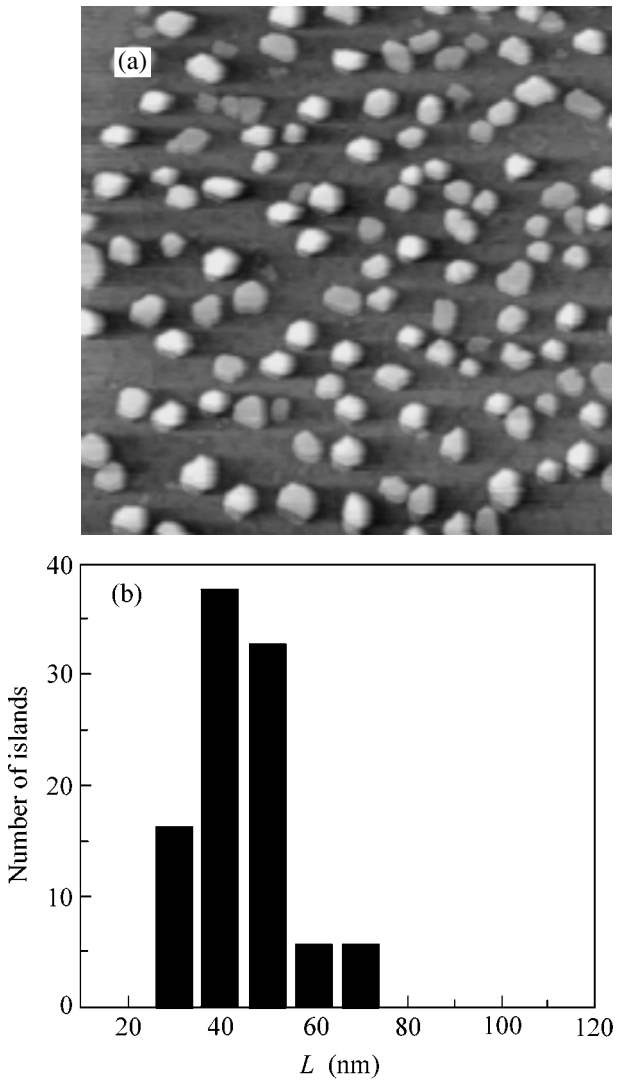

Fig. 2. The same as in Fig. 1 under conditions of pulsed (0.5 s) irradiation with low-energy (200 eV) Ge^+ ions during the heteroepitaxy.

The experiments with smaller amounts of deposited Ge give grounds to assume that the contribution from the ion-stimulated Ge-island formation dominates in the second-type experiments. For instance, the Ge islands are virtually not observed after growing 3.5 BL under conditions of conventional Ge heteroepitaxy on Si(111) (the Ge layer thickness is smaller than critical). The switching on of the pulsed irradiation in the course of heteroepitaxy (second-type experiments) gives rise to the islands with a concentration of 4.3×10^{-10} cm^{-2}.

The size ordering of the Ge islands is, most likely, caused by the following factors: (a) a single-event (within a pulse duration) nucleation of Ge islands after completing the growth of each layer followed by the enlargement of the islands and (b) a decrease in the growing layer roughness under the pulsed action of the Ge^+ ion beam, with an energy of about 200 eV, just before the growth oscillation maximum [9], likely because of an increase in the adatom diffusivity as a result of the ion-stimulated Ge(111)-surface reconstruction, namely, the transition from the (7×7) to the (5×5) superstructure.

We are grateful to V.A. Kudryavtsev for conducting experiments on heteroepitaxy and to S.A. Tiĭs and I.G. Kozhemyako for the STM measurements. This work was supported by the Russian Foundation for Basic Research (project no. 99-02-17196) and the program “Surface Atomic Structures” (project no. 4.2.99).

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Translated by V. Sakun