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ATOMIC STEPS ON THE SINGLE CRYSTAL SURFACE DURING EPITAXY, SUBLIMATION AND GAS REACTION

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Structural transitions on silicon surfaces during growth and oxygen exposure were investigated by in situ ultra-high vacuum reflection electron microscopy and atomic force microscopy. Consequent stages of gold three dimensional island ordering on silicon (111) surfaces were visualized over a wide temperature range of the substrate. Initial stages of surface oxidation and thermal etching were analysed in details.

Introduction

Surface structural transformations can be described in the frames of elementary acts of the point defects such as adatoms and vacancies. Thus clear understanding of behavior of adatoms and vacancies on crystal surface provides possibility to control surface atomic structure and morphology. Also it allows creating, so-called, low-scale systems or nanostructures which can demonstrate quantum phenomenon of charge carriers. Unfortunately a direct observation of the point defects on the surface is not possible in the most cases especially at the high temperatures. However behavior of point defects on the surface can be understood on the base of analysis of atomic steps which can be studied from *in situ* experiments at conditions far from equilibrium. In this paper we summarize recent results of atomic step analysis on the silicon surface.

Experiments were performed by combination of two surface-sensitive methods: ultrahigh vacuum reflection electron microscopy (UHV-REM) and atomic force microscopy (AFM) at the atmospheric conditions. UHV-REM was performed to carry out *in situ* experiments on the silicon surface. Scanning probe microscope (Solver P-47H, NT-MDT) was used for AFM examination of the sample being lifted from an UHV chamber of a reflection electron microscope.

Gold ordering

Analysis of the three-dimensional (3D) gold island nucleation, migration and coalescence on the silicon substrates with regular steps, step bunches and step anti-bunches was carried out to understand the self-organized processes on the stepped silicon surface. Also the influence of the heating electric current on the gold island formation was analyzed because electromigration is responsible for gold atom behaviour on the silicon surface [1].

Typical stages of surface modification during gold deposition are described below. Deposition 0.35ML of gold at 540^{o}C initiated partial inverting the contrast of monatomic steps in the REM-image from a dark to a bright one [2]. At the same time there were no changes in RHEED patterns after the deposition except of intensity decaying of 1/7 reflections in spite of the fact that the deposited amount at the crystal temperature is enough to initiate the (7x7)⇔(5x2) reconstruction [3]. The last could be explained by high rate of gold deposition [4]. The change of the step contrast seems to be associated with the incorporation of gold adatoms into monatomic steps. The deformation field of the crystal lattice around the edge of the monatomic step is probably changed by the gold adsorption, modifying the Bragg conditions for reflecting electrons.

The 3D-islands of gold appeared on the surface during further gold adsorption. These islands covered almost uniformly both the terraces and the step bunches. Chemical composition of the islands was not measured but Homma et al [5] estimated as 20% of silicon and 80% of gold at the similar condition. The average size of the 3D gold islands was determined by precision computer treatment of REM digitized images. Within the accuracy of our measurements, the average size of the 3D gold islands was equal to 50nm, assuming a spherical shape of the gold islands as shown in refs. [6,7].

Further gold deposition up to 1.4 ML resulted in an increase of the average island size up to 100 nm. The number of islands was constant and monatomic steps were not moved during deposition. It means that the deposited gold was incorporated mainly in the gold islands. Subsequent annealing of the sample at 540^{0}C for several minutes initiated the appearance of additional reflections in the diffraction pattern corresponding to the ($\sqrt{3}$ x $\sqrt{3}$) surface reconstruction.

REM observations revealed a dominating migration of the gold islands on the silicon (111) surface in the step-up direction. The direction of island migration was independent on the direction of the DC heating of the crystal. Although islands moved faster when the heating current was flowing in the step-down direction. The smaller gold islands moved faster than the larger ones due to smaller interaction energy with the silicon substrate [8]. During annealing the 3D-islands showed a tendency to orient along step bunch. Additional annealing of the sample at 650^{0}C led to an increased intensity of the ($\sqrt{3}$ x $\sqrt{3}$) reflections in the RHEED pattern. An essential redistribution was observed for the 3D-islands parked on the top part of bunch. One can see the chain of gold islands along step bunch. Fig.1 shows the 3D-islands redistribution on the surface containing step bunches and singular terraces between them after thermal annealing. The black spots in the REM image are 3D gold islands however the grey strips are two bunches.

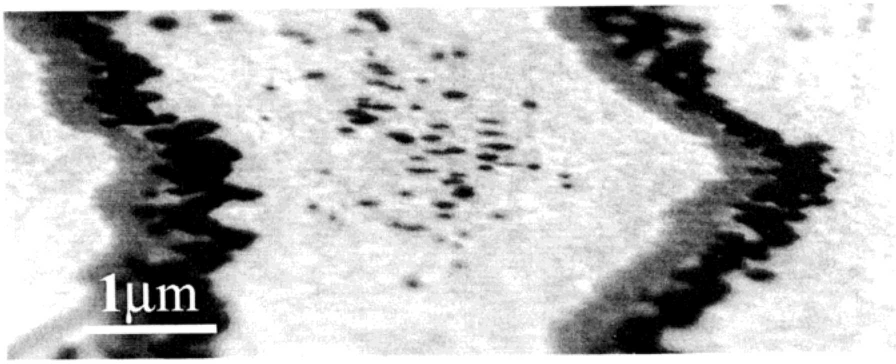

Fig.1 Typical REM-image of the silicon (111) surface after gold deposition 2ML at 540^{0}C.

REM observation of the surface with regularly distributed monatomic steps revealed also preferential drift of the gold islands in the step-up direction independently on the direction of the electric current during annealing. At the higher temperature the gold islands were uniformly distributed on the surface and no interaction between the gold islands and single monatomic steps was registrated. It is notable that REM observation showed the gold islands drift in the step-up direction when the alternative current heating is applied too. The same step-up direction of the island drift was observed during gold deposition on the surface morphology containing simultaneously two systems of monatomic steps with opposite sign of steps. Such surface morphology was realized during formation of bunches and anti-bunches on the silicon surfaces at sublimation [9]. The gold islands in bunch and anti-bunch moved in the opposite directions because the step-up direction for steps in bunches and anti-bunches are opposite. Direction of the electric current for steps in bunch and anti-bunch is opposite also, but the gold islands both on step bunch and anti-bunch drift in the step-up direction. As result, two parallel gold island chains with the distance between them equalled to the distance between bunch and anti-bunch were formed during heating treatment. The experiments with bunch- anti-bunch morphology also confirm that electromigration effect does not being responsible to the 3D gold island drift processes on the silicon (111) surface during thermal annealing.

Thermal oxygen etching

To analyse role of vacancies on the surface, the interaction of molecular oxygen with the silicon surface was examined in the wide temperature and oxygen pressure ranges. It is known that an oxygen-silicon interaction drastically depends on the substrate temperature and oxygen pressure [10]. At low temperatures and high oxygen flux, the silicon surface passives by a thin film of dioxide, whereas the surface was thermally etched at high temperature and low oxygen pressure. REM observation showed step movement in the step-up direction at low oxygen pressure. Monatomic step movement during oxygen treatment occurs due to nucleation, diffusion and interaction of vacancies with monatomic steps [11]. REM visualization showed that the step rate increased with the flux of oxygen and was linearly dependent on the width of neighboring terraces [12]. Steps with wide terraces move faster than steps with narrow terraces according to the classical BCF-theory. A linear dependence of step motion on the terrace width indicates that the vacancy diffusion length is larger than the interstep distance. This suggests that the main diffusion species on the silicon (111) surface during thermal etching is surface vacancies.

At high supersaturation of the vacancies on the surface the negative island nucleation and their enlargement occur during oxygen exposure. Fig.2 shows typical AFM image of the silicon surface with two-dimensional negative islands. Dark spots of triangular shape correspond to negative islands nucleated on the terrace during thermal etching of the silicon surface. Direct measurements of the depth of negative islands provided by AFM coincide with the monolayer height. It is notable the existence of a denuded zone free from islands was observed along atomic steps. All vacancies formed inside this zone disappeared through interaction with monatomic steps. Moreover, negative island nucleation was not observed at narrow terraces. Evidently, the occurrence of this two-dimensional mechanism of silicon thermal etching needs the terrace width to be more than twice the surface diffusion length of vacancies. Further exposure of the silicon surface to oxygen causes enlargement of negative islands and step motion in the step-up direction. The interaction of the moving step with islands leads to drastic changes of step shape, and islands disappear.

Nucleation of two-dimensional negative islands means removal of atom from top monolayer of the silicon surface. During etching process the intensity of the reflected beam of electrons will change, as the reflection coefficient of the surface changes from maximum (clean surface) up to a minimum (islands will take half of the surface area). Thus, oxygen exposure caused a periodic intensity change of the reflected electron beam from the silicon (111) surface. One period of oscillations of the reflected beam should correspond to the removal of one monolayer of substrate atoms. The period of oscillations does not depend on the substrate temperature at the range of 540^0 - 835^0C and it is proportional to the oxygen flux. It means that oxygen flux caused kinetic limitation of the surface reaction. It should be pointed out that the period of RHEED intensity oscillations depends on the oxygen pressure, while the temperature range of the two-dimensional mechanism of thermal etching of the silicon surface was related both to the oxygen pressure and the interstep distance.

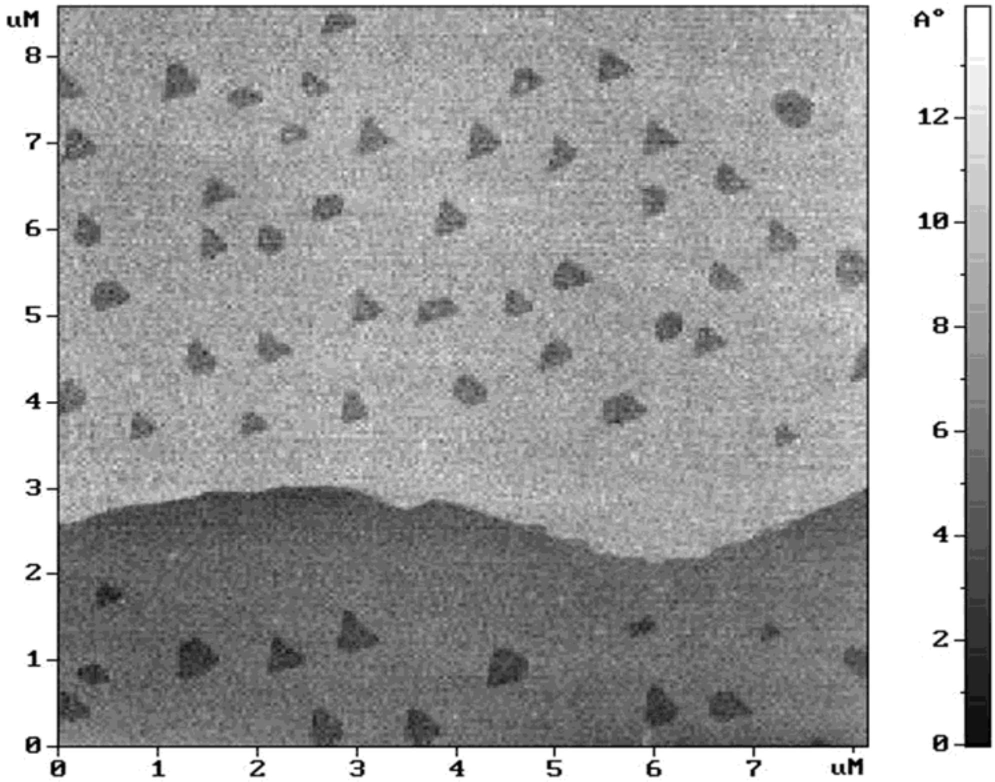

Fig. 2. Typical topographic AFM-image of the silicon (111) surface with negative islands.

Regular monatomic steps contain the same surface distribution during oxygen exposure. However step bunches were transform to the regular one at the oxygen atmosphere, where generation of surface vacancies occurs. Behavior of monatomic steps under supersaturation of surface vacancies is studied in details.

Step movement during thermal etching of the silicon surface by oxygen was accompanied by the roughening of step edges. That should occur according to the competition of two effects: free energy minimization and entropy increase. A straight monatomic step is unstable against step meandering. We carried out statistical analysis of step shape fluctuations and interstep distances on the silicon (111) surface by both UHV-REM and AFM. For fluctuation analysis the both separate step, regular step and step bunch were under consideration. At the same thermodynamical conditions the maximal amplitude of step fluctuation was registrated for monatomic step detaching from step bunch. Inside of step bunch step fluctuations have minimal amplitude due to restriction of step-step interaction. An increasing the amplitude of step fluctuations near bunch is result of interaction between monatomic step and step bunch by diffusion and strain fields. The same increasing of step fluctuation amplitude was found for atomic steps, which were detaching from bunch during homoepitaxial growth (Fig.3). An amplitude analysis of these fluctuations allows to deduce number of parameters for the adatoms, monatomic steps and surface such as step edge stiffness, diffusion constants and step-step interaction.

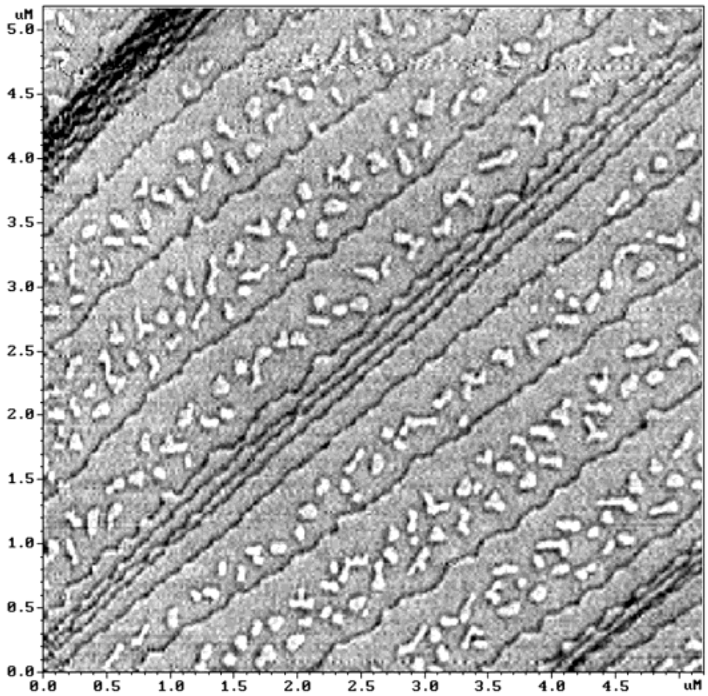

Fig.3. AFM image (phase contrast) of the stepped silicon (111) surface with step bunches after 0,2 ML silicon deposition at 740^{0}C.

Conclusion

In situ UHV-REM investigation was applied to study the initial stages of gold adsorption and thermal etching in an oxygen atmosphere on the silicon (111) surface at various crystal temperatures. REM observation revealed drifting of the gold 3D-islands in the step-up direction during thermal annealing. The same direction of island drift was seen for both DC directions while the drift rate was higher for heating current flowing in the step-down direction. These results provide new opportunities for better understanding metal-semiconductor growth and also for controlling self-assembled structural processes on the silicon surface. The main stages of surface vacancy formation on the silicon (111) surface under oxygen treatment were also visualized. An influence of surface vacancies on stability of monatomic step distribution was analysed.

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