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MECHANISMS OF SURFACE EVOLUTION DURING THE GROWTH OF UNDOPED NANOSILICON FILMS

The thickness dependence of the surface roughness and the grain size of nanosilicon films, produced by low-pressure chemical vapour deposition, has been found, by using atomic force microscopy. A correlation between the surface roughness, grain size, and transformation of a film structure from the equiaxial structure into a fibrous one is established. Possible mechanisms of surface evolution are analyzed.

Keywords: nanosilicon films, surface roughness, grain growth, mechanism of grain growth, atomic force microscopy.

1. Introduction

The wide application of polycrystalline silicon films in microelectronics (in MOS structures and others) and solar power engineering makes it necessary to study their structure. It is so, because the latter is responsible for all film properties important from the viewpoint of their practical application [1–4].

Defects of the crystal structure of silicon films are known to considerably influence the efficiency of solar cells [2, 3]. In particular, the character of a defect film structure governs the uniformity of the doping impurity distribution in them and, accordingly, the stability of photogeneration properties of the films and determines the effective ways for the charge carrier transport. Therefore, from the viewpoint of enhancing the efficiency of silicon materials, the researches aimed at improving the film structure are actual.

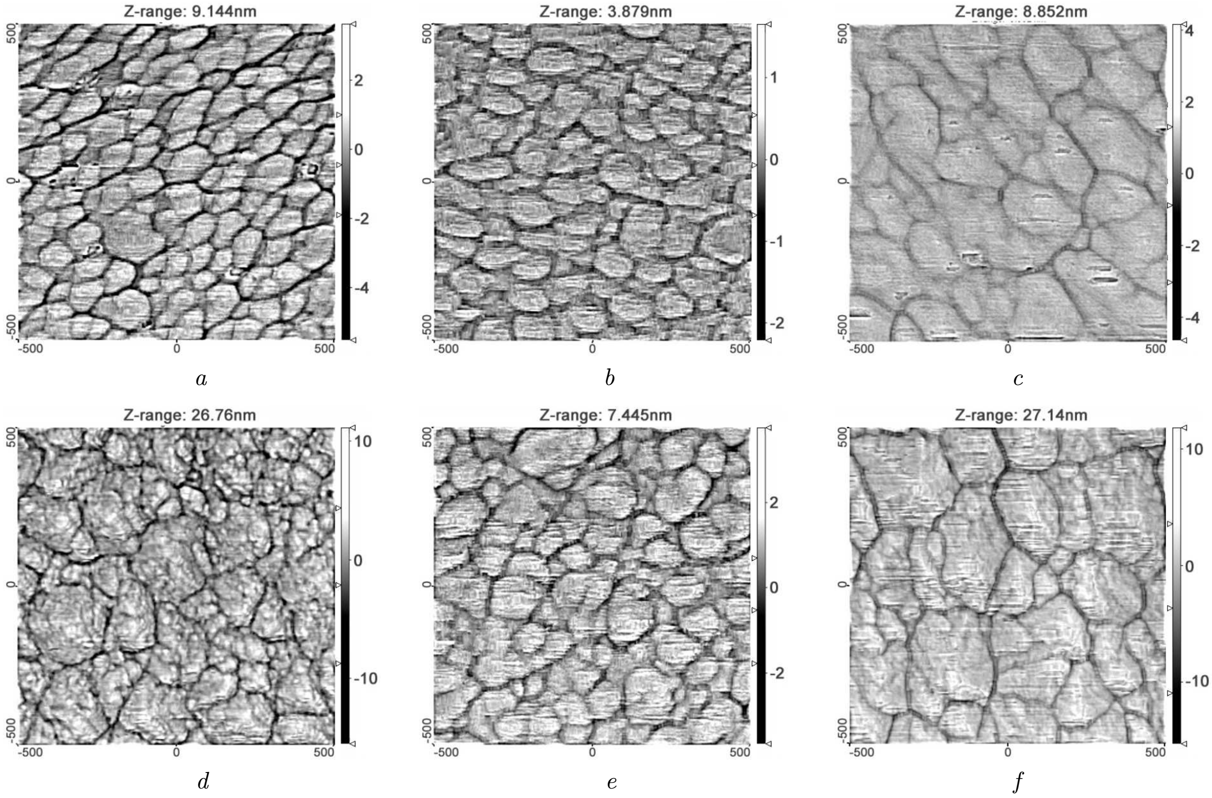
The structure of silicon films, depending on deposition conditions and subsequent technological treatments, was studied in works [5–14] in detail. Various structural modifications of the films were revealed – namely, equiaxial, fibrous, and dendritic – and conditions of their existence were determined [5]. In addition, the mechanisms of normal and abnormal grain growth in the films under annealing were found [5–7], and the phase modifications in films with fibrous and dendritic structures were studied [8, 9]. A significant attention was paid to researching the grain boundary structure of silicon films, in particular, the grain boundary faceting and the faceting of tips of twin plates in the interior of grains under annealing [10–13]. With regard for the practical applications of silicon films in microelectronics, the influence of the type of their structure on the oxidation processes in them were studied [14]. Almost all mentioned researches of a silicon film structure concern films 500–2000 nm in thickness, because just such films are used in microcircuit components.

However, the structure of nanocrystalline silicon films remains almost not studied. Only in work [15], the influence of the annealing on a microstructure of undoped polysilicon films 50 nm in thickness was analyzed. It was found that, depending on the annealing temperature, grain growth occurs through nondiffusive processes (glide of grain boundary dislocations) or diffusive processes (climb of grain boundary dislocations).

This work aims at researching the mechanisms of surface evolution in undoped nanocrystalline silicon films, as well as their dependence on the film thickness, by using the atomic force microscopy (AFM) methods.

2. Experimental Technique

Nanocrystalline silicon films were produced by low-pressure chemical vapour deposition from a silane/argon mixture. Single-crystalline silicon wafers with a SiO_2 oxide layer about 100 nm in thickness were used as substrates. The deposition temperature was equal to 630 °C. The thickness of silicon films varied from 10 to 100 nm.



a *b* *c*

d *e* *f*

Fig. 1. AFM topographic images of the surfaces of nanosilicon films with various thicknesses: 10 (*a*), 50 (*b*), 70 (*c*), 75 (*d*), 85 (*e*) and 100 nm (*f*)

The film surface was studied by means of AFM. The film surface images were obtained using a scanning atomic force microscope NanoScope IIIa in the periodic contact mode (Tapping mode) with a silicon probe of 10 nm edge radius.

3. Results and Their Discussion

Our previous transmission electron microscopy (TEM) and AFM researches showed that nanosilicon films have a homogeneous fine-grain structure, which does not change if the film thickness grows, whereas the surface undergoes considerable modifications [16]. Fine grains combined in large grain agglomerates, which form hillocks on the film surface. AFM researches showed that both the height and the horizontal size (the grain size|) of the hillocks change substantially, as the film thickness increases.

The two-dimensional AFM topographic images (Fig. 1) testify that the film thickness growth is accompanied by a modification of the film grain boundary structure and a considerable growth of the dimensions of grain agglomerates (hillocks). For instance, for a nanosilicon film 10 nm in thickness, the average grain size amounts to 24 nm, whereas this parameter for a film 100 nm in thickness reaches a value of 195 nm. The dependence of the average grain size on the film thickness is plotted in Fig. 2. One can see that, for films with the equiaxial structure (at a film thickness less than 70 nm), a moderate growth of the average grain size is observed. After the transition to the fibrous structure (at a film thickness of more than 70 nm), a considerable growth of the average grain size takes place.

The process of grain growth in nanocrystalline materials has specific features that distinguish it from the grain growth in coarse-grained polycrystals [17]. In polycrystals, the grains usually grow following the standard mechanism, by means of the migration of grain boundaries and their merging (coalescence) [17]. Those processes result in an increase of the average grain size and a reduction of the number of grains in the polycrystal. The grain growth through the migration and the merging of grain boundaries is usually considered as a unique mechanism of grain growth in coarse-grained polycrystals. The difference between the processes of grain growth in nanocrystalline materials and in ordinary polycrystals is associated with specific structural features of nanocrystalline materials, in particular, nanoscopic grain sizes and high densities of grain boundary ensembles and grain boundary triple junctions. Those structural features are responsible for the efficiency of the following factors that affect the grain growth [17]: (i) the inhibitory effect of triple junctions on the migration of grain boundaries, (ii) excess free volume as a result of the high density of ensembles of grain boundaries and their triple junctions, (iii) stabilization of a nanocrystalline structure owing to the elastic interaction responsible for the formation of low-energy defect structures in nanoscopic grain boundaries, (iv) a high average energy density (per unit volume) in the nanocrystalline material, which is connected with a considerable number of ensembles of grain boundaries and their triple junctions, and (v) the grain growth by means of the crystal lattice rotation in nanoscopic grains. The latter mechanism is connected with rotations of the crystal lattice in grains, which results in the disappearance of grain boundaries and the merging of neighboring grains. It should be noted that the indicated factors take place in coarse-grained polycrystals as well. However, their action in polycrystals is usually not pronounced and efficient.

The distributions of grain sizes in nanosilicon films with various thicknesses, which were determined from AFM topographic images, are depicted in Fig. 3. One can see that, for films 50 nm in thickness, the distribution has a one-mode character (Fig. 3, *a*). For a film thickness of 70 nm, the distribution has a two-mode character (Fig. 3, *b*). If the film thickness equals 85 nm, three maxima are observed in the distribution (Fig. 3, *c*). It is worth noting that the peak in Fig. 3, *a* corresponds to a grain size of 22 nm. In Fig. 3, *b*, the second peak appears, which corresponds to 44 nm. In Fig. 3, *c*, three peaks are observed at 30, 60, and 90 nm. It is evident that the maxima observed in Figs. 3, *b* and 3, *c* correspond to the increase of the grain size by a factor of two and three, respectively, in comparison with the initial one (Fig. 3, *a*). This fact testifies in favor of the mechanism of grain size growth by means of the rotation of grain's crystal lattice [17]. The reduction in the energy of grain bound-

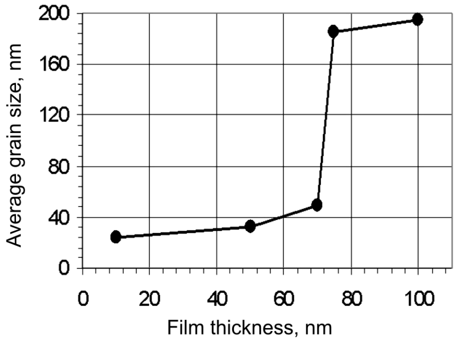
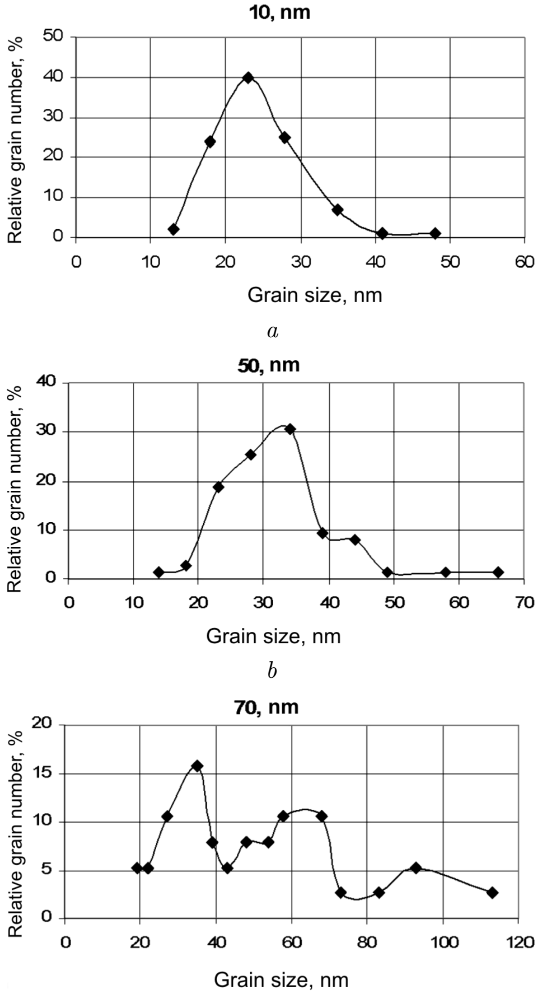


Fig. 2. Dependence of the average grain size on the nanosilicon film thickness



a

b

c

Fig. 3. Grain size distributions for films with various thicknesses

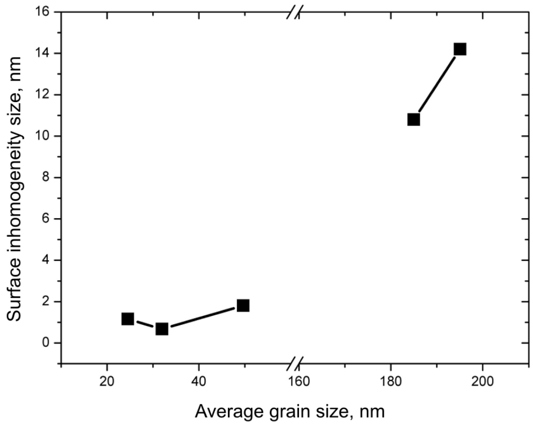


Fig. 4. Dependence of the surface inhomogeneity size on the average grain size in nanosilicon films

aries, when the parameters of a disordering change, is a driving force for rotations of the crystal lattice in grains. The rate and the geometrical parameters of a lattice rotation are governed by the grain boundary diffusion and the processes of dislocation transformations at the grain boundaries. In particular, if the grain sizes decrease, the crystal lattice rotations governed by the grain boundary diffusion become more intense [17]. The rotation of the crystal lattice in a grain results in the disappearance of the grain boundary and the merging of neighboring grains. So, this is a micromechanism of grain growth, which is alternative to the standard micromechanism of grain growth through the migration and the merging of grain boundaries.

The dimensions of surface roughness and grains are known to be related to one another. The researches on metal films, in particular, Au films, showed that the evolution of surface roughness is accompanied by changes in both the orientations and the average sizes of grains [18]. In this case, one may observe either an increase or a reduction of the surface roughness sizes, as the grain sizes grow. In particular, while studying Au films at the annealing, it was found that, at low annealing temperatures, the sizes of surface roughness decrease, as the grain sizes increase. If the temperature and the grain sizes grow further, the dimensions of roughness remain constant and start to increase at sufficiently high temperatures. This character of the evolution of the structure and the morphology of the film surface results from a competition between two factors responsible for structural modifications, namely, the mobility of grain boundaries and the surface diffusion. The mobility of grain boundaries prevails at low temperatures, whereas the surface diffusion is responsible for the adjustment of a surface microrelief in correspondence to the grain sizes at high temperatures.

For nanosilicon films, the dependence of the surface inhomogeneity dimensions on the average grain size is plotted in Fig. 4. The growing character of this dependence testifies that the surface diffusion is the mechanism of surface evolution.

4. Conclusions

(i) As the thickness of undoped nanocrystalline silicon films grows from 3 to 100 nm, the sizes of surface roughnesses increase, as well as the grain sizes.

(ii) A drastic growth in the dimensions of microrelief roughness on the film surface and in the grain size at film thicknesses larger than 70 nm correlates with the change of the film structure type from the equiaxial type to a fibrous ones.

(iii) Rotation of the crystal lattice of grains is the mechanism of grain growth with increasing the film thickness.

(iv) Surface diffusion may be a probable mechanism of surface microrelief modification, as the film thickness increases.

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МЕХАНІЗМИ ЕВОЛЮЦІЇ ПОВЕРХНІ ПРИ РОСТІ НЕЛЕГОВАНИХ НАНОКРЕМНІЄВИХ ПЛІВОК

Резюме

Методом атомної силової мікроскопії досліджено вплив товщини нелегованих нанокремнієвих плівок, що отримані методом хімічного осадження в реакторі зниженого тиску, на характеристики їх поверхневого мікрорельєфу. Встановлена кореляція між зростанням розмірів неоднорідностей мікрорельєфу поверхні плівок та зміною типу структури плівок від рівноосьової до волокнистої. Проаналізовано імовірні механізми еволюції поверхні нанокремнієвих плівок.