Functional Materials 13, No.2 (2006)

Features of special joints of grain boundaries in polysilicon films of equiaxial and dendritic structures

N.G.Nakhodkin, N.P.Kulish, P.M.Lytvyn, T.V.Rodionova*

T.Shevchenko Kyiv National University,
64 Volodymyrska St., 01033 Kyiv-033, Ukraine
*V.Lashkaryov Institute of Semiconductor Physics, National Academy of Sciences of Ukraine, 41 Nauky Ave., 03028 Kyiv, Ukraine

Comparative analysis of special grain boundary joints in polysilicon films with equiaxial and dendritic (undoped and phosphorus-doped) structure, prepared by low-pressure chemical vapor deposition, has been carried out using atomic force microscopy and transmission electron microscopy. The formation mechanisms of special grain boundary joints have been analyzed for different film structures. The effect of phosphorus on formation of grain boundary joints has been analyzed.

Методами атомной силовой микроскопии и просвечивающей электронной микроскопии проведен сравнительный анализ специальных стыков границ зерен в поликристаллических пленках кремния с равноосной и дендритной структурой (нелегированных и легированных фосфором), полученных методом химического осаждения из газовой фазы в реакторе пониженного давления. Рассмотрены механизмы формирования специальных стыков в пленках разных структурных модификаций. Проанализирована роль фосфора в формировании стыков границ зерен.

Polycrystalline silicon films are used widely as the gate electrode material for metal-oxide-semiconductor (MOS) devices, the contact material for various devices and other applications in microelectronics. The film structure is known to define significantly the mechanical, optical, and electrical properties thereof.

Joints of grain boundaries are a characteristic feature of polycrystalline film structure and are defined as a peculiar kind of linear defects [1–3]. It is evident that the joints influence the film structure formation and properties of the films. This effect is different for various joints and is structure-dependent. Usually, the grain boundaries of general type interact to each other forming in equilibrium triple 120° joints. Special triple joints result from interaction of special grain boundaries. Studies of special grain boundaries have shown that these boundaries may form stable multiple (fourfold and fivefold) joints of grain boundaries [4, 5]. Of particular interest is the investigation of special grain boundary joints, because these joints possess a low energy, weak trend to impurity adsorption, low diffusive penetration and so on. As compared to joints of general type, the special joints are more stable, because mobility of special grain boundaries is low [4, 5].

It is well known [6] that in polysilicon films, the relative amount of special joints is defined by the film structure type (equiaxial, dendritic, fibrous). However, the grain boundaries and their joints in polysilicon films of different structure modifications are not considered in detail. It was shown for dendritic polysilicon films that the number of special grain boundaries is influenced by the presence of phosphorus [7]. In this work, a comparative study of special grain boundary joints and surface roughness in polysilicon films of equiaxial and dendritic structures have been carried

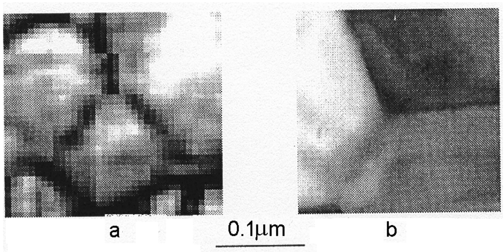


Fig. 1. AFM image (a) and TEM micrograph (b) of a triple grain boundary joint in a polysilicon film.

out by means of atomic force microscopy and transmission electron microscopy.

The polysilicon films were prepared by low-pressure chemical vapor deposition. Films were deposited on thermally oxidized (0.1 µm oxide thickness) (100) single crystal silicon wafers. The films were 0.5 µm thick. Some films were doped with phosphorus. The dopant concentration was 10^{21} cm^{-3}. Depending on formation conditions, the films had different structures: equiaxial or dendritic. The equiaxial structure was studied in phosphorus-doped polysilicon films deposited at 630°C and annealed at 1150°C in nitrogen atmosphere. Dendritic structure was observed in undoped and phosphorus-doped polysilicon films deposited in amorphous phase (the deposition temperature 560°C) and annealed at temperature ≤1000°C. The film structure depending on the preparation conditions and post-treatments is considered in [8] in detail.

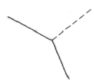
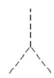
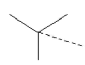
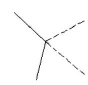
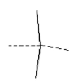
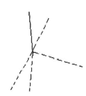
Atomic force microscopy (AFM) has been used to obtain a set of statistical data in studies of grain boundary joints. The film surface images were obtained using a scanning atomic force microscope NanoScope IIIa in the periodic contact mode (Tapping Mode) with a silicon probes of 10 nm edge radius. 264 grain boundary joints have been analyzed for equiaxial films (films with equiaxial structure), 391 joints for undoped dendritic films, and 328 joints for phosphorus-doped dendritic films.

More detailed information concerning the nature of grain boundary joints, especially multiple joints, and grain boundaries has been obtained from transmission electron microscope (TEM) data. The samples for TEM investigations were prepared by chemical etching. Fig. 1 shows the example of images of triple grain boundary joint obtained using AFM and TEM.

The AFM studies have shown (Fig. 2) that triple grain boundary joints dominate throughout the entire examined films. However, as is seen from Table, most of triple joints in undoped dendritic films are special joints, that is, these joints are composed of special grain boundaries (joints such as 3b 3c (the description of joint types is considered [6] in detail)). At the same time, in equiaxial films and in phosphorus-doped dendritic films, most of triple joints are formed by grain boundaries of general type. The number of multiple special joints is higher in undoped films, too.

A more detailed information concerning the nature of grain boundary joints has been obtained from TEM studies. Unfortunately, this method gives no way to investigate as large area as with the use of AFM. Thus, not all joint types which being observed in AFM-images are presented in Fig. 3. However, Fig. 3 demonstrates that

Table. Relative number of triple and multiple special grain boundary joints in polysilicon films of equiaxial and dendritic structure, per cent

Structure type	Types of grain boundary joints and their schematic representation						
	Triple			Multiple			
	3a 	3b 	3c 	4a 	4b 	4c 	5 
Equiaxed	89.8	2.7	2.7	0.4	3.8	0.4	0.4
Dendritic (phosphorus-doped)	80.2	10.1	3.0	1.8	2.8	1.5	0.6
Dendritic (undoped)	19.9	24.6	18.7	7.9	15.3	9.5	4.1

--- — grain boundary of general type,
- - - — special grain boundary

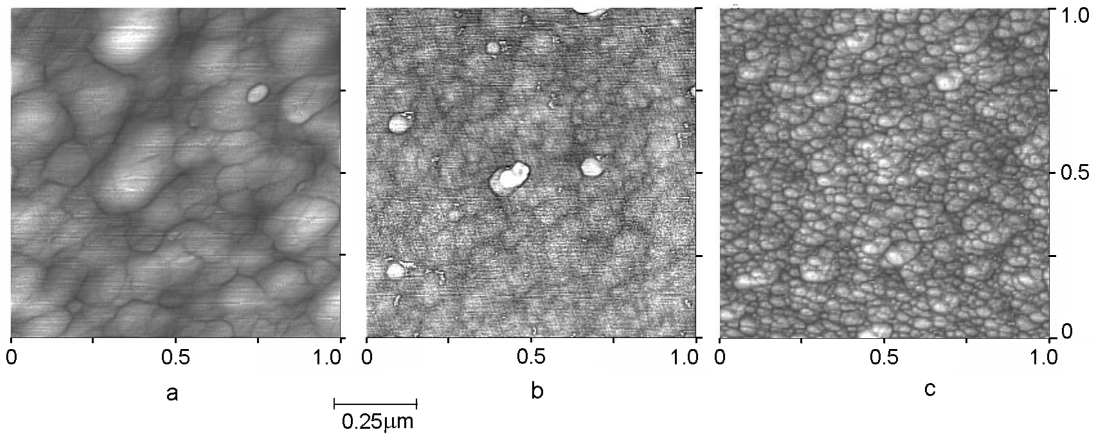


Fig. 2. AFM-images of polysilicon films: equiaxial (a); undoped dendritic (b); phosphorus-doped dendritic (c).

in equiaxial polysilicon films, the joints consist of large-angle grain boundaries (such boundaries appear as solid lines in TEM images, Fig. 3,a–d). At the same time, in dendritic films, both undoped and phosphorus-doped, some part of the multiple joints (in particular 4a, 4c) are formed by low-angle grain boundaries with dislocation structure (dashed lines in TEM images, Fig. 3g, 3i).

There are several formation mechanisms of special grain boundary joints in polycrystalline films. First, the grain boundary splitting is possible. In materials with low energy of stacking faults, to which silicon belongs also, the grain boundary splitting is observed often enough [1]. The grain boundaries with orientation defined by sum of turns resulting in formation of low-energy grain boundary, may become split. Second, a random meeting of special grain boundaries may occur. In the third case, special grain boundary joints may arise from multiplex twinning. The multiplex twinning is a characteristic feature of a face-centered crystal with low energy of stacking faults. Interaction of twins of various orders may result in multiple grain boundary joints formation.

The morphology analysis has shown that all three mechanisms of special grain boundary joint formation take place in both equiaxial and dendritic polysilicon films. However, each structural modification has its own peculiarities. In case of equiaxial structure, it is possible to suggest, by analogy with metals [1], that most probable mechanism of special joint formation is splitting of grain boundary under annealing.

As is shown in [9], the dendritic structure of polysilicon films results from multiple twinning. The dendritic grains are complexes of the twins with $\sum 3^n$ boundaries. Interaction of twin boundaries results in formation of special grain boundary joints. For example, it is known [4] that when two $\sum 3$ boundaries spaced at different (111) planes are in contact, the third boundary in the joint always is a $\sum 9$ boundary. In this case, special triple joint of 3c type is formed. Further interaction of $\sum 9$ boundary with next $\sum 3$ boundary, nonparallel with first two ones, causes $\sum 27$ boundary, and so on. In addition, a large number of twin boundaries in dendritic films result in larger probability (as compared to equiaxial film structure) of their random meeting and special joint formation.

The distinction in the number of special grain boundary joints and grain boundary joints of general type in undoped and phosphorus doped polysilicon films is caused by influence of phosphorus on the film structure. The presence of phosphorus is known [8, 10] to be accompanied by increasing number of charge vacancies that results in increased grain boundary mobility and the dislocation motion rate. Motions of dislocations are immediately connected with formation of grain boundaries and grain boundary joints. The dislocation structure of grain boundary (Fig. 3g,3i) in dendritic films indicates that internal stresses take place in the films. As is known [10], the phosphorus-doped dendritic films are characterized by significantly lower internal stress level as compared to undoped films. It is believed that the increased dislocation

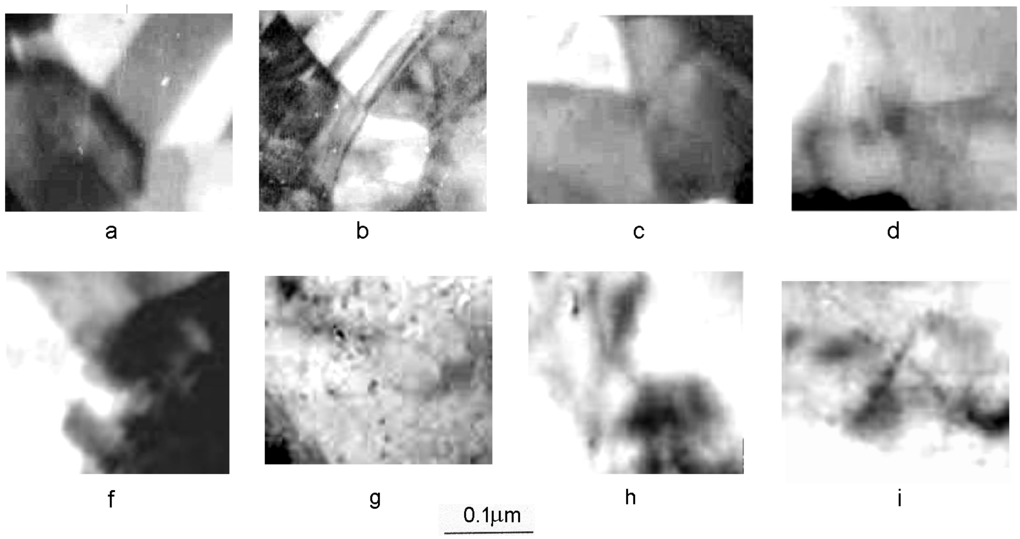


Fig. 3. TEM micrographs of some grain boundary joints in polysilicon films: 4a joint in equiaxial structure (a); 4a joint in dendritic structure (b) (dash lines point to dislocation structure of grain boundary).

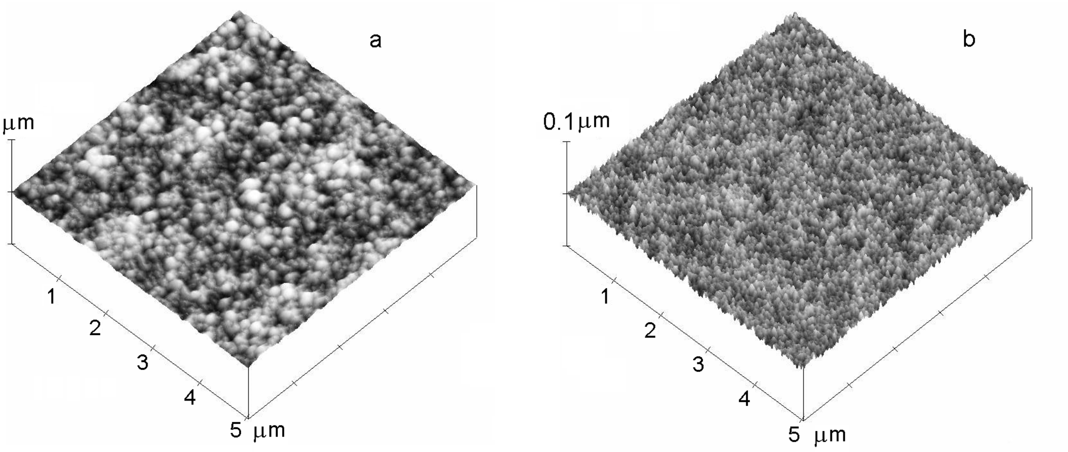


Fig. 4. Three-dimensional AFM images of surface of polysilicon films with equiaxed (a) and denfritic (b) structure.

mobility promotes stress relaxation in the films. This process is followed by a realignment of grain boundary, namely, low-angle grain boundary having long-range field of elastic stresses are transformed to large-angle boundaries where the stress approaches zero. This is due to the fact that principle of boundary is changed. For large-angle boundaries, it consist in local displacements of lattice points, which cannot be described by the Hooke law. Hence, the stress relaxation can be considered as driving force of grain boundary realignment. In this case, the mechanism of grain boundary realignment in dendritic polysilicon films may be thought to be dislocation gliding, as for metals. The increased number of large-angle boundaries results in increasing number of general type triple joints (such as 3a, Fig. 3a, 3f) in phosphorus doped dendritic polysilicon films. As for surface roughness, the AFM studies have shown (Fig. 4) that in equiaxial polysilicon films the surface roughness is nearly three times more than in dendritic ones.

To conclude, the AFM and TEM studies of special grain boundary joints and surface roughness in polysilicon films with equiaxial and dendritic structure have shown what follows. Both in equiaxial and dendritic (undoped and phosphorus-doped) polysilicon films, the triple grain boundaries joints of general type predominate. However, in undoped dendritic polysilicon films, in contrast to equiaxial and phosphorus doped dendritic ones, most of the joints are special ones, i.e. contain special grain boundaries. The number of multiple special joints is higher in undoped films. Splitting of grain boundaries under annealing is the most probable mechanism of special joint formation in polysilicon films of equiaxial structure. In dendritic films, special joints are a result of multiple twinning that occurs during the film formation. Moreover, a large number of twinning boundaries in these films results in a higher probability of their random meeting and special joint formation. The decreased number of special grain boundary joints in phosphorus doped dendritic polysilicon films as compared with undoped dendritic ones is caused by an effect of phosphorus doping. Phosphorus stimulates relaxation processes that are accompanied by a realignment of grain boundaries. Surface roughness in equiaxed films is nearly three times more than in dendritic films.

This research was supported in part by Ukrainian Ministry for Education and Science, grant M/240-2004, and by T.Shevchenko Kyiv National University (№ 01BF052-02).

References

1. Ch.V.Kopetsky, A.N.Orlov, L.K.Fionova, Grain Boundaries in Pure Materials, Nauka, Moskow (1987) [in Russian].
2. S.G.Protasova, V.G.Sursaeva, L.S.Shvindlerman, *Fiz. Tverd.Tela*, **45**, 1402 (2003).
3. A.L.Kolesnikova, I.A.Ovid'ko, A.A.Fedorov, *Pisma v Zh. Tekhn. Fiz.*, **29**, 7 (2003).
4. G.D.Suhomlin, Ch.V.Kopetsky, A.V.Andreeva, *Fiz. Metal. Metalloved.*, **62**, 349 (1986).
5. G.D.Suhomlin, A.V.Andreeva, *Fiz. Metal. Metalloved.*, **66**, 509 (1988).
6. N.G.Nakhodkin, N.P.Kulish, P.M.Lytvyn, T.V.Rodionova, in: Coll. of Sci. Works on Nanosystems, Nanomaterials, Nanotechnologies, Kiev, **2**, (2004), p.793 [in Russian].
7. M.G.Nakhodkin, M.P.Kulish, P.M.Lytvyn et al., *Visnyk Kyiv Nat. Univ. im.T.Shevchenka, Ser. Fiz.-matemat. nauky*, **2**, 359 (2005).
8. N.G.Nakhodkin, T.V.Rodionova, *Phys. Stat. Sol. (a).*, **123**, 431 (1991).
9. N.G.Nakhodkin, N.P.Kulish T.V.Rodionova, *Poverkhnost': Rentgen., Sinkhrotron. i Neitron. Issled.*, No.6, 19 (2003).
10. D.Waechter, N.G.Tarr, *Can. J. Phys.*, **65**, 1030 (1987).
11. V.M.Kosevich, V.M.Ievlev, L.S.Palatnik, A.I.Fedorenko, Structure of Intercrystallite and Interphase Boundaries, Metallurgia, Moskow (1980) [in Russian].

Особливості спеціальних стиків границь зерен в полікремнієвих плівках з рівноосьовою та дендритною структурою

М.Г.Находкін, М.П.Куліш, П.М.Литвин, Т.В.Родіонова

Методами атомної силової мікроскопії та просвічуючої електронної мікроскопії проведено порівняльний аналіз спеціальних стиків границь зерен та поверхневих неоднорідностей у полікристалічних плівках кремнію з рівноосьовою та дендритною структурою (нелегованих і легованих фосфором), отриманих методом хімічного осадження з газової фази в реакторі зниженого тиску. Розглянуто механізми формування спеціальних стиків у плівках різних структурних модифікацій. Проаналізовано роль фосфору у формуванні стиків границь зерен.