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MODELING OF THE MORPHOLOGY OF ALUMINUM FILMS BEFORE AND AFTER DIFFERENT KINDS OF THERMAL TREATMENT

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This paper considers the process of change in the structure of aluminum films on silicon in metallizing integrated microcircuits under the conditions of both prolonged and fast thermal treatment. We propose an analytical model of the formation and recrystallization of condensed aluminum films. It is based on the application of the Monte Carlo method and describes the change in the size and angles at triple points depending on the temperature and time of thermal treatment.

At the present time, in developing very large-scale integrated (VLSI) circuits with submicron sizes, much consideration is given to the formation of a metallized wiring made with films from aluminum alloys, which, however, have a number of important disadvantages connected with the recrystallization processes proceeding in them when they are heated. Among them are the formation on the film surface of abnormally high bumps and the silicon segregation at the grain boundaries of aluminum and at the aluminum–silicon interface. Their presence leads to a short circuit between different levels of metallization and an increase in contact resistances [1]. To reduce such disadvantages, methods lowering the intensity of the recrystallization processes proceeding in the films are needed. Modeling of this process makes it possible to establish the dependence of grain sizes and angles at triple points on the temperature and time of thermal treatment of aluminum films and, consequently, to control the recrystallization processes.

To model the process of nucleation and formation of a continuous film on the substrate surface and that of the change in its morphology under various kinds of heat treatment, let us consider briefly the main phenomena accompanying this process [2]. As the film being deposited on the substrate from a source with evaporation condenses, its growth includes the following stages: nucleation of grains (there appear small grains of ~0.5 nm statistically distributed on the surface); their growth (grains grow on large spatially separated islets); merging of islets (interconnections of islets form a net containing empty channels); filling of channels.

The mechanism of grain formation and growth is as follows. Incident particles with kinetic energies corresponding to temperatures higher than the substrate temperature are usually captured by the substrate surface and move on it as a 2D gas until they lose their excess energy and condense into a solid substance. The latter takes place only in the case where there exists a grain where adsorbed particles can liberate the excess energy. Grains are mainly formed on dislocations, portions with impurities, and other irregularities of the substrate's crystal lattice. In the course of evaporation, the number of islets (more than 0.5 nm in size) increases with a random distribution of their positions until saturation of the islet density is reached in the range from 10^{10} to 10^{12} at/cm^2, i.e., at their size of 10–100 nm. With further evaporation the islets grow and their density decreases due to their mutual coalescence. When the substrate is filled with grains, spatial islets begin to grow on them. Coalescence of grains can occur at their mutual contact. Then new grains are formed with an orientation corresponding to the orientation of the larger of the two grains.

The recrystallization processes at thermal treatment of polycrystalline films will proceed exactly in the same manner as does the interaction between grains, causing, on the whole, an increase in the grain sizes.

Taking into account that the processes of formation and recrystallization of condensed films have a probabilistic character, to model them, we make use of the Monte Carlo method, namely, the single-particle method of

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modeling the transition of particles from one state to another. The point of such an approach is the consideration of the random travel of one particle followed by the ensemble and time averaging of the particle travels.

In using the single-particle method to model diffusion processes, the calculation algorithm consists in that for a particle in a grain or at its boundary the transitions to adjacent states are calculated. On the plane, these can be transitions up, down, to the right, to the left or the particle position remains unchanged. The probability of these events is equal to

$$P_1 = \frac{1}{t_1 P_5}, \tag{1}$$

$$P_2 = \frac{1}{t_1 P_5} + \frac{1}{t_2 P_5}, \tag{2}$$

$$P_3 = \frac{1}{t_1 P_5} + \frac{1}{t_2 P_5} + \frac{1}{t_3 P_5}, \tag{3}$$

$$P_4 = \frac{1}{t_1 P_5} + \frac{1}{t_2 P_5} + \frac{1}{t_3 P_5} + \frac{1}{t_2 P_5}, \tag{4}$$

$$P_5 = \frac{1}{t_1} + \frac{1}{t_2} + \frac{1}{t_3} + \frac{1}{t_4} + \frac{1}{t_5}. \tag{5}$$

In each direction, depending on the diffusion coefficient D, the transition times of the particles t_1, t_2, t_3, and t_4 are calculated from the expression

$$t = \frac{a^2}{4D}. \tag{6}$$

The direction of particle motion is determined by comparing the randomly generated number q uniformly distributed on the interval [0, 1] with the calculated probabilities P_1, P_2, P_3, P_4, and P_5.

The chosen approach imposes restrictions on the dimensions of the object being modeled. In using the Pentium III-based computing technique, modeling of a thin-film metallization fragment representing a layer of grains of thickness up to 1 μm and area up to 100 μm^2 on the assumption that the grain sizes obey the logarithmically normal distribution law seems to be real. In particular, for a median grain size 0.65 μm their number may be as large as 200. For such a fragment, the number of potential participants may amount to $6.026 \cdot 10^{11}$ atoms. This means that, taking into account the possibilities of modern techniques, it is impossible to model the behavior of each atom. Therefore, the model has a number of assumptions: as a particle, it is suggested to use a cluster of atoms, which simultaneously participates in the interaction and goes from one state to another; the above process proceeds throughout the whole thickness of the film and, in modeling, it is considered that it proceeds between the particles in the plane; it is assumed that if the interaction goes along the grain boundary throughout the thickness, then the cluster represents a parallelepiped having a height equal to the thin-film element thickness and a square at its base.

Considering that the interaction of particles proceeds throughout the whole thickness of the film, the modeling can be reduced to the consideration of the recrystallization in the plane. In this case, the cluster projection on the plane will represent a square in which clusters of atoms having an analogous configuration are situated, and each cluster has a clearly defined center with respect to which the further calculation is carried out. The actual size of the cluster for which modeling was carried out was 0.02×0.02 μm.

The modeling process can be broken up into three main stages: formation of the stochastic structure of the film, modeling of the process of grain growth, and statistical processing of results.

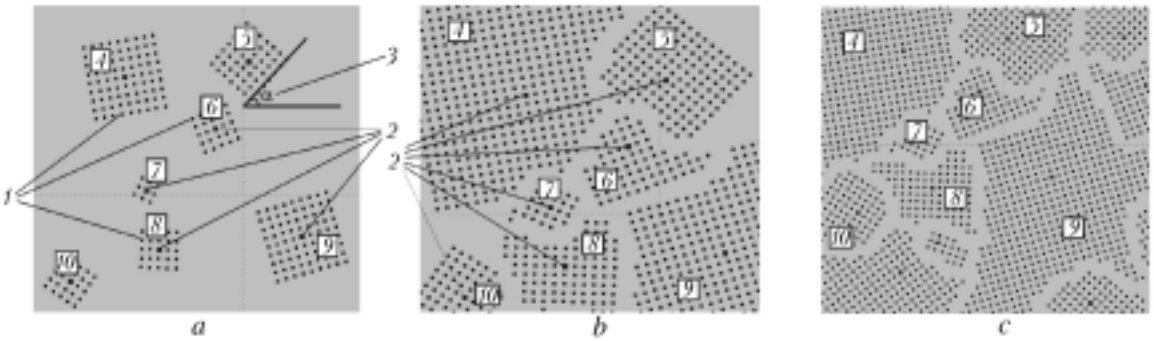

Fig. 1. Scheme of formation of the stochastic structure of aluminum films: a) in the initial stage of deposition; b) on completion of the grain growth 7; c) on completion of the process of structure formation in general; 1) clusters; 2) grain-formation centers; 3) angle α of grain orientations 5; 4–10) individual grains.

The algorithm of stochastic structure formation relies on the following principles: the grain sizes and nucleation centers around which grains begin to grow are arranged randomly and distributed according to the logarithmically normal law; each grain has an angle of orientation of the uniformly distributed crystal lattice; coalescence of grains upon their formation is impossible.

For each grain, the position of its center and the area in which the probability of the center growth is determined are drawn. The grain grows until it collides with a neighboring grain. Thus, a stochastic structure similar to the nonequilibrium structure obtained in depositing a film is formed.

In the initial stage of formation of the stochastic structure of the film, grains grow uniformly around all centers. As their size increases, the grains begin to contact one another and grow only in the direction where there is a vacancy (Fig. 1b). On completion of the growth of grains their boundaries experience bends characteristic of the nonequilibrium structure (Fig. 1c). In so doing, each grain has its own angle of orientation of the crystal lattice — between the *x*-axis and the crystal lattice cell (a cluster having a square form) (Fig. 1a).

In constructing a model of the recrystallization process, it is assumed that the dominant mechanism is the diffusion process, as a result of which the near-boundary atoms move from one grain to another. In the nonequilibrium film, this process has a mass character and involves a large number of atoms, which permits, in modeling by the single-particle method, considering a particle as a cluster.

We believe that the recrystallization process is realized in the near-boundary region whose size measures to be from two to three intercluster distances. Atoms in this region have a higher probability of interaction. In the model, such a process is determined by the energy state of atoms and depends on the bulk and surface energies of the grains. The probability of grain growth in terms of the bulk energy P_6 is determined from the expression

$$P_6 = \frac{G_1}{G_1 + G_2}. \tag{7}$$

The surface energy is taken into account in terms of the grain shape in the region of its growth. The probability of grain growth with regard for this energy is calculated in terms of the relation between the projections of the mass centers of the clusters lying in the growth region and of the attached clusters.

For all clusters lying in the near-boundary region, their lifetimes τ are calculated. Proceeding from the assumption of the diffusion nature of the recrystallization process, the cluster lifetime τ is determined from expression (6), taking into account that in this case α corresponds to the intercluster distance.

The cluster with the minimum lifetime defines the region with the most probable interaction. For all clusters, at their boundaries the lifetimes are drawn:

$$\Delta t = -\tau \ln q. \tag{8}$$

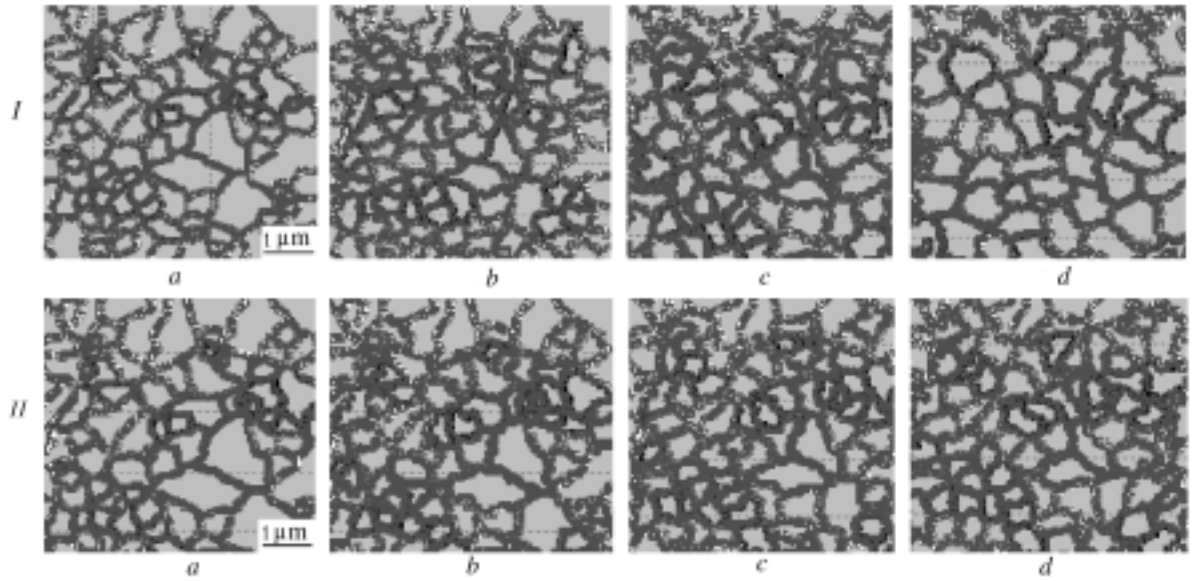

Fig. 2. Results of modeling of the surface morphology of the initial aluminum film (*I*) and silicon-doped aluminum film (*II*) of size 5 × 5 μm (a) and its view upon FTT for 50 msec at $T = 510^{o}$C (b) and for 1.7 sec (c), and after prolonged thermal treatment for 10 min at $T = 510^{o}$C (d).

Then the cluster having the shortest lifetime is selected. For this cluster, the following possible events are calculated: an increase in the grain size (attachment at this site of the neighboring grain clusters or clusters moving along the boundary) and the absence of change in its size.

If, as a result of this sampling, the grain grows in size, then in this region the grain extends around the cluster with the minimum lifetime by one cluster with regard for the crystal lattice orientation. For new boundary grain clusters, their lifetimes are drawn with regard for the grain growth time.

Then the process continues by analogy: a cluster with the minimum lifetime is found, the probabilities of growth are calculated, the event of grain growth is drawn, etc. In principle, the whole process of modeling proceeds in steps equal to the minimum lifetime. Summing these times, we obtain the current time.

The above algorithm permits modeling pure aluminum films. But the aluminum films used in microelectronics contain impurities of up to 1–1.5% of silicon. The silicon thereby is uniformly distributed throughout the crystal lattice. If the silicon is at the boundary, between grains, it prevents their growth. This factor should be taken into account in modeling the grain-growth process. To this end, we determined the probability that at a given instant of time at the grain boundary a silicon cluster appears. The possibility of such an event was calculated under the condition that at the initial instant of time the silicon was uniformly distributed over the film area. In this case, the least probability that the silicon will go to the boundary takes place when the silicon is in the grain center, and the highest probability takes place when it is near the boundary. On the average, it turns out that for the silicon to go diffusely to the boundary, it should cover a distance equal to a quarter of the median grain size. As soon as the silicon cluster finds itself at the grain boundary, its growth moderates.

To model the recrystallization process at a different character of the temperature action, we took into account the temperature change with time with a corresponding correction of the lifetime and diffusivity. Moreover, since in the case of fast thermal treatment (FTT) the dominant process is the grain-boundary diffusion and in the case of prolonged treatment — the bulk diffusion, the lifetime of a cluster is calculated depending on the duration of thermal treatment by either the boundary or bulk parameters of the diffusion process.

For modeling, the following parameters of the regime of treatment are given: modeling time; initial temperature of the sample; silicon concentration in aluminum; temperature to which heating is carried out; time of temperature stabilization (if any); and time of sample cooling down to the initial temperature. Depending on what diffusion mechanism in the recrystallization is dominant, the corresponding values of the diffusion-process parameters E_a and D_0 are given. It should be noted that the value of these parameters also depends on the film composition, i.e., in our case —

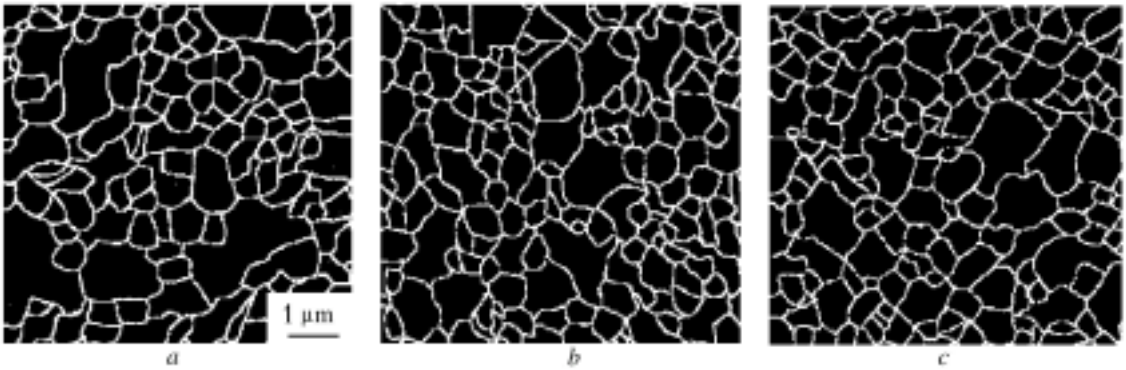

Fig. 3. Surface morphology of the silicon-doped aluminum film (digital method of image processing): a) upon deposition; b) upon deposition and prolonged thermal treatment for 10 min at $T = 510^{o}$C; c) upon deposition and FTT for 1.7 sec at $T = 510^{o}$C.

on the percentage of silicon in the aluminum film. The values of these parameters in the modeling were as follows [3]: for pure aluminum film in the case of grain-boundary diffusion $E_a = 0.8$ eV, $D_0 = 10^{-7}$ cm^2·sec^{-1}, and in the case of the bulk diffusion $E_a = 1.25$ eV, $D_0 = 10^{-7}$ cm^2·sec^{-1}; for aluminum films doped with silicon, in the case of the grain-boundary diffusion $E_a = 0.9$ eV, $D_0 = 2\cdot10^{-7}$ cm^2·sec^{-1}, and in the case of the bulk diffusion $E_a = 1.4$ eV, $D_0 = 2\cdot10^{-7}$ cm^2·sec^{-1}.

Modeling is carried out as long as the number of growing grains is greater than zero. Upon its completion, statistical processing of the results obtained is performed, namely, the grain sizes and angles at triple points (points at which contact of three grains takes place) are determined. The grain size is determined proceeding from the parameters of the rectangle in which it is inscribed. To estimate the value of angles at triple points, the grains forming these angles are selected. The obtained statistical data on the grain size and angles are output in the form of histograms, as well as in the form of tables containing the basic parameters of these data.

The modeling of the surface morphology for pure aluminum films (Fig. 2, *I*) has made it possible to establish the following regularities: the initial aluminum film (Fig. 2, Ia) has a mean grain size of 0.47 μm and a spread (0.1–0.4 μm) with its distribution maximum in the region of 0.43 μm; the distribution of angles at triple points has a wide maximum (99–136^{o}) and a spread (54–196^{o}), which points to a low ordering of the structure of the initial aluminum film.

Treatment of this film by the FTT method (Fig. 2, *I* b, c) leads to an ordering of the structure and an increase in the mean grain size. For instance, in treating with millisecond pulses (Fig. 2, *I* b) it increases to 0.58 μm with a distribution maximum in the region of 49 μm and a spread from 0.3 to 1.1 μm, which is much less than in the initial film. The distribution of angles at triple points has a maximum in the region of 120^{o} and has become less blurred (108–128^{o}). In the case of using second-length pulses (Fig. 2, *I* c), the increase in the mean grain size is somewhat greater than for millisecond pulses and is 0.63 μm with a maximum in the 0.64–0.77 μm range and a spread from 0.37 to 1.0 μm. In so doing, the distribution of angles at triple points has a sharp maximum in the region of 116^{o}. The given data for second pulses point to the fact that in this case the aluminum film structure ordering proceeds more intensively than for millisecond pulses. One feature of the grain growth in the case of FTT is noteworthy: this process proceeds not only during the heating but also during the cooling of the plate. For the millisecond pulses, the grain growth is observed mainly at the stage of film cooling-down, and for the pulses of second durations the heating–cooling cycles make equal contributions to the time interval of the grain growth.

In the case of prolonged heat treatment, the mean grain size is 0.82 μm with a distribution maximum in the region of 0.85 μm and a spread from 0.49 to 1.3 μm (Fig. 2, *I* d). In so doing, the value of the maximum in the distribution of angles at triple points in the region of 120^{o} has decreased from 40 to 33%, i.e., under a prolonged thermal treatment, besides the structure-ordering process, its disordering takes place when the structure contains both abnormally large and small grains.

Analysis of the results of the modeling for silicon-doped aluminum films has shown (Fig. 2, *II*) that the changes in the grain caused by different kinds of thermal treatment have the same character as for pure aluminum

films with the only difference being that the mean grain size in the film is smaller. For instance, at prolonged thermal treatment the mean grain size increased from 0.47 to 0.62 μm (Fig. 2, *II* d), in the case of using second pulses — from 0.47 to 0.57 μm (Fig. 2, *II* c), and for millisecond pulses — from 0.47 to 0.53 μm (Fig. 2, *II* b), i.e., in the last two cases the increase in the grain size is comparable to that for pure aluminum films. This is due to the fact that only at prolonged thermal treatment is there enough time for practically complete diffusion of silicon from the grain bulk towards the boundaries and blocking of the grain growth.

Comparison of these results of modeling with the results of experimental investigation of the recrystallization processes in aluminum films after different kinds of thermal treatment [1] (Fig. 3) reveals their complete identity, which verifies the reliability of the proposed model of aluminum film recrystallization.

NOTATION

t_1, t_2, t_3, and t_4, times of possible transitions up, down, to the right, and to the left, respectively, sec; t_5, time of rest, sec; P_1, P_2, P_3, P_4, and P_5, probabilities of transition up, down, to the right, to the left, and/or probability that the particle position remains unchanged, respectively; $D = D_0 \exp(E_2/kT)$, diffusivity, $cm^2 \cdot sec^{-1}$; α, distance between lattice nodes or intercluster distance, cm; D_0, diffusivity constant, $cm^2 \cdot sec^{-1}$; E_a, activation energy, eV; k, Boltzmann constant, eV/^{o}C; T, temperature, ^{o}C; P_6, probability of grain growth as to bulk energy; G_1 and G_2, bulk energies of interacting grains, eV; τ, cluster lifetime, sec; q, randomly generated number uniformly distributed over the interval [0, 1]. Subscripts: a, activation; 0, corresponds to the diffusivity at $T \to \infty$.

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