Monte Carlo Simulation of Silicon Nanocrystals Formation in Annealed Nitrogen Doped Silicon Thin Films

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***Abstract*—In this work, we deal to simulate the formation of silicon nanocrystals in annealed nitrogen doped silicon thin films by using Monte Carlo methods. To do this, we propose an impurities distribution pattern in the simulation matrix able to describe the influence of nitrogen presence in the amorphous silicon matrix, on the crystallization kinetic during the thermal annealing. Thus, the image of simulated crystallite structure, the numerical data reflecting crystallite size distribution, have been produced by the program according to the sites number reserved for the impurities. Results show the strong dependence of nanocrystalline size and density on the nitrogen amount.**

Keywords-Monte Carlo simulation; nanocrystals; nitrogen

I. *Introduction*

A great deal of interest has been devoted recently to silicon nanocrystals (Si-nc) from both the experimental and the theoretical point of view. These nanomaterials have a huge potential in technological applications in many fields because Si-nc is expected to exhibit a quantum size effect. Several methods tried to form silicon nanocrystals in silicon based materials among them the crystallization of amorphous nitrogen heavily doped silicon (SiN_x) thin films [1, 2]. Indeed, the wavelength of luminescence can be controlled by changing the size of silicon nanocrystals that could be controlled by variation of nitrogen amount in thin films [2]. Therefore, the size control of silicon nanocrystals is very important for novel display applications [2]. It was reported [3] that the presence of strong Si–N bonds of which the density increases with the amount of nitrogen prevented the coalescence between the smaller crystallites. To simulate this phenomenon by using the Monte Carlo (MC) method, we propose a pattern for the distribution of nitrogen impurities that can model the noncoalescence of the silicon crystallites.

II. *Simulation procedure*

For the simulation of the kinetic of crystallization and grain growth phenomenon, a basic MC algorithm based on Potts model can be found in detail in the literatures. First, the simulation is applied in a two-dimensional discrete lattice represented the microstructure. Each of the lattice sites is assigned a random orientations number between 1 and Q, where Q is the total number of grains orientations. A contiguous group of lattice sites that all have the same orientations number are part of the same grain. Grains boundary segment is defined to lie between two sites of unlike orientations number. The energy of a lattice site i is given by

$$E = -J \sum_{i=1}^{N} \sum_{j=1}^{Z} \left(\delta_{Q_i Q_j} - 1 \right) \cdot \qquad (1)$$

Where J is a positive constant which sets the scale of the grains boundary energy expressed in (eV/m), Q_i the orientations number at the randomly selected site i, and Q_j the orientations numbers of its nearest neighbors. Note that the number of nearest neighbors Z is eight for a square lattice and six for a triangular lattice. δ is the Kronecker delta function, which is 1 if $Q_i = Q_j$ and 0 otherwise.

To simulate the kinetic of crystallization, a lattice site is selected randomly from the simulation matrix. Note that each lattice site must be chosen just once in a MC Step (MCS) [8]. Its energy (E_1) is calculated according to (1). Then, a new random orientations number is assigned to this lattice site from its nearest neighbors and its energy (E_2) after reorientation is again calculated. The reorientation is accepted when ($E_2 < E_1$). Otherwise it is accepted with the Boltzman probability

$$p = \exp\left(-\Delta E / K_B T \right) \cdot \qquad (2)$$

Where

$$\Delta E = E_2 - E_1, \tag{3}$$

k_B is the Boltzman constant and T is the temperature. One MCS comprises N such reorientation attempts. Here, N refers to the total number of lattice sites in the system.

The technique of MC is used to simulate the phenomenon of the silicon nanocrystals formation on a network of (200x200) sites, and an orientation number Q of 64. To model the phenomenon of noncoalescence caused by the presence of strong Si-N bonds, the impurities distribution in the simulation matrix is described by using a new pattern assuming that the distribution of impurities has not a random aspect as in the literature [4, 5]. In our approach, the impurities occupy the nearest sites to the selected site (the nucleation site) and do not participate in the reorientation process as it is illustrated in Fig. 1(b). Thus, by increasing the number of nucleation sites surrounded by impurities (increase in the number of impurities introduced into the matrix); the nucleation sites become increasingly isolated from each other preventing the coalescence phenomenon. Consequently, the formation of crystallites with small sizes can be simulated using MC method.

III. Results and discussion

We apply our approach to simulate the formation of silicon nanocrystals in nitrogen doped silicon thin films according to the nitrogen tenor in films. In first time we study an experimental case [1] basing on the grains size distribution obtained via scanning electron microscopy (SEM) observation, of nitrogen doped silicon thin films containing 7% nitrogen content and annealed under a temperature of 850°C during 2 hours. A good statistical agreement between theoretical and experimental grains size distributions and the microstructures of film (Figs. 2 and 3 respectively) is obtained by using a MCS of 1000, and a number of sites occupied by impurities equal to 37131.

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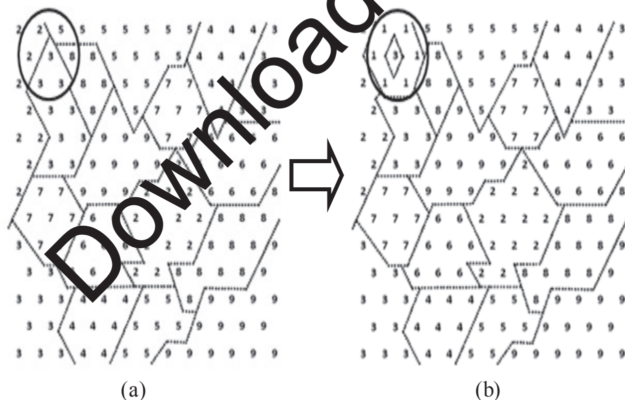

(a) (b)

Figure 1. Schematic design of simulation matrix (a) without impurities, (b) with impurities. The lines represent the grains boundaries when the integer numbers are the crystallographic orientations numbers. 3 in the circle (Fig. 1(b)) is the orientations number of a nucleation site selected primarily for the reorientation process. 1 is the orientations number of the impurities sites.

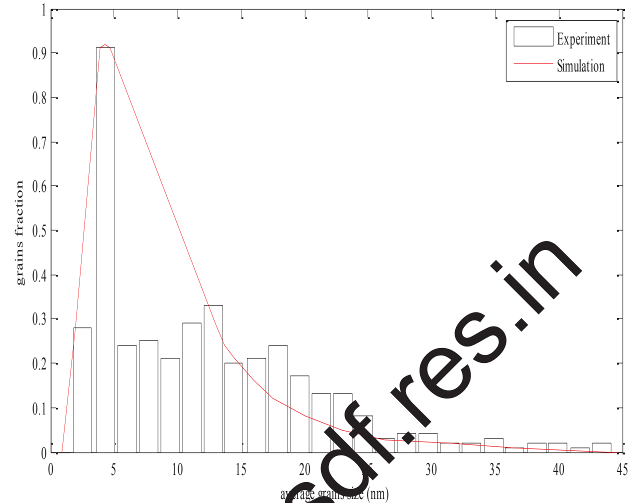

Figure 2. Superposition of experimental and theoretical grain size distributions.

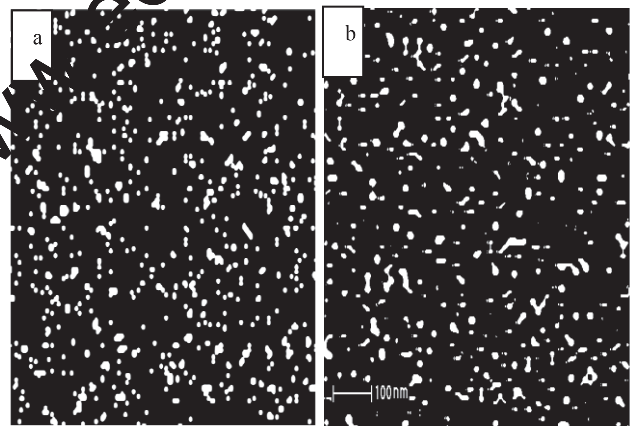

Figure 3. Microstructure images (a) MC simulation, (b) SEM observation.

To simulate the influence of nitrogen content on the film morphology in terms of nanocrystals size and density, we keep the same temperature (850°C), the same MCS (1000) and we proceed to a change in the number of sites reserved for impurities.

The MC simulation results are represented in Figs. 4, 5 and 6. We observe a shift toward the small sizes combined with a narrowing of GSDs when nitrogen content increases. These results show that the density of crystallites of small sizes increases with the increase of impurities, highlighting the role of nitrogen in the formation of silicon nanocrystals in good agreement with the experimental observations.

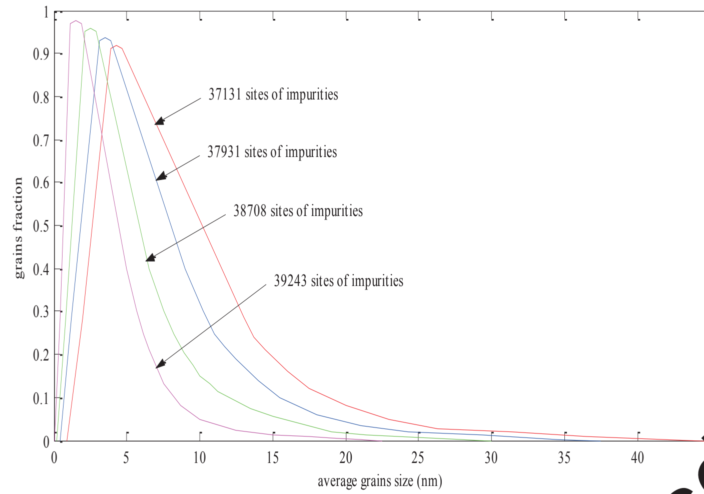

Figure 4. Grain size distributions obtained by MC simulation as a function of impurities sites number.

Figure 5. Microstructures obtained by MC simulation as a function of impurities sites number.

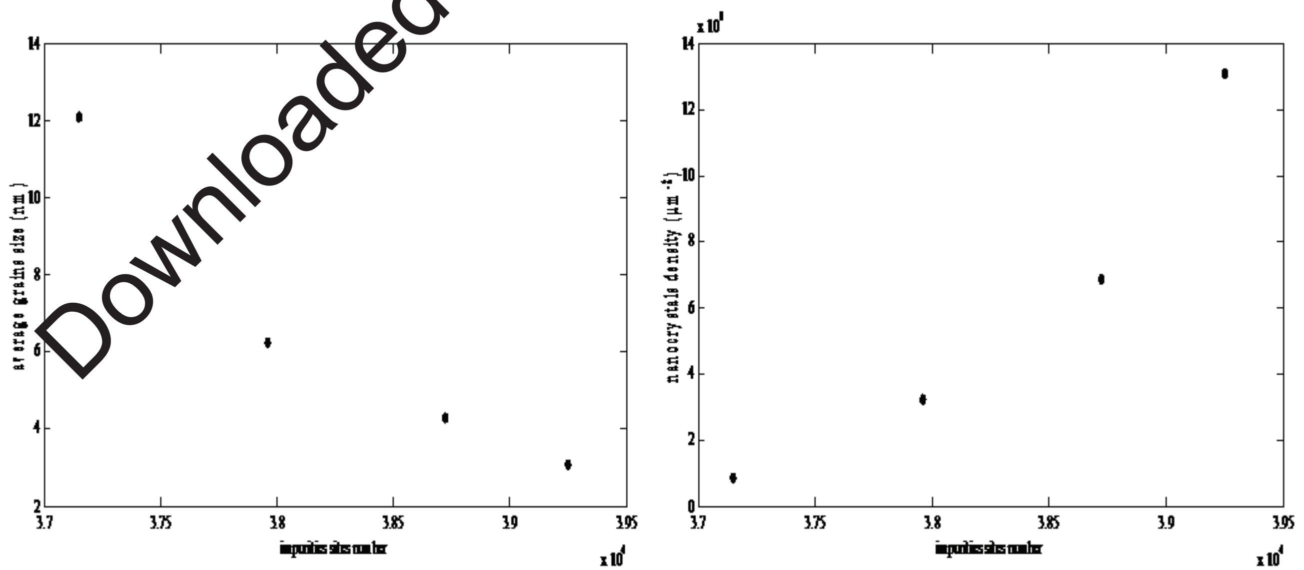

Figure 6. Variation of silicon nanocrystals size and density as a function of impurities sites number.

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IV. Conclusion

The silicon nanocrystals formation according to the nitrogen content present in silicon thin film has been studied in the present work, using Monte Carlo simulation based on Potts model. The noncoalescence between silicon crystallites caused by the presence of Si-N bonds has been modeled by using a particular pattern of the impurities distribution in the basic simulation matrix. Results show that an increase of impurities content leads to an increase of nanocrystals density combined with a decrease in their sizes, highlighting that the proposed impurities distribution aspect describes well the experimental observations.

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