



Full Length Article

Evolution of medium-range order and surface compositions by mechanism-driven model with realistic network

Yixiong Zhang^{a,b,**}, Hong Wang^a, Yanqing Gou^b, Shaoji Jiang^{a,*}^a School of Physics, State Key Laboratory of Optoelectronic Materials and Technologies, Sun Yat-sen University, Guangzhou 510275, People's Republic of China^b Department of Physics and Electrical Engineering, Hanshan Normal University, Chaozhou 521041, People's Republic of China

ARTICLE INFO

Keywords:

Fluctuation transmission electron microscopy
Structural topology
Atomic-mechanism driven
Disorder to order structure

ABSTRACT

To elucidate the evolution of structural topology with the medium-range order of growing silicon films fabricated by plasma enhanced chemical vapor deposition with highly diluted hydrogen, we established a the model for a realistic network of growth films based on continuous random networks. We combined the kinetic Monte Carlo method with the interactions of various atomic-scale mechanisms from first-principles density-functional-theory computations and molecular-dynamics computations. To quantitatively characterize the short- and medium-range order, in addition to the higher order of the structural network, we applied a fluctuation transmission electron microscopy simulation and the pair correlation function to measure information about local order regions in film network. Interestingly, we found that the inflexion temperature of surface SiH₃ coverage directly affected the silicon hydride crystallization process by forming much chemisorption from physisorption of surface hydride species. More interestingly, based on the atomic-scale chemical mechanism of the growth process of film, our results first realistically rendered the continuous disorder-order phase transition from structural topology, which is differed from previous knowledge that the amorphous to polycrystalline transition is a discontinuous short-range-order to long-range-order phase transition. Especially, we predicted the temperature dependence of evolution of structural network of the film and elucidated that the growing silicon film formed a nanocrystalline structural network at very low temperatures, relative to those of thermal annealing. This result was ascribed to interactions and competitions between various mechanisms and critically due to the dissociation mechanism and H-induced crystalline mechanism of silicon hydride, which isn't depend on only one of the model mechanisms of surface diffusion model, etching model and annealing model. These results provide significant new physical insight into an experimentally relevant process for optimizing a strategy of deposition condition.

1. Introduction

A fundamental understanding of the chemical and physical behaviors of the evolution of elaborate structural networks based on atomic-scale mechanisms, is needed to fabricate technologically important novel materials [1–3]. Previously, much work has focused on the experimental and theoretical studies of the chemical mechanism about the transition of the disorder-to-order structure network of growth silicon film [4–7]. At low temperatures, the atomic-scale mechanisms that form the local order regions in the silicon film have attracted great interest in the field of semiconductor nanometer materials, as this understanding would enable new technologies, including inexpensive flexible electronics and solar cells [6,7]. Furthermore, three

predominant models describe the transition mechanisms from the amorphous-to-crystalline in silicon, including the surface diffusion model, the selective-etching model and the chemical annealing model. These have been proposed to explain the evolution of a-Si:H films containing nanocrystals or the growth of nc-Si:H, providing indirect interpretations of the crystallization mechanism. These models are still controversial, moreover, some key processes are still questionable [6,8–10].

To identify an accurate atomic-scale topological structural model and develop a necessary understanding of growth of silicon films, some models for a-Si have been proposed. The ideal continuous random network (CRN) for a-Si structural topology well reproduces the experimental pair correlation function obtained by diffraction. However,

* Corresponding author.

** Corresponding author at: School of Physics, State Key Laboratory of Optoelectronic Materials and Technologies, Sun Yat-sen University, Guangzhou 510275, People's Republic of China.

E-mail addresses: zhyxiong@mail2.sysu.edu.cn (Y. Zhang), stsjsj@mail.sysu.edu.cn (S. Jiang).<https://doi.org/10.1016/j.apsusc.2018.09.090>

Received 28 July 2018; Received in revised form 7 September 2018; Accepted 10 September 2018

Available online 11 September 2018

0169-4332/ © 2018 Elsevier B.V. All rights reserved.

the model is inconsistent with variance data about the medium-range order (MRO) from fluctuation transmission electron microscopy [4,11,12]. The revert Monte Carlo (RMC) scheme was developed for computer generation of amorphous structures [13,14]. But, the well-known RMC includes experimental data constraints in the revert model, which cannot describe the deposition growth mechanism of the films. More recently, Treacy et al. proposed that paracrystallites of local ordered regions are embedded within the a-Si matrix, describing the structure as containing the medium-range structural (5–30 Å) materials [4]. Law et al. also reported that paracrystallites are localized regions of higher ordered Si material, which act as nucleation sites in a-Si:H during solid phase crystallization [4,11,15,16]. Very recently, to revealed important information to the structural properties or electronic characteristics of a-Si:H films, V. L. Deringer et al. reported an accurate structural models of a-Si can be obtained using a machine-learning-based interatomic potential. In addition, D. Igram et al. fabricated several models of various sizes to analyze the structural, electronic and vibrational characteristics [17,18]. These studies provided important information about the crystallization mechanism and the evolution of atomic-scale topological structures in the growth nanocrystalline silicon films.

2. Methods

To characterize the structural network of the growing film at the critical stages of originating incubation, forming nucleation, producing nanocrystalline, we probed structural topology with measurements that were sensitive to MRO. Although the pair correlation function was an important probe of short-range-order (SRO), that is, within a distance of the two nearest neighbor bonds, it was relatively insensitive to the presence of MRO [4,19,20]. Moreover, it suggests probabilities that different atomic networks have the same radial distribution function, resulting in a lack of unique atomic structural model solutions for the only pair correlation function, $g(r)$, constrained. Therefore, it was necessary to add higher order correlation functions to the pair correlation function for the characterization of model in joint matching and correlate with experimental results. Fluctuation transmission electron microscopy (FEM) has been repeatedly verified as a powerful “fingerprint” to evaluate the information of high-order correlation functions about medium-range order in structures [11], and they calculated statistical fluctuation variance about the diffracted intensity from the probed material. FEM probed both the statistical mean diffraction data and the statistical variance of diffraction data, which provides explicit information about four-body correlation functions, beyond the radial distribution-function of the pair-body correlation function. The FEM simulation variance was calculated from a set of microdiffraction pattern dark field image intensities, $I(p, k, R)$, via

$$V(k, R) = \frac{\langle I^2(p, k, R) \rangle_p}{\langle I(p, k, R) \rangle_p^2}, \quad (1)$$

where p denotes the location of the probe, and R is the probe width associated with spatial resolution, the angular brackets represented averaging over all probe locations, and k is the scattering vector [4,21].

To establish a description of realistic structural evolution of a growing film, we proposed a local definite CRN structure combination with the kinetic Monte Carlo (KMC) method, based on an atomic-scale mechanism-driven film growth model [22–27]. This enabled us elucidate the evolution of structure of films, which was attributed to the atomic interactive behaviors of film growth, such as diffusion, H-transfer-mediated dissociation, H-mediated etching, and H-induced crystallization of chemical annealing processes in plasma enhanced chemical vapor deposition (PECVD) when growing a-Si:H thin film with silane by diluted strong H_2 . Based on the continuous network model, the bond length, bond angle and dihedral angle of deposited species on film were determined, by reacting various atomic-scales mechanisms

and drives the deposited sites of these species toward local non-crystalline or crystalline positions [4,5].

To analyze the essential features of the crystallization mechanism and the topological structure of silicon films, the predominant silyl radical precursor and H elementary reaction procedures were the primary focus in the simulation. The elementary chemical reactions and transport processes [11,22,28–30] emphasized during the simulations of H and silyl radical impingement on a-Si:H films, included (1) H-induced crystallization behavior, (2) H-transfer-mediated Si incorporation procedure, (3) H atom etching adsorbed SiH_3 radical process, (4) silyl radical diffusion, and (5) Hydrogen atom diffusion on the H-passivated a-Si:H surface. Hydrogen was preferentially incorporated in the dangling bonds of a-Si:H film. Our model represented the activation energy barriers of H-insertion strained Si–Si bonds behavior, H-transfer-mediated Si incorporation in surface valleys, H-transfer-mediated Si incorporation on surface terraces, H atom etching silyl hydride, SiH_3 radical diffusion, and H atom diffusion by E_{hi} , E_{vi} , E_{hb} , E_{et} , E_{sd} , and E_{hd} , respectively.

The corresponding process rate of thermally activated kinetic processes or reaction events could be estimated by the standard Arrhenius rate equation $\nu_{i \rightarrow j} = \nu_0 \exp[-(E_{i \rightarrow j} + E_s + nE_b)/k_B T]$, where $E_{i \rightarrow j}$ represented the activation energy barrier for the above various growth process of the reaction, considering the minimum-energy pathway from state i to state j . E_s is the additional Ehrlich-Schwoebel barrier energy for silyl radical motion for downward at the island edge, k_B is the Boltzmann constant, T is the substrate temperature, n and E_b are the number of nearest neighbor radicals and the nearest neighbor radicals binding energy, respectively [22,31]. The attempt frequency prefactor ν_0 was described as $\nu_0 = 2T/h$, and h is Planck's constant.

With the aperiodic boundary conditions, the KMC simulation based on CRN was performed on an ideally amorphous flat glass substrate with a surface area $60 \times 60 \text{ \AA}^2$. Moreover, SiH_3 radicals and H atoms from plasma with the hydrogen dilution ratio R_H (denoted as $R_H = H_2/[SiH_4 + H_2]$) were deposited onto the surface at a given silicon deposition rate D_{si} . The activation energy barrier E_{hi} for the H insertion strained Si–Si bonds and relaxed Si–Si bond after insertion were characterized by approximately 0.60 eV, which was based on first-principles density-functional-theory (DFT) computations and MD simulations [5,32]. We assumed the activation energy barrier for Si incorporation in surface valleys and on surface terraces with $E_{vi} = 0.35 \text{ eV}$ and $E_{hi} = 0.90 \text{ eV}$, respectively [28,31]. The range of activation energy barriers E_{sd} for SiH_3 radical diffusion on the a-Si:H surfaces were taken as 0.15–0.30 eV [33,34]. H atom etching of the silicon hydride weakly adsorbed with surface back to the gas phase process activation barriers E_{et} is 0.13–0.39 eV and H atom diffusion activation barriers $E_{hd} = 0.3 \text{ eV}$ [28,34,35]. We assumed the bond energy between two nearest-neighbor radicals was $E_b = 0.1 \text{ eV}$ reflecting the interaction of atoms in the vicinity. Furthermore, we took the additional Ehrlich-Schwoebel barrier energy E_s for the additional activation barrier of silyl radical motion downward at the island edge $E_s = 0.11 \text{ eV}$ [31,34]. In our simulations, under an optimization of strategy [32], the silicon radical deposition rate, D_{si} , was fixed to a low deposition rate of 2ML/s and the hydrogen dilution ratio was set as $R_H = 95\%$, denoting the heavily diluted of SiH_4 by H_2 to provide sufficient H atoms for the H-induced crystallization event.

3. Results and discussion

In Fig. 1, the results of mechanism-driven CRN-KMC simulations demonstrate the temperature dependence of the evolution of the network and surface chemical compositions of film fabricated with PECVD. At the relatively low temperature of 273 K, which is shown in Fig. 1(a), the structural network of the film clearly lacks crystallized atoms in the film matrix, which manifestly characterizes the structure of growing film with approximately perfect amorphous phase structure. The temperature dependence of the surface coverage ($C_{(i)}$), T -C, of various

Download English Version:

<https://daneshyari.com/en/article/9569598>

Download Persian Version:

<https://daneshyari.com/article/9569598>

[Daneshyari.com](https://daneshyari.com)