

Effect of MeV electron irradiation on Si-SiO₂ structures

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Abstract: The effect of 20-MeV electron-beam irradiation on Si-SiO₂ structures was studied. The Si-SiO₂ samples were fabricated on n-type <100>-oriented Si wafers with resistivity of 4.7 Ω cm. Following a standard cleaning procedures, thermal oxidation in dry O₂ + 8 % HCl ambient was performed at 900 °C to produce oxide layer with a thickness of 20-nm. After oxidation, the samples were cooled at a rate of 1° C s⁻¹ in the ambient where the oxidation was carried out. The samples were exposed to a beam of 20-MeV electrons with a flux of about 1.2×10¹⁵ cm⁻². The oxide thickness was measured by ellipsometry before and after electron irradiation, which showed that the SiO₂ thickness in the irradiated Si-SiO₂ structures increases. This result can be connected with the increase of the oxygen content at the Si-SiO₂ interface. If one takes into account the defect generation in the whole Si-SiO₂ structure, it is reasonable to expect that the oxygen motion through the SiO₂ oxide defects will be stimulated during MeV electron irradiation. The changes on the SiO₂ surface roughness induced by the high-energy electron irradiation of the Si-SiO₂ structures were observed by atomic force microscopy (AFM). It was seen that these changes consisted in the formation of nanocrystals at the SiO₂ surface. We assume that the MeV electron irradiation breaks the Si-O bonds, the free oxygen moves through the radiation defects and creates conditions whereby Si nanostructures are generated in the oxide SiO₂ and at the SiO₂ surface of the structures.

1. Introduction

The dissociation of SiO₂ layer in Si heterostructures using high-energy electron irradiation has been discussed by many authors [1-7]. Xi-wen Du et al. observed transformation of amorphous SiO₂ into crystalline Si (c-Si) by keV electron irradiation at ambient temperature [3]. The authors assumed that this transformation takes place in two steps: the first one involves the transformation of amorphous SiO₂ to amorphous silicon (α-Si), while the second one is the crystallization of amorphous silicon. They reported that α-Si nanodots are produced under keV electron irradiation, but c-Si nanodots are formed by heating the electron-irradiated Si heterostructure to 850 K [3]. But, the understanding of the dissociation of SiO₂ by high-energy electron irradiation is very limited and it is not clear whether c-Si nanodots can be obtained by electron irradiation only.

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We present a study of the changes in amorphous SiO₂ layers during 20-MeV electron irradiation at room temperature. Using atomic force microscopy, we observed dramatic macroscopic changes on the SiO₂ after irradiation. At the same time, the oxide thickness of the samples increased (as evidenced by ellipsometry before and after MeV electron irradiation). These results correlate with our previous studies of SiO₂ layers of Si-SiO₂ structures during MeV electron irradiation [4, 5, 7]. We presumed that transformation of amorphous SiO₂ to crystalline silicon can be achieved also by MeV electron irradiation only. In this work, we investigate silicon nanostructures generation in the Si-SiO₂ structures by MeV electron irradiation. To study the effect of MeV electrons on formation of nanostructures at the SiO₂ surface, Si-SiO₂ structures were analyzed by AFM before and after electron irradiation with a flux of about $1.2 \times 10^{15} \text{ cm}^{-2}$.

2. Experimental details

The studies were performed with n-type <100> oriented silicon wafers with resistivity of 4.7 Ω cm. The wafers were cleaned and then oxidized in dry O₂ + 8 % HCl at 900° C to produce an oxide layer of a thickness of 20 nm. The samples were cooled in the same oxygen ambient (in which oxidation was carried out) at a rate of 1° C s⁻¹. Then the samples were exposed to 20-MeV electron-beam irradiation perpendicular to the SiO₂ surface. This energy is sufficiently high for the electrons to penetrate through the entire SiO₂ and Si wafer and create radiation defects in the whole structure. The electron irradiation with a flux of $1.2 \times 10^{15} \text{ cm}^{-2}$ was carried out on the Microtron MT-25 (equipment in the Flerov Laboratory of Nuclear Reactions of the Joint Institute of Nuclear Research - FLNR, JINR, Dubna, Russia), in a vacuum chamber under pressure of about 1×10^{-2} Pa. The beam current was about $I_e = 9 \text{ μA}$. The sample temperature was controlled during the entire process and kept close to room temperature. The distance between the Microtron window and the samples was 150 mm.

The oxide thickness of the samples was determined by ellipsometry before and after each electron irradiation. The morphology of the SiO₂ surfaces of the Si-SiO₂ structures before and after irradiation was observed by taking AFM images over areas of 1 μm² with scanning probe microscope CP-II in air at room temperature.

3. Results and discussion

The ellipsometric measurements showed that the oxide thickness of all samples increased after 20-MeV electron irradiation – from 20 nm (before) up to 21,7 nm after irradiation with a flux of $1.2 \times 10^{15} \text{ cm}^{-2}$. This is in agreement with our earlier studies, when we found that this increase depends on the radiation dose and the ambient in which the irradiation is carried out [5]. A possible explanation of this result is the increase of the oxygen content at the Si-SiO₂ interface. In [6] we reported that, as a result of high-energy electron irradiation, the oxygen concentration at the Si-SiO₂ interface increases. Further, nuclear reaction analysis and X-ray emission spectroscopy demonstrated that MeV electron irradiation of Si-SiO₂ samples leads to an increase in the oxygen surface density. A possible mechanism of the radiation-stimulated oxidation may be oxygen motion through the radiation defects in the oxide layer in order to reach the silicon surface. If we take into account the defect generation in a result of MeV electron irradiation, it is reasonable to expect that the oxygen motion will be stimulated.

The microscopic morphologies of the SiO₂ surfaces of the Si-SiO₂ structures were observed by an atomic force microscope. Figure 1 is a typical AFM image of a SiO₂ film observed before irradiation. As can be seen, the sample did not exhibit any ordered surface structures.

After 20-MeV electron irradiation with the dose quoted, the AFM images revealed a roughing of the surface (shown in figure 2), with a formation of clusters, an increase in the crack density, crack healing, and, finally, a substantial modification, namely, the irradiation resulted in accumulation of nanostructures at the SiO₂ surfaces.

We suppose that the MeV electron irradiation breaks the Si-O bonds and the free oxygen moves through the radiation defects in the SiO₂ (as generated by the high-energy electrons), i.e., MeV electron irradiation creates conditions whereby Si nanostructures are generated in the oxide. This is in

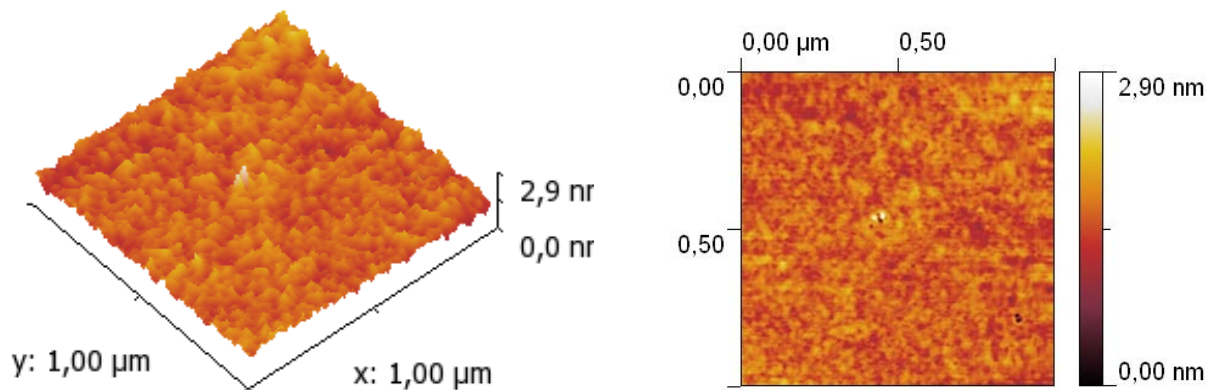


Figure 1. Typical AFM images of the Si-SiO₂ samples before electron irradiation.

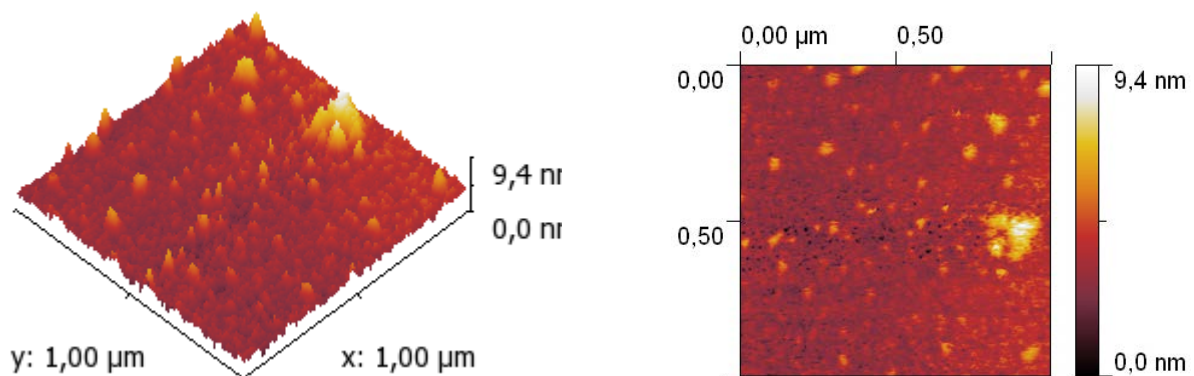


Figure 2. AFM results after 20 MeV electron irradiation with a flux of $1.2 \times 10^{15} \text{ cm}^{-2}$.

good agreement with earlier results [4, 6]. We assume that the radiation defects (created by MeV electron irradiation) play also the role of sources of Si nanostructures generation in the SiO₂ [7]. Xi-wen Duet al. proposed that SiO₂ is first transformed into α -Si and then α -Si is transformed into c-Si during electron irradiation [1]. They showed the change in number and type of chemical bond during irradiation. The authors assumed that most of the α -Si islands are transformed in c-Si islands and suggested that the content of the Si-Si bond gradually increases from 0% in SiO₂ to 100% in pure Si as the time of irradiation is raised. We assume that our results (shown at figure 2) able to confirm their suggestion.

4. Conclusions

Using ellipsometry method and atomic force microscopy (AFM), it has been shown that the morphology of SiO₂ is changed as a result of the interaction between MeV electrons and the Si-SiO₂ structure. A possible reason for the observed oxide thickness increase of the irradiated samples is the increase of the oxygen concentration at the Si-SiO₂ interface. Our results demonstrate that the radiation defects (created by MeV electron irradiation) stimulate the oxygen motion through the oxide layer to reach the silicon surface, so that the oxide thickness increases and radiation-stimulated oxidation in a result of high-energy electron irradiation takes place. We assume that the radiation defects in the SiO₂ oxide act also as sources of Si nanostructures generation. MeV electron irradiation breaks the Si-O bonds, the free oxygen moves through radiation defects and creates conditions under which Si nanostructures are generated in the oxide SiO₂. Finally, we proved that

nanocrystals can be created in the SiO₂ layer and at the SiO₂ surface of the Si- SiO₂ structures by MeV electron irradiation only.

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