Si nanoclusters generated in Si-SiO₂ structures implanted with different doses of Si ions

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Abstract. Topography changes of a SiO₂ surface and formation of Si nanoclusters, induced by high-energy electron irradiation of ion implanted Si-SiO₂ structures, are studied by atomic force microscopy (AFM) measurements. Si ions with energy of 15 KeV and doses of 10¹² or 10¹⁶ cm⁻² are implanted in the Si-SiO₂ structures with an oxide thickness of 20 nm. The ion energy is chosen to produce maximum ion damage at the Si-SiO₂ interface. Some of the implanted samples are simultaneously irradiated by 20 MeV electrons with a flux of about 1×10¹⁵ cm⁻². AFM measurements indicated that the SiO₂ topography does not vary significantly only due to the ion-implantation. Si nanoclusters are observed in a SiO₂ layer after MeV electron irradiation. The measurements show that the size and shape of the Si nanoclusters depend significantly on the dose of the previously implanted ions.

1. Introduction
Silicon nanocrystals generated in a SiO₂ layer have attracted great interest due to their excellent optical and electronic properties [1-2]. The most important property of these nanocrystals is their intense photoluminescence in the visible and red/near-infrared region of the optical spectrum [2-3]. This is why Si nanocrystals are potential candidates for Si-based optoelectronic devices. Klimenkov et al. investigated the influence of the electron irradiation energy on Si-SiO₂ structures implanted with different ion doses. They showed that high-energy electron irradiation causes movement of clusters or atoms on the surface [4] and, alternatively to the thermal treatment, cluster formation can be stimulated by high-energy electron irradiation [5].

In this paper we demonstrate the effects of the high-energy electron irradiation on the topography of SiO₂ samples implanted with a dose of 1.5×10¹² and 1.0×10¹⁶ cm⁻² Si ions (Si⁺). It is shown that Si accumulates and Si nanoclusters are formed on the SiO₂ surface after high-energy electron irradiation.

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We assume that free oxygen is generated during MeV electron irradiation as a result of Si-O bond breaking and radiation-stimulated movement through defects takes place.

In addition, we followed the effect of the Si+ dose on the shape, height and density of Si nanoclusters.

2. Experimental details
The native oxide and contaminations from the surface of n-type CZ Si <100> wafers were removed by HF cleaning and the wafers were then oxidized in dry O2 at 950 °C. The SiO2 thickness was determined to be 20 nm by ellipsometry.

After oxidation, the SiO2-Si samples were implanted through the oxide by 15 KeV Si+ with a dose of 1.5×10^{12} and 1.0×10^{16} cm^{-2}. The ion energy is chosen so as to produce maximum ion damage at the Si-SiO2 interface.

The samples were irradiated with electrons in a Microtron MT-25 system (Flerov Laboratory of Nuclear Reactions, Joint Institute of Nuclear Research - Dubna, Russia). Some of the implanted samples were simultaneously exposed to 20 MeV electrons perpendicular to the SiO2 surface in a vacuum chamber under a pressure of about 1×10^2 Pa. Because of the high energy of the electrons, they penetrated through the whole sample (SiO2 and Si wafer), creating radiation defects. The flux of the electrons was 1.2×10^{15} el/cm². The sample temperature was controlled during the irradiation; it remained near room temperature. The distance between the Microtron window and the samples was 150 mm.

The surface topography of the implanted and irradiated Si-SiO2 structures was studied by an atomic force microscope (AFM) (CP II, Veeco Instruments) in tapping mode. The AFM images were evaluated using Gwyddion software [6]. The size of the surface area investigated was 1 μm × 1 μm for Si+ implantation with a dose of 1.5×10^{12} cm^{-2} and 5 μm × 5 μm for a dose of 1×10^{16} cm^{-2}. The SiO2-Si structures were measured before and after MeV electron irradiation.

3. Results and discussion
Figure 1 presents the AFM images of Si+ samples implanted with a dose of 1.5×10^{12} cm^{-2} before (a) and after 20 MeV electron irradiation (b). It can be seen that after Si+ implantation the SiO2 surface is rough, but unambiguous nanostructures cannot be observed (figure 1a). It is possible for implanted Si atoms to migrate and form small precursor precipitations, because implanted ions and implantation-induced damages are in some activated state. The following 20 MeV electron irradiation (with a flux of 1.2×10^{15} el/cm²) generates silicon nanoclusters, which are observed by AFM on the SiO2 surface (figure 1b). The mean lateral size (l_{mean}) of these nanostructures is in order of 50 nm and the mean height (h_{mean}), between 2.3 to 4.2 nm (table 1). We presume that MeV electron irradiation break Si-O bonds and the free oxygen moves through radiation defects so that Si nanostructures are generated in the SiO2. This is in good agreement with our earlier results [8-9], where it was shown that oxygen accumulates at the SiO2-Si interface after MeV electron irradiation of SiO2-Si structures. In our opinion, the nanoclusters created by MeV electron irradiation have the same structure as the nanoclusters generated by thermal annealing [10] and, therefore, have crystal structure. One of the advantages of MeV electron irradiation compared to thermal annealing is that Si nanoclusters can be observed at the SiO2 surface without additional temperature treatment.

The topography of a high-energy electron irradiated SiO2-Si sample implanted with Si+ with a dose in the order of 10^{16} cm^{-2} is shown in figure 2. In this case l_{mean} of the Si nanoclusters is 35 nm, which is lower than l_{mean} of the Si nanoclusters in the sample implanted with a lower dose of Si. On the contrary, h_{mean} of the Si nanoclusters increases in comparison to those implanted with a lower dose of Si and has values in the range of 9 to 16 nm. The comparison between the AFM images in figure 1 b) and figure 2 show that the nanocluster size depends on the dose of the Si ions implanted in SiO2-Si structures. The variation of the nanocluster size can be related to the reduction of the oxygen...
concentration on the oxide surface. The height of the nanoclusters increases, but the lateral size and the surface density decrease with increasing the dose of implanted Si atoms, as can be seen in figure 2. The ion implantation breaks many of the Si-O bonds and also may knock out atoms from their sites. However, most of the atoms, silicon or oxygen, bond again shortly after being dislodged. One can thus explain the presence of nanoclusters with larger sizes observed after electron irradiation in silicon implanted samples with dose $10^{16}$ cm$^{-2}$ in figure 2 and in table 1.

Figure 1. AFM images of surface topography of Si$^+$ implanted Si-SiO$_2$ sample (a dose of 1.5×cm$^{-2}$): (a) before and (b) after electron irradiation with acceleration voltage of 20 MeV.

Table 1. Mean height and mean length of the Si nanoclusters, created by MeV electrons.

| Sample        | $l_{\text{mean}}$ [nm] | $h_{\text{mean}}$ [nm] |
|---------------|------------------------|------------------------|
| Si$^+10^{12}$+20 MeV | 50                     | 2.3 to 4.2             |
| Si$^+10^{16}$+20 MeV | 35                     | 9 to 16                |
Figure 2. AFM images of Si+ implanted Si-SiO2 sample with a dose of $10^{16}$ cm$^{-2}$ and 20 MeV electron irradiation.

4. Conclusion
The effect of MeV electron irradiation on the surface topography of Si-SiO2 structures implanted with silicon ions with a dose of $1.5 \times 10^{12}$ and $1.0 \times 10^{16}$ cm$^{-2}$, is characterized by AFM measurements. The SiO2 surface topography demonstrates that high-voltage electron irradiation (at a flux of $1 \times 10^{15}$ cm$^{-2}$) creates Si nanoclusters not only in the bulk of SiO2 layers, but also on its surface in both groups of silicon implanted samples. The shape and the density of the Si nanoclusters depend on the dose of implanted ions. The lateral size and the surface density of the nanoclusters are smaller and their height, larger for the samples implanted at a higher dose of Si ions.

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