Surface processing of amorphous optical materials by argon cluster ion beam

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Abstract. The production of a high-quality surface of fused silica is an important task for advanced optical technologies. In this work, the surface of fused silica has been processed by argon cluster ion beam having mean cluster size $N_{\text{mean}}$ ranged from 180 to 1000 atoms/cluster and energy $E$ ranged from 5 to 23.5 keV. The analysis of surface morphology using atomic force microscopy and the spectral power density (PSD) function shows a noticeable smoothing of roughness in different spatial frequency ranges, depending on the cluster ion parameters. To evaluate the processing efficiency, the dependence of the etching rate of SiO$_2$ on the parameters of cluster ions has been investigated. It is shown that the etching rate $v_{\text{etch}}$ is determined by the energy per atom in the cluster $E/N_{\text{mean}}$ and it varies from 0.2 to 20 nm/min with an energy change from 5 to 130 eV/atom.

1. Introduction
The surface roughness is one of the most important parameters of optical materials, largely determining their performance characteristics, including the reflection and scattering of light, the laser damage threshold etc. At present, a number of different techniques, e.g., mechanical polishing, physical sputtering, chemical etching and their combinations, are used to smooth of optical materials surface. In order to obtain qualitative results, it is necessary that the structure is not damaged during the processing and there is no chemical contamination of the material being processed. One of the promising ways to reduce the roughness of different materials is to process their surfaces by the accelerated cluster ion beams formed from supersonic gas jets [1, 2]. It is believed that the cluster interacts with a large number of particles of the target surface in the case of a collision with a solid. Such a collective interaction leads to non-linear effects, lateral sputtering and the possibility of smoothing the surface to a subnanometer level. The use of an inert gas in a high vacuum completely eliminates the contamination of the material during processing.

Although amorphous optical materials are the preferred materials for many applications and devices, there are still no works concerning morphology evolution of glassy materials during processing by cluster ions. In this work, the influence of the argon cluster ion beam on the surface roughness of fused silica is studied experimentally.
2. Experimental results and discussion

2.1. Experimental details
The experiments were performed using equipment described in Ref. [3]. In this work, we used the cluster ion beam formed from a supersonic argon jet under optimum conditions [4]. The experiments have been carried out with the mean cluster ion sizes $N_{\text{mean}}$ of 790 and 1000 atoms/cluster and the cluster ion energy $E$ of 11 and 22 keV at the normal angle of incidence of the cluster ions to the target. The time-of-flight technique has been used to determine the parameters of cluster ion beam [5]. Samples of fused silica were processed by three processing modes: 1) $N_{\text{mean}}=790$ atom/ion; $E=22.5$ keV; 2) $N_{\text{mean}}=1000$ atom/ion; $E=22.5$ keV; 3) $N_{\text{mean}}=790$ atom/ion; $E=11$ keV. The atomic force microscope (AFM) Ntegra Prima HD has been used to observe the surface morphology before and after processing. The AFM measurements were carried out at different scan sizes ($10 \times 10 \, \mu\text{m}^2$ and $80 \times 80 \, \mu\text{m}^2$) with a resolution of 512×512 pixels. The irradiation dose was approximately the same ($1.0 \times 10^{16}$ ion/cm$^2$) for all treatment modes.

2.2. Morphology evolution of fused silica surface
In the first processing mode, the clusters with the highest energy per atom (28.5 eV/atom) were used. The results of polishing of the fused silica by argon cluster ions are illustrated in figure 1, which shows the 3D images of surface at different scales and corresponding values of the root-mean-square roughness $R_q$. At that, $Z=0$ nm corresponds to the level of the lowest point of the profile. A noticeable smoothing of surface irregularities is observed at the smallest dimension of the measurement region, while the roughness decreases slightly at the large region.

For the second processing mode, the mean cluster size $N_{\text{mean}}$ in the beam has been increased from 790 to 1000 atom/ion, which reduced the energy per atom from 28.5 to 22.5 eV/atom, respectively. In the third mode, the treatment was performed at the minimum energy 13.9 eV/atom. After the third processing mode, the total height of the measured profile decreased from 6 nm to 5 nm for field size $80 \times 80 \, \mu\text{m}^2$ and from 5 nm to 3 nm for $10 \times 10 \, \mu\text{m}^2$, as shown in figure 2. Before processing, there were scratches on the surface of the fused silica with a characteristic size of about 1 μm, but such a structure is not observed on the small scan area after processing. The high-frequency roughness has significantly decreased on a small scan area after treatment.

![AFM images](image-url)

**Figure 1.** AFM images of the fused silica surface before (a) and after (b) processing at $N_{\text{mean}}=790$ atom/ion; $E=22.5$ keV.
Figure 2. AFM images of the fused silica surface before (a) and after (b) processing at $N_{\text{mean}}=790$ atom/ion; $E=11$ keV.

The power spectral density (PSD) function is the Fourier transforms of the profile height distribution measured by AFM in the selected scan area, and it is especially helpful for a more detailed description of the roughness of the optical materials. The spatial frequency $\nu$ is a measure of how often the same height repeats per unit of distance [6]. The PSD functions obtained from AFM data of the fused silica surface before and after by argon cluster ions in different modes are shown in figure 3. For all processing modes, the PSD functions decrease, starting from about 0.2 μm$^{-1}$ and the maximum reduction in roughness is observed in the high spatial frequency range ($\nu > 1$ μm$^{-1}$). This corresponds to a reduction in the root-mean-square roughness $R_q$ by 43% at the scale of 10×10 μm$^2$. In the middle-frequency range ($\nu \sim 0.1÷1$ μm$^{-1}$), the roughness decreases by an average of 2.5 times in the same way after treatment in modes 1 and 2 and significantly more (up to 8 times) in mode 3 with the lowest energy per atom in the cluster. The roughness $R_q$ decreased by 3÷5 times in the whole range after processing in mode 2, while, after the 3rd mode the roughness $R_q$ decreased by an average of more than 1 order of magnitude. The change in roughness after different processing modes is more pronounced in the high-frequency range ($\nu = 1÷10$ μm$^{-1}$). These results indicate that the processing by argon cluster ions allows effective smoothing the surface of fused silica in the middle- and high-frequency roughness ranges. Although further experimental data are still required, it can be assumed that by combining the processing by clusters with different characteristics (mean cluster size and kinetic energy) can obtain effective smoothing of the surface in a wide range of spatial roughness frequencies.

2.3. Determination of etching rate

It is known that the etching rate is the important parameter for the materials processing. The fused silica is the purest amorphous form of SiO$_2$, and therefore, 400 nm thick SiO$_2$ films obtained by dry thermal oxidation of silicon wafer were used as targets. The film thickness was measured by laser ellipsometer. The etching of SiO$_2$ films has been carried out using Ar cluster ions by varying either the mean cluster size (from 150 to 1000 atoms/cluster) or the cluster energy (from 5 to 23 keV) at the normal angle of incidence of the cluster ions to the sample. Since the sputtering yield depends on the energy per atom in the cluster [7], the dependencies of the etching rate on $E/N$ were analyzed. Since the beams of the different cluster sizes are focused differently, the current ion density was varied depending on the processing mode. Therefore, in order to correctly compare the etching rates in different modes, they were recalculated for the same current density $j = 1 \mu$A/cm$^2$. 

\begin{align*}
\text{Rq} &= 0.89 \text{ nm} \\
\text{Rq} &= 0.71 \text{ nm} \\
\text{Rq} &= 0.79 \text{ nm} \\
\text{Rq} &= 0.54 \text{ nm}
\end{align*}
As can be seen in figure 4, the energy per atom in cluster varies within the range from 5 to 130 eV/atom. These limitations are related to the maximum permissible ranges of accelerating cluster energy and mean cluster size for our equipment. The etching rate varies nonlinearly and it increases by 2 orders of magnitude, with a change in the energy per Ar atom by a factor of 26. In [8], it was shown...
that the etching rate of fused silica by a beam of argon monomer ion at the current density of 0.5 mA/cm² was varied from 15 to 40 nm/min with a change in the ion energy from 500 to 1500 eV. A comparison with our results shows that the cluster ion beams give a commensurate etching rate at a much lower energy of the particles due to the collective effects of interaction with the surface.

3. Conclusion

The influence of accelerated argon cluster ion beam on the surface morphology of fused silica has been studied experimentally. The maximum reduction in the root-mean-square roughness $R_q$ has reached 18% at scan area of 80×80 μm² and 80% at 10×10 μm². The roughness has been maximally reduced in the high spatial frequency range ($ν = 1÷10$ μm⁻¹). It was shown that by changing the parameters of cluster ion beam (mean cluster size and energy per atom in the cluster), it was possible to effectively smooth the surface of fused silica within a wide range of spatial frequencies.

The etching rates of silicon dioxide have been measured at different energies per atom in the cluster for a more detailed description of the effect of cluster ions on the fused silica. It was confirmed that the cluster ion beam can provide the etching rate of fused silica comparable with the conventional ion beam at a much lower particle energies and, thus, at the low damage of the treated surface.

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