Formation of nanostructures on the surface of KTP single crystals by argon cluster ion beam

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Abstract. In this work, the surface of potassium titanyl phosphate (KTP) single crystals has been processed by argon cluster ions having low energy per atom $E/N_{\text{mean}} = 12.5$ eV/atom. The formation of periodic nanostructures has been studied using atomic force microscopy (AFM) and the power spectral density (PSD) functions. To evaluate the processing efficiency, the dependence of the etching rate of KTP on the incident angles of cluster ions has been investigated. It is shown that the average etching rate $\langle v_{\text{etch}} \rangle$ varies between 0.46 and 1.1 nm/min with an incident angle change from 0° to 70°.

1. Introduction
Surface self-organizing nanostructures are of wide interest for various practical applications [1]. It was previously shown that such structures can be formed on the surface of various materials using monomer ion [2–5] and cluster–ion beams [6]. Potassium titanyl phosphate (KTiOPO$_4$, KTP) single crystals are currently widely used in optical elements. Effective functional KTP characteristics allow KTP single crystals to be used in the frequency converters of high–power lasers and electro–optical modulators, parametric generators of visible and infrared radiation range, integrated–optic elements, etc. [7, 8].

2. Experimental results and discussion
2.1. Experimental details
In this work, chemical-mechanical polished KTP single crystals were used as targets. The description of the experimental equipment is given in Ref. [9]. Taking into account the fact that the lower energy per atom contributes to a smaller disturbance of the subsurface layer [10], the surface of a KTP single crystal was treated with a cluster–ion beam formed of a supersonic argon jet at an energy of $E = 10$ keV with an average cluster size $N_{\text{mean}} = 800$ atoms/cluster. Thus, the energy per atom in the cluster was only $E/N_{\text{mean}} = 12.5$ eV/atom. The processing of the surface target was carried out at different incident angles of the cluster–ion beam: 0°, 15°, 30°, 45°, 60°, 70°. The maximum irradiation dose was $3.7 \times 10^{15}$ ions/cm$^2$. The time-of-flight technique has been used to determine the parameters of cluster ion beam [11]. The atomic force microscope (AFM) Ntegra Prima HD has been used to observe the surface morphology after processing. The AFM measurements were carried out at the same scan sizes (2×2 μm) with a resolution of 1024×1024 pixels.
2.2. Formation of nanostructures on the KTP surface

Figure 1 shows 3D images of the KTP surface after processing by argon cluster ions and the corresponding roughness parameters $R_q$. The height of the profile irregularities is counted from the lowest point of the profile ($Z = 0$ nm). It can be seen that the surface profile changes slightly when the incident angles of the cluster ions are less than 45°.

![AFM images of the KTP surface after processing at $N_{\text{mean}}$=800 atoms/cluster; $E$=10 keV and the incident angle of cluster ions: (a) 0°, (b) 15°, (c) 30°, (d) 45°, (e) 60°, (f) 70°.](image)

Pronounced periodic nanostructures are formed at incident angles of 45° and 60°. It should be noted that the root–mean–square roughness $R_q$ increases significantly: from 0.27 to several nm. As it is shown in Ref. [12], such nanostructures contribute to the quality improvement of the antireflection coatings.

For the analysis of surface morphology, the power spectral density (PSD) function that is the Fourier transforms of the profile height distribution was used. The profile height distribution has been measured by AFM in the selected scan area. The spatial frequency $\nu$ is a measure of how often the same height repeats per unit of distance [13]. PSD functions of the KTP surface after processing by argon cluster ions at the different incident cluster angles are shown in Figure 2. It is seen that the PSD functions before the formation of nanostructures differ slightly in accordance with the 3D AFM images and $R_q$. The main difference in the processing at small angles ($\leq 30°$) is observed in the range of spatial frequencies from 5 to 30 $\mu$m$^{-1}$, where the roughness grows gradually. The formation of periodic nanostructures is accompanied by a significant increase in roughness over the entire range of spatial frequencies. The peaks of the PSD functions (marked by arrows in Fig. 2) correspond to the frequency of nanostructures.
on the sample surface. It should be noted that with an increase in the angle of incidence, the characteristic size (wavelength) of the nanostructure increases significantly and it is 167, 200 and 334 nm for angles of 45°, 60°, and 70°, respectively.

Figure 2. PSD functions of the KTP surface before and after the formation of periodic nanostructures at the different incident angles of the cluster ions.

2.3. Determination of etching rate
In addition to analyzing the surface morphology, the angular dependence of the etching rate of KTP surface has been investigated. Since the current density in the experiments at different incident cluster angles varied slightly, the etching rates were recalculated for the average current density $\langle j \rangle = 0.63 \mu A/cm^2$. As it can be seen in Figure 3, the average etching rate $\langle v_{etch} \rangle$ varies nonlinearly and it increases by 2.4 times of magnitude with a change in the incident angle from 0° to 45°. Interestingly, the angle of the highest etching rate corresponds to the angle of formation of periodic nanostructures. It should be noted that a noticeable increase in etching rate begins at a more acute angle (30°), although the surface morphology remains almost unchanged as compared with the incident angles of 0° and 15°.

Figure 3. Angular dependence of etching rate at the same current density.
Summary
The influence of accelerated argon cluster ion beam on the surface morphology of KTP has been studied experimentally. It was shown that periodic nanostructures were not formed at small incident angles of the cluster ions (≤ 30°). Since the etching rate increases 1.6 times at the angle of 30°, and the surface roughness remains almost unchanged, the thirty–degree processing can be used to etch the surface effectively without much changing in the surface morphology. Surface processing at angles of 45° and 60° is effective for forming the most uniform nanostructures with a wavelength of 167 and 200 nm, respectively. Such periodic nanostructures can be used on substrates to improve the quality of antireflective coatings.

Acknowledgments
This work has been supported by the Ministry of Education and Science of the Russian Federation (Grant No 11.1402.2017/4.6). The experimental results were obtained by the use of the facilities of the Shared Equipment Center NSU “Applied Physics”.

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