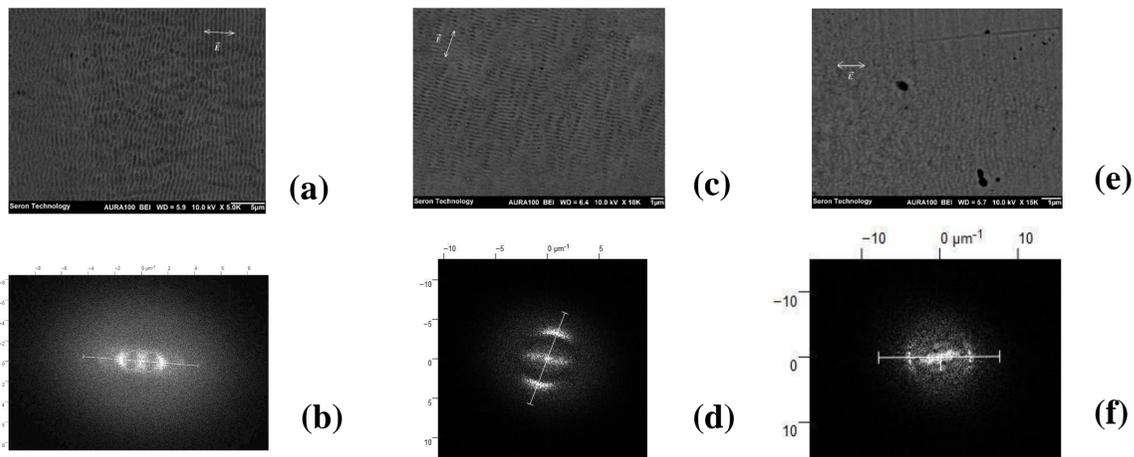


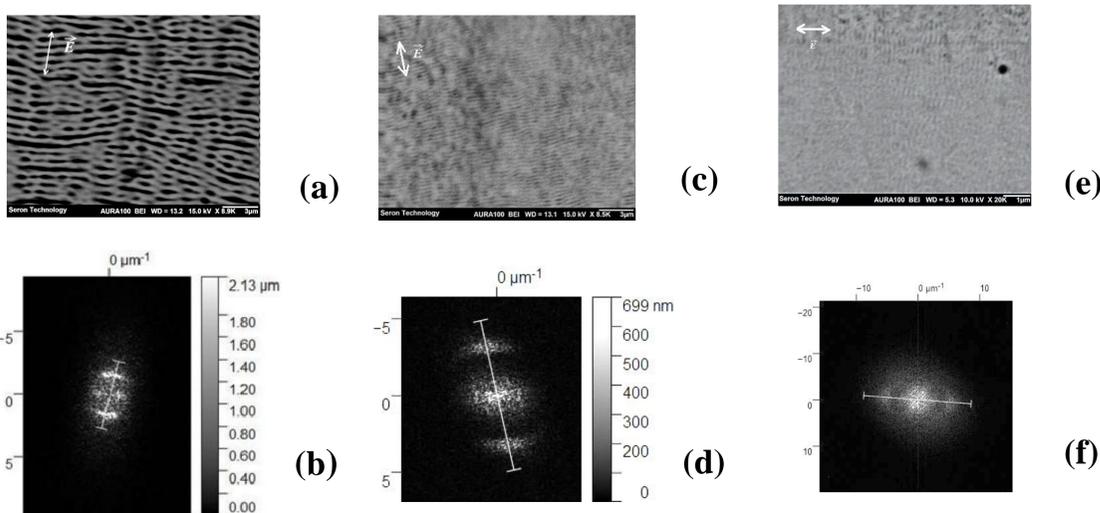
Report deals with the impact of femtosecond (fs) radiation with different wavelengths (fundamental wavelength (800 nm), the second (400 nm) and the third (266 nm) laser harmonics) on the efficiency of the formation of laser-induced periodic surface structures (LIPSS) on the metal surfaces (Ag, Cu, W) for its application in plasmon nanoresonator structures [1]. The fs-laser treatment was realised in the multi-pulse regime in air or argon environment. The influence of fs-laser radiation on the sample morphology has been analysed by scanning electron microscopy (SEM) and 2D Fourier transform of SEM images.



- **Silver (Ag) surface** was treated with linearly polarized fs-laser radiation with pulse duration of ~ 130 fs at the repetition rate of 1 kHz with mean power of 270, 135 and 33 mW for wavelengths of 800, 400 and 266 nm, respectively.

Fig. 1: SEM images of fs-laser treated Ag surface and 2D Fourier transform of these SEM images: (a), (b) – for the wavelength of 800 nm (the pulse irradiation energy density of $2.45 \text{ mJ}\cdot\text{cm}^{-2}$); (c), (d) – for the wavelength of 400 nm (the pulse irradiation energy density of $14.2 \text{ mJ}\cdot\text{cm}^{-2}$); (e), (f) – for the wavelength of 266 nm (the pulse irradiation energy density of $13 \text{ mJ}\cdot\text{cm}^{-2}$). Samples have been treated in air.

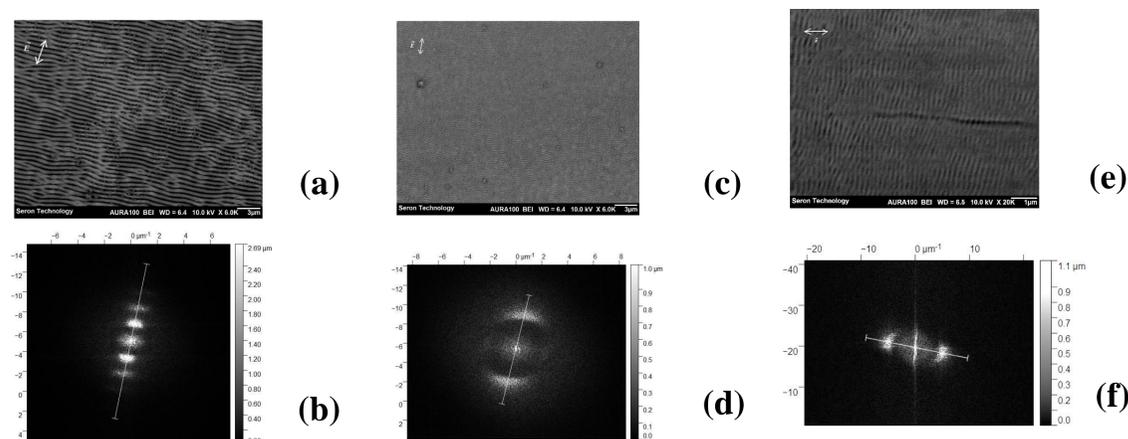
- **Results:** Fs-laser radiation with a wavelength 800 nm forms LIPSS orientated perpendicular to the laser beam polarization with a period of 649 ± 46 nm, quasi-grating orientated along \vec{E} with period of $2.15 \mu\text{m}$; fs-radiation with a wavelength 400 nm also causes similar periodic structures with periods 302 ± 16 nm ($\perp \vec{E}$) and $2.15 \mu\text{m}$ ($\parallel \vec{E}$); under the third laser harmonic (266 nm) LIPSS with a period of 257 nm orientated perpendicular to the laser beam polarization has been formed.



- **Copper (Cu) surface** was treated with linearly polarized fs-laser radiation with pulse duration of ~ 130 fs at the repetition rate of 1 kHz with mean power of 270, 150 and 40 mW for wavelengths of 800, 400 and 266 nm, respectively. Samples have been treated in air.

Fig. 2: SEM images of fs-laser treated Cu surface and 2D Fourier transform of these SEM images: (a), (b) – for the wavelength of 800 nm (the pulse irradiation energy density of $4.1 \text{ mJ}\cdot\text{cm}^{-2}$); (c), (d) – for the wavelength of 400 nm (the pulse irradiation energy density of $2.27 \text{ mJ}\cdot\text{cm}^{-2}$); (e), (f) – for the wavelength of 266 nm (the pulse irradiation energy density of $0.61 \text{ mJ}\cdot\text{cm}^{-2}$).

- **Results:** It has been observed LIPSSs orientated perpendicular to the laser beam polarization with periods of 639.8 ± 7.8 nm, 331 ± 6 nm and 195 ± 1.5 nm - for irradiation wavelength 800 nm, 2nd and 3d laser harmonics, respectively; quasi-gratings orientated along \vec{E} with period of $1.9 \mu\text{m}$ and $\sim 0.5 - 0.9 \mu\text{m}$ - for irradiation wavelengths 800 nm and 400 nm, respectively.



- ✓ **Tungsten (W) surface** was treated with linearly polarized fs-laser radiation with pulse duration of ~ 130 fs at the repetition rate of 1 kHz with mean power of 160, 97 and 30 mW for wavelengths of 800, 400 and 266 nm, respectively.

Fig. 3: SEM images of fs-laser treated W surface and 2D Fourier transform of SEM images: (a), (b) – for the wavelength of 800 nm (the pulse irradiation energy density of $1.6 \text{ J}\cdot\text{cm}^{-2}$); (c), (d) – for the wavelength of 400 nm (the pulse irradiation energy density of $0.97 \text{ J}\cdot\text{cm}^{-2}$); (e), (f) – for the wavelength of 266 nm (the pulse irradiation energy density of $0.3 \text{ J}\cdot\text{cm}^{-2}$). Samples have been treated in air.

- ✓ **Results:** It has been revealed LIPSSs orientated perpendicular to the laser beam polarization with period of 578 ± 99 nm and additional feature with periodicity of 383 nm - for irradiation wavelength 800 nm, 285 ± 43 nm and 193 ± 32 nm - for the 2nd and 3d laser harmonics, respectively.

CONCLUSIONS: Controlled formation of LIPSS on studied metals under the fs-laser 3rd harmonic treatment, to our knowledge, was achieved for the first time. Additional surface features and the slight period reduction could be caused by the temperature effects and the hydrodynamic instabilities of the melt during the laser processing. Breakthrough improvement of the homogeneity filling of LIPSS in argon environment compared to the treatment in air was not observed that confirms the effectiveness of processing in air.

[1] O.A. Yeshchenko, V.Y. Kudrya, A.V. Tomchuk, I.M. Dmitruk, N.I. Berezovska et al. Plasmonic nanocavity metasurface based on laser-structured silver surface and silver nanoprisms for the enhancement of adenosine nucleotide photoluminescence // ACS Applied Nano Materials. – 2019. V. 2. – No 11. – p. 7152-7161.

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