Nanostructuring of Rough Aluminum Surface by Ultrashort Laser Pulses: Influence of Laser Fluence

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Abstract: Compared to noble metals, aluminum is unfavorable for plasmonics. Proper nano-structuring of aluminum surface improves its plasmonic properties even for contaminated and rough original surface. This method allows production and repair of cheap plasmonic elements.
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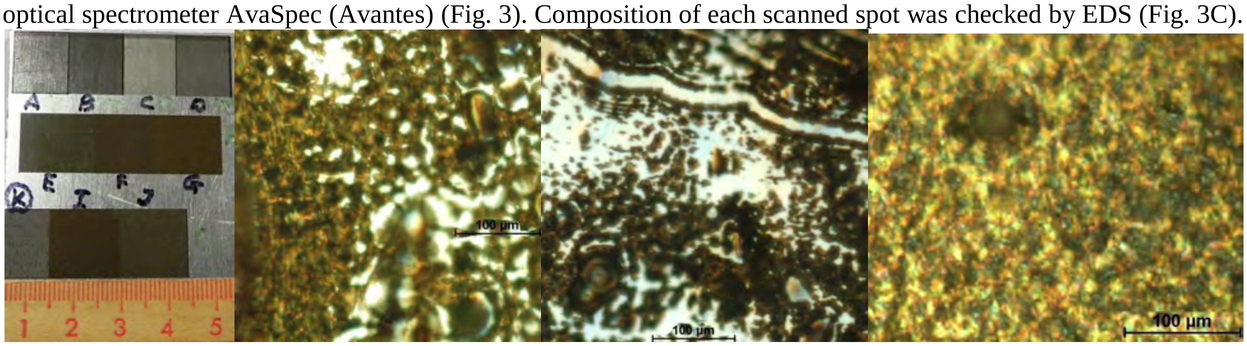
1. Introduction

Nanostructuring of metal surfaces by ultrashort laser pulses significantly modifies their fundamental properties, e. g., reflectivity spectrum, work function, and wetting properties for various liquids [1-4].

Modification of the properties of nanostructured metal surfaces is attributed to laser-induced micro- and nano-structures [1-4]. The fundamental mechanisms of their formation by ultrashort laser pulses are still not clear. The most popular approach considers formation of laser-induced periodic surface structures (LIPSS) by interference of incident laser pulses and scattered waves at metal surface that creates periodic interference pattern [1-7]. This model explains formation of the periodic surface ripples, but does not explain formation of nanostructures observed in most experiments [1-6]. Therefore, additional insight into the basic mechanisms of LIPSS formation is still required. In this connection it is notable that the traditional approaches to study ultrafast LIPSS meet a significant conceptual problem: a majority of experiments [1-6] have been performed on polished metal surfaces without evident sources of the scattered waves to produce the interference pattern of fluence/intensity distribution. To fix this gap, we have performed experiments on LIPSS generation on a metal surface with artificial roughness produced by chemical etching in acids. Many applications of the nano-structured metal surfaces are associated with intensive usage of the surfaces in chemically aggressive environment [1-4] that damages metal surfaces. The laser-induced nanostructures are destroyed by the environment. This problem motivates our development of approaches to restoration of nano-structured surfaces after the environmental damage. We present proof-of-concept experiments on restoration of specific optical properties of the nanoscturctured surfaces.

2. Experiment details

Aluminum plate was first polished to mirror quality. Its reflectivity spectrum was measured by integrated sphere and was utilized as a reference to characterize reflectivity modification by the following procedures. The plate was made rough by 50-μm sand paper followed by etching in sulfur acid for 3 minutes to mimic surface decay in a chemically aggressive environment. After washing in water and cleaning with methanol and acetone, the plate was scanned with ultrashort laser pulses (CPA-2210 (Clark MXR); Ti:sapphire regenerative amplifier; 150 fs; 772 nm; repetition rate 1.0 kHz, linear polarization; linear scanning speed 1 mm/s; inter-line step 0.1 mm; variable fluence) to produce areas of different color on the rough surface (Fig. 1A) at normal incidence. The scanned areas were studies with SEM and optical microscope (Fig. 1, 2). Reflectivity spectra were taken in the range 250 - 1050 nm with integrated sphere and optical spectrometer AvaSpec (Avantes) (Fig. 3). Composition of each scanned spot was checked by EDS (Fig. 3C).

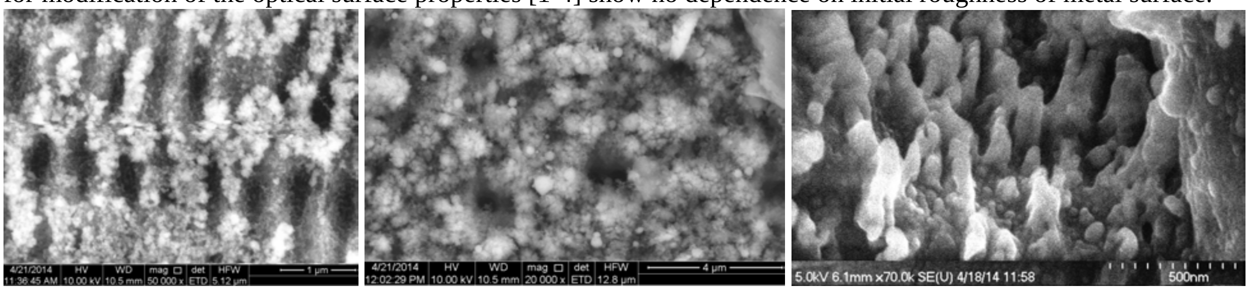


A B C D

Fig. 1. Aluminum sample (A) with 10 scanned areas and structure of scanned areas A (fluence 0.10 J/cm^2) (B), D (fluence 0.09 J/cm^2) (C), and G (fluence 0.16 J/cm^2) (D) delivered by high-resolution Nomarski optical microscope.

3. Discussion and conclusions

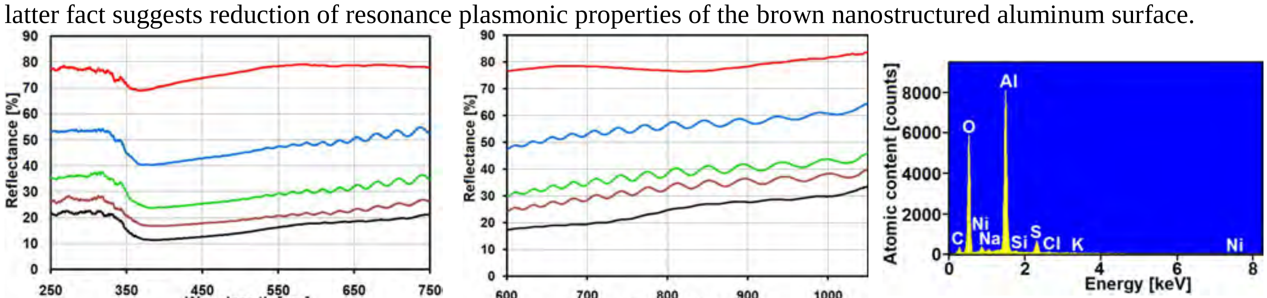
The chemical etching produced multiple shallow pits and spots on the metal surface with irregular shape of edges. Size of the etched structures varied from 100 nm up to 100 μm with density about 50 1/mm^2. The pits served as chaotic scattering centers for incident laser pulses. As long at laser fluence was low enough to produce grey colors of scanned areas (areas A through D in Fig. 1A), LIPSS were produced by scattering and interference of light at the roughness in agreement with the traditional model. Evident signatures of the interference include periodic ripples at extended linear roughness and circular structures in the vicinity of round spots (Fig. 1). Total map of the micro- and nanostructures on the surface treated with low-fluence pulses is a result of interference of multiple waves scattered by all roughness objects within a laser spot. The low-fluence scanning by femtosecond pulses also produced specific periodic ripples of reflectivity spectra (Fig. 3 A and B) attributed to plasmon resonances [7]. SEM detected the same structures reported for polished metal surfaces [5,6]: residual markers of scanning tracks of laser beam, cloud-type nanoaggregates, and periodic ripples covered with nanostructures (Fig. 2). Therefore, the nanostructures responsible for modification of the optical surface properties [1-4] show no dependence on initial roughness of metal surface.



A B C

Fig. 2. SEM images of the nanostructures from areas A (A), D (B), and area G (C).

Scanning with higher fluence (areas E through J Fig. 1) significantly erased the evident interference patterns and produced golden-brown color of the surface (Fig. 1D). SEM showed the major families of the laser-induced nano-structures were the cloud-type aggregates, porous and pillar-type structures (Fig. 2C) combined with the residual markers of scanning tracks of laser beam. Reflectance of the brown areas was significantly reduced compared to the reference signal from aluminum mirror, but reflectance spectra were smooth and showed no ripples (Fig. 3). The latter fact suggests reduction of resonance plasmonic properties of the brown nanostructured aluminum surface.



A B C

Fig. 3. Reflectivity spectra (A and B) of the un-scanned rough surface (red), area A (brown), area C (fluence 0.080 J/cm^2) (blue), area D (green), and J (fluence 0.15 J/cm^2) (black). Fig. C shows a representative EDS spectrum from laser-treated areas of the aluminum surface.

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4. References

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