



# Evolution of aluminum surface irradiated by femtosecond laser pulses with different pulse overlaps



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## ABSTRACT

We report the modification of aluminum surfaces by femtosecond laser pulses with different pulse overlaps in ambient air. Several kinds of structural colors can be generated on the aluminum surfaces by varying the pulse overlap from –27% to 99%. The SEM images reveal that the structural colors of the modified area depend on the surface morphology. Furthermore, X-ray photoelectron spectroscopy and X-ray diffraction spectra show that the content of Al<sub>2</sub>O<sub>3</sub> increases while that of Al(OH)<sub>3</sub> decreases with the increase of the pulse overlap. Only Al<sub>2</sub>O<sub>3</sub> is formed on the aluminum surfaces when the pulse overlap is larger than 93%.

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## 1. Introduction

The modification of metal surfaces with femtosecond laser irradiation has become a hot research topic and attracted lots of interest in recent years. This novel method has the advantage of producing metallic nanostructures with unique properties, which have potential applications in life sciences [1,2], optical data storage [3], microfluidic devices [4,5], powder metallurgy [6] and structural colors [7–11]. For the successful metal surface processing with femtosecond laser pulses, it is essential to optimize the laser processing parameters. These parameters include laser fluence, pulse overlap, the distance between adjacent scanning lines, laser incidence angle and so on. It has been reported that aluminum films with gray, golden and black colors can be fabricated by adjusting the laser fluence and the number of exposure pulses to induce nanostructures, microscale cloudy aggregates, and slight periodic marks [12]. The laser incidence angle can also affect the diffraction grating period [13]. Compared to other processing parameters, fewer investigations have been carried out on the evolution of modifying surface by different pulse overlaps.

Aluminum is a common energetic material, and it has the characteristics of low melting point. It can be easily oxidized when exposed to air. In this work, we modified the aluminum surfaces

by scanning the femtosecond laser beam over the surfaces. The influence of the pulse overlap on processing result was studied in detail through observing the induced micro/nano structure, and analyzing the composition of the modified surface. Results demonstrated that various structural colors can be generated by properly controlling the pulse overlap. Such an investigation is supposed to be able to provide a promising method for producing controllable structural colors.

## 2. Experimental setup

A regenerative amplified Ti: Sapphire femtosecond laser system (Spectra-physics) with the pulse duration of 120 fs, the central wavelength of 800 nm and the repetition rate of 1 kHz was applied to process the aluminum surfaces (99.99% purity). The experimental setup is schematically illustrated in Fig. 1. The pulse energy could be continuously varied by using the combination of a  $\lambda/2$  wave plate and a Glan-Taylor polarizer. The average power of the laser pulse is measured by a power meter (Coherent, FM10), which is set behind the polarizer. The laser beam is focused by a biconvex lens with 50 mm focal length, and the diameter of the focal spot is about 15  $\mu\text{m}$ . The aluminum sheets are mounted horizontally on a high-precision computer controlled linear translational stage. A series of pulse overlaps in the  $x$ -direction on the aluminum surfaces are obtained through changing the speed ( $v$ ) of the linear translational stage. Before the laser treatment, the aluminum sheets are cleaned with acetone to remove organic dopants on the surface.

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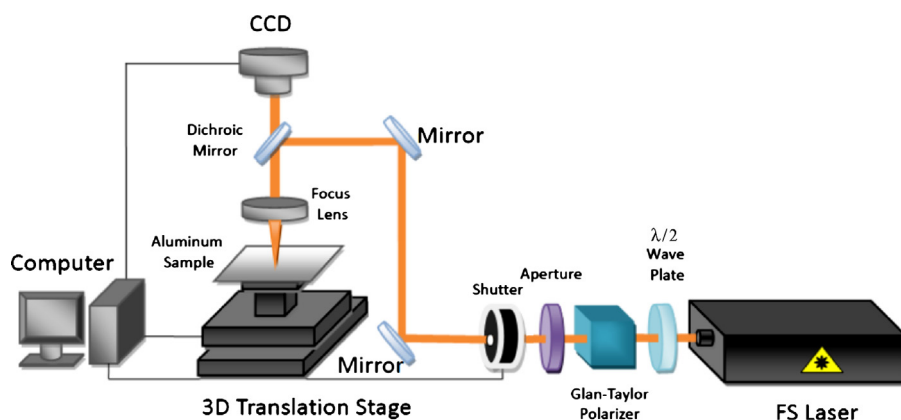


Fig. 1. Schematic illustration of the experimental setup.

The experiments are conducted by scanning the laser focused spot across the aluminum surface in air environment while controlling the pulse overlap [13,15]. The morphology of the modified surface is imaged by using a field emission scanning electron microscopy (FESEM) (JSM-6700F). The X-ray photoelectron spectrum (XPS) of the aluminum surface is measured with ESCALAB 250 (Thermo-VG Scientific). The X-ray diffraction spectra are obtained with a Bruker D8 Advance X-ray diffractometer, which is equipped with Ni-filtered Cu  $K\alpha 1$  radiation at room temperature.

### 3. Results and analysis

Firstly, the possibility of producing structural colors on aluminum surfaces by altering the pulse overlap is examined. In experiments, the average laser fluence is  $5.1 \text{ J/cm}^2$ . The pulse overlap in the  $y$ -direction is chosen to be  $-233\%$  (hatch distance =  $50 \mu\text{m}$ ). The aluminum surfaces appear golden, white, gray and black when the overlap is set as  $-7\%$ ,  $33\%$ ,  $93\%$ , and  $99\%$ , respectively. The corresponding scan rate is 16, 10, 1, and  $0.1 \text{ mm/s}$ . The typical results are demonstrated in Fig. 2. It is found that the induced structural colors become brighter and more vivid with the decrease of the pulse overlap. All these structural colors exhibit the same color under various viewing angles.

To identify how the surface structures are affected by the pulse overlaps, a detailed SEM study is performed on the aluminum sample surface. Fig. 3 shows the SEM images of an unexposed aluminum surface. Figs. 4–7 reveal the SEM images of irradiated targets at various pulse overlaps, which are  $-7\%$ ,  $33\%$ ,  $93\%$  and  $99\%$ , respectively.

It is clear from Fig. 4(a) that adopting the pulse overlap in  $x$ -direction of  $-7\%$  endows the modified aluminum surface with a smooth surface as compared to the untreated surface in Fig. 3. The more detailed structures can be seen from Fig. 4(b) to (d). Many nanoparticles, whose diameters are in the order of hundreds of nanometers, are embedded on the substrate surface. As shown in Fig. 4(c), these nanoparticles can be generally classified into two shapes. One is like a flower with diameter of approximately three

hundred nanometers in diameter, and the other is like a sphere with diameter of approximately one hundred nanometers. Fig. 4(d) confirms that these nanoparticles contain many protrusions and micro scale aggregates, which are formed by fusing nanoparticles together on the surface.

When the pulse overlap increases to  $33\%$ , slight stripe structures dispersed on the substrate surface appear as shown in Fig. 5(a). Nonuniform cracks of different scale are also formed, as evidently displayed in Fig. 5(b). When the surface morphology is magnified 20,000 times as shown in Fig. 5(c), block-like structures and grainy structures can be found clearly. This is the evidence that the liquid phase exists in the laser ablation process [14]. Higher pulse overlap makes more surface materials vaporize, therefore both the quantity and the size of spheres decrease and more fluffy structures are generated as shown in Fig. 5(d).

Fig. 6(a) represents the surface topography of representative groove structures generated by scanning a laser beam horizontally across a sample surface at a  $93\%$  overlap. The period of the groove is  $50 \mu\text{m}$ , which is determined by the distance of adjacent horizontal scanning lines. The width of groove is about  $15 \mu\text{m}$ , which is approximately equal to the diameter of the focused laser beam. Compared with the case of  $33\%$  overlap, the scale formed spherical and block-like nanoparticle aggregates much bigger as shown in Fig. 6(b). More fluffy porous can be generated, which can be found in Fig. 6(c) and (d).

When the overlap reaches  $99\%$ , the processing lines can produce deeper grooves on the aluminum surface as shown in Fig. 7(a) and (b). From the cross-sectional SEM image, Fig. 7(e), the depth of the grooves is measured to be ranging from  $89$  to  $125 \mu\text{m}$ . Fig. 7(b), (c) and (d) demonstrate the black aluminum surface has a rich variety of structures including types of nano- and micro-scale voids, nanoprotusions, microscale aggregates formed by coalescence of nanoparticles [8].

In order to further study the influence of the pulse overlap on surface modifications, we also considered the case of the pulse overlap of  $12\%$  ( $v = 13 \text{ mm/s}$ ). Under such a condition, another kind

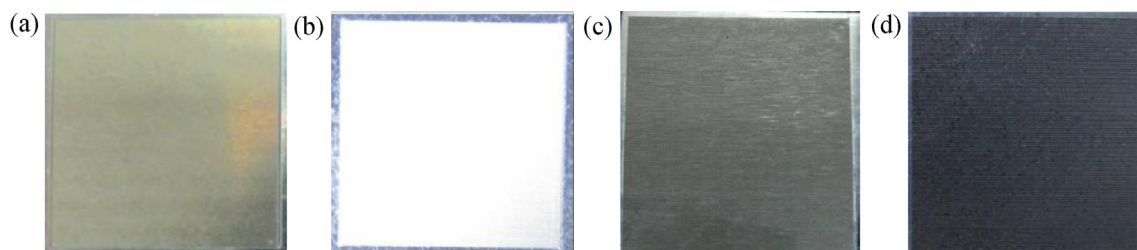


Fig. 2. Photograph of modified aluminum: (a) golden; (b) white; (c) gray; (d) black.

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