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Time dependency of the laser-induced nanostructuring process of chromium layers with different thicknesses on fused silica

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ABSTRACT

Nanostructures exhibit a raised importance in manifold application fields like electronics and optics. The laser irradiation of thin metal layers allows the fabrication of metal nanostructures induced by a melting and deformation process where the resultant structures are dependent on the laser and metal layer parameters. However, for an optimization of this process a detailed physical understanding is necessary. Therefore, the dynamics of the metal layer deformation process was measured by time-dependent reflection and transmission as well as shadow graph measurements at different KrF excimer laser parameters (laser fluence and number of laser pulses) and metal layer thicknesses were used. Magnetron-sputtered thin chromium films with a thickness from 10 to 100 nm on fused silica substrates were studied. Based on the optical measurements the liquid phase lifetime of the metal was estimated and compared with the calculated lifetime using a simple thermodynamic model.

1. Introduction

Nanostructures exhibit a raised commercial interest, e.g. in microelectronic applications. However, the fast and cost-effective production is a big technological challenge where laser methods using self-organization have an outstanding potential.

Different laser methods using self-organization processes of surfaces were developed: 3-dimensional fs laser metal nanostructuring [1–3] and ripple structures formation [4–7] as well as nanostructuring of metal layers [8–12].

This study is focussed on the nanosecond nanostructuring process of thin metal layers. This process is dominated by two physical effects: the laser–solid interaction as well as the mass transport in the liquid. The laser–solid interaction [13–19] can be theoretically described by a heat equation [15,16] as well as the mass transport in the liquid by a Navier–Stokes equation [10,20–26]. The consideration of both effects allows a good theoretical description of the laser-induced melting process [11,27].

A multipulse low laser fluence treatment of the thin metal layer results in the formation of metal droplets [10] where the position of the metal droplets can be controlled by the laser beam profile down to the sub-μm range [28]. The resultant metal droplets dependent on the metal layer thickness is extensively studied [9,23,26,29,30].

Furthermore, the laser beam scanning procedure [31] as well as the lateral limitation of the metal film [32] influences the nanostructuring process.

However, for the prediction and defined fabrication of nanostructures, in the case of an incomplete metal droplet formation process [26,27,29], the knowledge of the time dependency of the laser-induced liquid metal film is necessary.

Among other things the droplet formation process can be used for the nanostructuring of the substrate surface [11,12].

In this study, the time dependency of a KrF excimer laser irradiated thin chromium layer at different layer thicknesses on fused silica was measured by the detection of the time-dependent transmission and reflection signal using a probe cw laser.

2. Experimental details

The laser-induced modification of a magnetron-sputtered chromium film with different layer thicknesses $d = 10$ nm, 20 nm,

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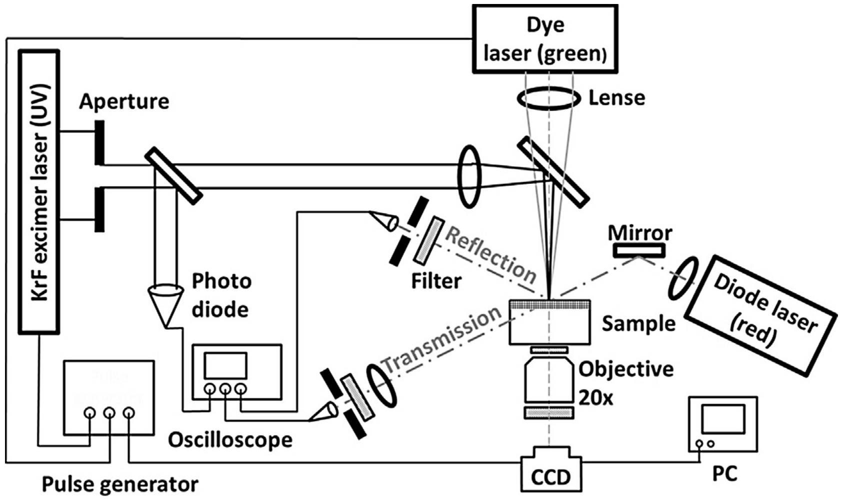

Fig. 1. Schematic illustration of the experimental set-up.

50 nm, and 100 nm on fused silica was studied, where the chromium film changes was induced by a KrF excimer laser irradiation with a wavelength of $\lambda = 248$ nm and a pulse duration of $\Delta t_p = 25$ ns. The time-dependent behaviour of the melting characteristics of thin metal layers on dielectric substrates was studied with a pump-probe set-up. The used experimental set-up is schematically illustrated in Fig. 1.

Furthermore, the metal layer/fused silica system was irradiated by a red cw diode laser with a wavelength of $\lambda = 660$ nm and a laser power of $P = 130$ mW, where the transmitted and reflected signal of the diode laser was detected by a photodiode with a time resolution of $\Delta t_{ph} = 1$ ns. The time-dependent signal of the photodiode was recorded by a 1 GHz oscilloscope, where the excimer laser beam was used as reference signal. To reduce the influence of the UV laser radiation onto the measured reflection and transmission signal an aperture/filter combination was used. In Fig. 2 an exemplary transmission and reflection signal is shown. Furthermore, the experimental set-up allows the direct time-dependent optical imaging of the metallic surface structure. Therefore, the Cr/fused silica system was additionally irradiated by an electronically delayed N_2 laser pumped Coumarin 153 green dye laser ($\lambda = 543$ nm, $\Delta t_p = 1$ ns, $E = 1$ mJ) and the image of the surface structure was displayed by a 20× objective with optical correction of the fused silica thickness into an externally triggered CCD camera. The signal was improved by a band pass filter system for blocking UV and red laser radiation. The time delay $\Delta\tau$ between the KrF excimer laser, the dye laser, and the recording point of the camera could be adjusted by a freely programmable pulse generator. For this study the delay time between both pulsed lasers was fixed at 20 µs.

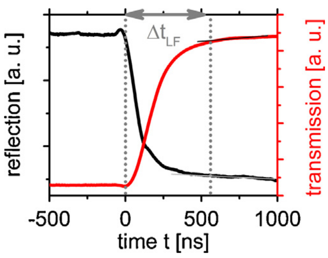

Fig. 2. Exemplary reflection and transmission signal at 20 nm Cr, $N = 1$, $\Phi \sim 930$ mJ/cm^2 (the reflection and transmission signal were smoothed by a low pass filter). (For interpretation of the references to colour in the text, the reader is referred to the web version of this article.)

3. Experimental results

The irradiation of the chromium layer on fused silica with low laser fluences results in a distinct modification of the surface topography induced by a mass transport in the liquid phase [11,33]. In order to analyze the time behaviour of the deformation process the reflection and transmission signal were detected.

In Fig. 2 the exemplary variation of the reflection and transmission signal of 20 nm Cr on fused silica induced by the irradiation of the sample with approx. 930 mJ/cm^2 is shown. The irradiation of the 20 nm Cr sample at the first pulse results in a distinct increase of the transmission signal and a decrease of the reflection signal.

The variation of the reflection and transmission signal directly starts at the beginning of the irradiation with the ultraviolet laser pulse at $t = 0$. Besides the irradiation-induced decrease of the reflection signal, the signal also slightly increased at $t \sim 0$. This effect can be most likely explained by a minor detection of the UV laser light by the photodiode and, further, an interaction of the electromagnetic field of the KrF excimer laser with the photo diode and the coaxial cable cannot be completely excluded.

The absolute value of the slope of the optical signals decreased at increasing time and finally both signals converged to an almost constant value. The time distance between the beginning of the UV laser pulse and the time position of the convergence is called Δt_{LF} (see Fig. 2). However, the defined and repeatable determination of the convergence point is very difficult due to the signal noise and a low frequency modulation of the signal, e.g. based on a small movement of the sample surface relative to the red laser beam. Therefore the signals were plotted by a linear function (see thin black and grey line in Fig. 2) at a higher time values. The experimentally used convergence point is defined by the point where the signal presented a divergence from the linear plot (see Fig. 2).

At $\Phi \sim 930$ mJ/cm^2, for the irradiation of a thin metal layer $d = 20$ nm (see Fig. 2), the main variation of the optical signal can be found at the first pulse. For a thicker layer $d = 50$ nm (see Fig. 3(b) black square) the distinct variation of the transmission signal can be detected at a higher number of laser pulses.

Besides the variation in the optical signal, a distinct modification of the surface morphology induced by the laser irradiation was detected. In Fig. 3(a) the shadow graph images dependent on the number of laser pulses N for a 50 nm thick chromium film irradiated with ~930 mJ/cm^2 are shown. All images were observed 20 µs after the laser pulse. The irradiation induces the formation of randomly distributed holes in the metal layer. This process starts with the first