Thin Solid Films 571 (2014) 102–107

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Impact of capillarity forces on the steady-state self-organization in the thin chromium film on glass under laser irradiation

Mindaugas Gedvilas *, Bogdan Voisiat, Kęstutis Regelskis, Gediminas Račiukaitis

Center for Physical Sciences and Technology, Savanoriu Ave. 231, LT-02300 Vilnius, Lithuania

ARTICLE INFO

Article history:
Received 18 February 2014
Received in revised form 25 August 2014
Accepted 24 September 2014
Available online 7 October 2014

Keywords:
Laser
Self-organization
Capillarity convection
Thermo-capillarity
Marangoni
Chromium thin film
Curvature radius

ABSTRACT

Metal films on transparent substrates are widely used for mask production in lithography, and lasers are frequently applied for their patterning. Steady self-organization of a chromium thin film on the glass substrate to parallel metal lines under irradiation with partially overlapping highly astigmatic nanosecond laser pulses above the ablation threshold has been observed. Transformations in a chromium film were investigated experimentally and numerically. The theoretical model of the steady self-organization is presented and discussed. It was demonstrated that the capillarity convection force was responsible for the transformation process in the molten metal. It was shown that the thermo-capillarity (Marangoni) shear stress and the stress originating from a variation in the radius of curvature along the structure were equally important in the case of the steady self-organization process.

1. Introduction

Metal films on glass are widely used in industry for diverse applications. A chromium film on glass is an important material for photomask production in lithography [1,2], as well as for the production of diffraction gratings [3], and linear optical encoders for metrology [4,5]. Lasers are frequently applied to pattern the metal film instead of the wet-chemical or plasma etching [6,7]. When performing micromachining using the lasers, bursts of low-energy pulses are employed to minimize heating of the film. However, nanoparticle arrays [8], ripples [9] and the laser-induced periodic surface structures, are observed at intensities near the ablation threshold [10,11]. Such transformation of the metal film under the laser beam irradiation into a regular structure is to some extent an undesirable effect but sometimes it is a promising method for micro- and nano-structuring of the functional surfaces [12].

In our previous research, the beam of a nanosecond laser tightly focused to a line was applied for the back-side ablation of the thin chromium film on the glass substrate [13–15]. The stripe-like area ablated with a single laser pulse had cylindrical ridges of the melted metal. While, the partially overlapping laser pulses transformed chromium film into parallel, quasi-parallel or irregular metal lines, depending on the processing parameters. Regular structures of the resolidified chromium metal were formed when laser fluence was above the single-pulse removal threshold and the pitch between pulses was less than the half width of the stripe ablated with a single laser pulse. The regular self-organized metal lines were located periodically with the period of 2.5–4 μm. Chromium lines were orientated perpendicularly to the long axis of the beam spot and their length increased with every shifted pulse. Formation of regular diffraction gratings was experimentally implemented by using the above-mentioned technique [16]. The Rayleigh–Plateau instability was introduced as an initiator of the self-organization of a flat metal film into parallel metal lines [17–19]. The physical phenomenon responsible for the steady continuation of the self-organization of the chromium thin film on the glass substrate into the regular structures was still unknown.

In this work, the theoretical model of the steady continuation of the self-organization is presented. It is demonstrated that the capillarity convection force is responsible for the steady continuation of the self-organized metal lines. It was shown that the thermo-capillarity (Marangoni) shear stress and the stress originating from a variation of the curvature radius along the ridge structure were equally important in the case of the steady self-organization process.

2. Experimental setup and procedures

Experiments on the laser ablation were performed by using the diode pumped nanosecond Nd:YAG laser NL202 (Ekspla Ltd.) with the Gaussian intensity profile. The acylindrical lens specially designed for the tight aberration-free focusing through the glass substrate to the backside of the chromium thin film was used in the experiments (Fig. 1). The astigmatic laser spot at the focal position was a high

* Corresponding author.
E-mail address: mgedvilas@ftmc.lt (M. Gedvilas).

http://dx.doi.org/10.1016/j.tsf.2014.09.047
0040-6090/

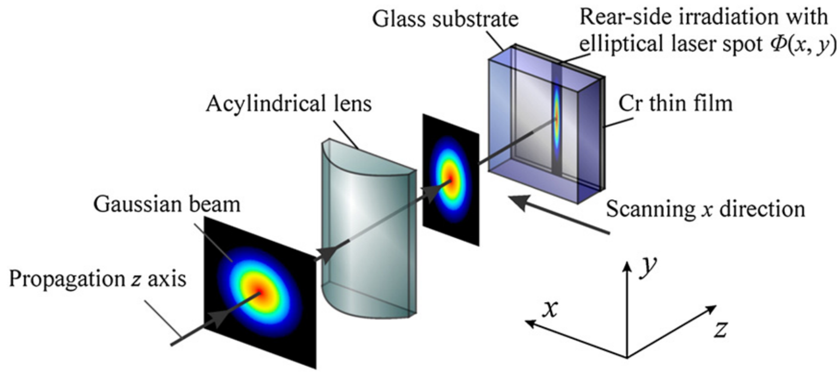

Fig. 1. Principal scheme of the experimental setup.

aspect-ratio stripe with the dimensions of $w_{x0} = 2.5$ μm by $w_{y0} = 2.5$ mm. The spatial energy density distribution in the spot is given by:

$$\Phi(x, y) = \Phi_0 \exp\left[-2x^2/w_{x0}^2 - 2y^2/w_{y0}^2\right], \quad (1)$$

where x and y are the spatial coordinates, $\Phi_0 = 2E_p/(\pi w_{x0} w_{y0})$ is the peak laser fluence at the center of the beam, and E_p is the laser pulse energy.

The laser generated the radiation with the wavelength of $\lambda = 1064$ nm, and the pulse duration full width at half maximum (FWHM) was $\tau_p = 10$ ns. The pulse energy was up to $E_p = 2$ mJ. The laser was operating at the pulse repetition rate of $f_{Rep} = 1$ kHz in the regime of the position-synchronized output, controlled by the positioning system. A detailed description of the experimental setup can be found in [13].

The chromium thin film was deposited by vacuum evaporation on the float glass substrate. The thickness h of the films used in experiments was 100 nm. The thickness of the glass substrate was 4.8 mm.

Samples were placed on the high-precision stage ALS25020 (Aerotech) and were irradiated with a sequence of partially overlapping laser pulses (Fig. 2). The distance Δx between the overlapping laser pulses (pitch) was precisely controlled by the motion controller and a computer. Morphological investigations of the structures were performed by using CPII (Veeco) atomic force microscope (AFM). AFM was used in contact mode by using a standard cantilever with the tip radius of curvature of 60 nm.

3. Numerical simulations

The aim of the modeling was to investigate the temperature distribution in the self-organized structures at the location and time of the last laser pulse in the sequence. The goal was to determine the temperature gradients in the heated structure and to estimate the time span while the chromium metal stays in a liquid state. The modeling was performed by using COMSOL Multiphysics software.

4. Background: capillarity convection

4.1. Thermo-capillarity (Marangoni) convection in liquid metal

Surface tension γ is a thermodynamic property of the liquid which depends on the temperature and other parameters such as chemical composition and surface cleanliness [20]. If the temperature difference is small, the temperature dependence of $\gamma(T)$ can be linearized in such a way that $d\gamma/dT$ is a constant. It is usually a negative value for liquid metals [21]. When the temperature varies substantially along the free surface, the gradient in the surface tension $d\gamma/dx$ results in a shear force, which causes fluid to move from the hot region to the cold region [22,23]. This phenomenon is called thermo-capillary or Marangoni convection [24]. The velocity of a liquid metal can be calculated using the following equation [25]:

$$u_{Ma} = \frac{1}{\eta}\frac{d\gamma}{dT}\frac{dT}{dx}h, \quad (2)$$

where η is the dynamic viscosity of a fluid, h is the thickness of a liquid metal and dT/dx is the temperature gradient. In thin films irradiated with the Gaussian beam, the Marangoni flow results in the formation of dry areas. The surface tension gradient due to the non-uniform heating induces a flow of the molten liquid metal away from the center of the irradiated area, leading to the formation of dry areas on the substrate [26,27].

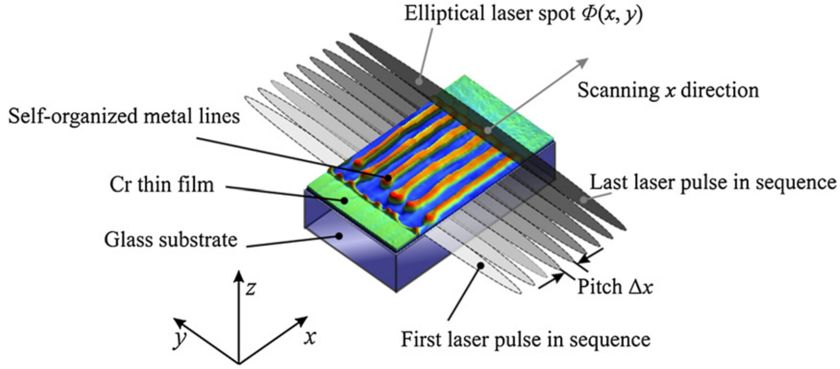

Fig. 2. Representation of the experimental procedure. Cr thin film is irradiated with the sequence of laser pulses and transformed to self-organized metal lines.