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Fabrication of sub-micron surface structures on copper, stainless steel and titanium using picosecond laser interference patterning

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ABSTRACT

Picosecond direct laser interference patterning (ps-DLIP) is investigated theoretically and experimentally for the bulk metals copper, stainless steel and titanium. While surface texturing with nanosecond pulses is limited to feature sizes in the micrometer range, utilizing picosecond pulses can lead to sub-micrometer structures. The modelling and simulation of ps-DLIP are based on the two-temperature model and were carried out for a pulse duration of 35 ps at 515 nm wavelength and a laser fluence of 0.1 J/cm^2. The subsurface temperature distribution of both electrons and phonons was computed for periodic line-like structures with a pitch of 0.8 μm. The increase in temperature rises for a lower absorption coefficient and a higher thermal conductivity. The distance, at which the maximum subsurface temperature occurs, increases for a small absorption coefficient. High absorption and low thermal conductivity minimize internal heating and give rise to a pronounced surface micro topography with pitches smaller than 1 μm. In order to confirm the computed results, periodic line-like surface structures were produced using two interfering beams of a Yb:YAG-Laser with 515 nm wavelength and a pulse duration of 35 ps. It was possible to obtain a pitch of 0.7 μm on the metallic surfaces.

1. Introduction

Surface engineering is considered as an essential technology for many key industry sectors, e.g. automotive industry, "green" energy, life science, etc. It is also connected to a variety of social themes of the 21st century like climate, energy, mobility and healthcare. In order to meet the technical requirements to succeed in a specific market, it is necessary to develop and establish new manufacturing processes to produce smaller geometric structures on large areas.

Grooves, dimples and pillar-like structures can be generated with different technologies. Besides lithographic methods such as photolithography [1], interference lithography [2,3], nanoimprint lithography [4,5] and deep X-ray lithography [6], short (nanosecond and sub-nanosecond) and ultrashort pulsed (pico- and femtosecond) laser-based techniques have been established to fabricate well-defined surface micro pattern over the last decades [7–10]. The effect of such micro structures improves, for example, the anti-adhesive properties of cutting tools [11], reduces friction of automotive components [12,13], generates hydrophobic or superhydrophobic surfaces [14–16], enhances cell proliferation on biomaterials [17–19], boosts solar cell efficiency [20] and promotes adhesive bonding [21].

Current lithographic techniques require expensive equipment, use hazardous materials and are time consuming due to serial processing, while laser techniques allow in contradiction a high level of flexibility. Both nanosecond- and sub-nanosecond pulsed laser systems in the range <100 ps have provided the opportunity to trigger specific material properties by impinging an engineered surface micro topography [22–26]. Although current direct laser writing systems are generally sufficient to produce microtopographies down to 10–15 μm, they exhibit significant limitations with respect to process speed and feature sizes, especially below 5 μm. An approach to generate periodic pattern with subwavelength feature sizes is known as LIPSS – laser induced periodic surface structures. At laser pulses below 500 fs, surface ripples develop on the surface after the irradiation of solid materials like semiconductors, insulators and metals [27–30]. LIPSS methods are currently limited to small area processing due to speed limitations. Also, period and complexity of the ripples are inconsistent across the

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ablation spot. Overall, the underlying physical mechanisms are not yet fully understood.

The interference of two or more coherent laser beams can be applied to produce periodic grooves, grids or dimples with pitches (periods) in the micrometer and sub-micrometer range on a variety of materials, such as metals, polymers and thin metallic films [31–33]. This technique is called direct laser interference patterning (DLIP). For the case of bulk metals, DLIP is based on a photothermal process that involves local melting and/or selective ablation at the interference maxima. For nanosecond laser pulses, the primary material removal mechanism is ablation but substantial melting occurs for metallic materials. The surface temperature is determined by thermal conductivity, thus laser ablation is an equilibrium process which obeys Fourier's law of heat conduction [34,35]. The minimum achievable pitch is therefore limited by the thermal diffusion length, which is approximately 1 μm for stainless steel and titanium, and 2 μm for copper [36,37]. When the laser pulse duration reduces to the order of picosecond or femtosecond, little thermal damage is observed and pitches below 1 μm should be feasible. DLIP with sub-nanosecond laser pulses <100 ps offers therefore the possibility to fabricate precisely defined surface topographies with pitches in the sub-micron range. These sub-micrometer topographies can potentially be used to generate an additional specific surface function, such as antireflection or dirt-repellent effects.

In this study, DLIP is used to fabricate periodic grooves with pitches below 1 μm on the bulk metals copper, stainless steel (type: 304), and titanium (TiAl6V4). The metals were chosen as the focus of this study due to their relevance to the industry, e.g. for medical applications (Cu, Ti), the printing industry (Cu, stainless steel) and the food industry (stainless steel). The patterning is achieved using a Yb:YAG laser system, where frequency doubled laser beams at 515 nm wavelength overlap to form an interference pattern with a peak-to-valley architecture. In addition, thermal simulation by finite element method (FEM) was carried out modelling the photothermal laser interactions. The two-temperature model (TTM) is considered for the first time to represent the heat transfer under picosecond DLIP. It describes the time-dependent excitation of the electrons and the subsequent energy transfer from the electrons to the phonons (lattice) in the metallic material. The temperature profiles of both electrons and phonons of bulk copper, iron (substitute for stainless steel) and titanium irradiated by a single-shot picosecond laser pulse are determined solving the one-dimensional differential equations according to the TTM. The temperature dependence of the thermal conductivity is taken into account and examined via FEM for copper.

2. Experimental

2.1. Thermal model

When laser radiation strikes matter, the energy is partly reflected, transmitted or absorbed. In metals, the laser energy of an incident laser pulse is firstly absorbed by the electrons, leaving the phonons "cold" for an initial time period of femtoseconds [38]. The heat is subsequently transmitted by electron-phonon collisions. This energy exchange between electrons and phonons is characterized by the "relaxation time", which describes the decrease of energy upon the laser pulse until a thermal equilibrium between electrons and phonons is established, and can be described by the linear differential equations

$$c_i \frac{\partial T_i}{\partial t} = -c_e \frac{\partial T_e}{\partial t} = \gamma (T_e - T_i), \tag{1}$$

where t is the time, c_i, c_e and T_i, T_e are the specific heat capacity and temperature of the phonons and electrons, respectively. The parameter γ depends on the material, and describes the strength between the electron-phonon-coupling. Applying Fourier's law of heat conduction, two equations for the lattice and electron temperature can be derived. For the one-dimensional case (z is the direction perpendicular to the metal surface) they become [39]

$$c_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z} \kappa_e \frac{\partial T_e}{\partial z} - \gamma (T_e - T_i) + \alpha Q(z,t) \tag{2}$$

$$c_i \frac{\partial T_i}{\partial t} = \frac{\partial}{\partial z} \kappa_i \frac{\partial T_i}{\partial z} + \gamma (T_e - T_i). \tag{3}$$

The coefficients κ_e and κ_i are the thermal conductivity of the electrons and phonons, α is the absorption coefficient of the material, and the source term $Q(t, z)$ is associated to the energy of the laser pulse. Since the thermal conductivity of the electrons is considerably higher than the thermal conductivity of the photons, the heat conduction term in Eq. (3) can be neglected. While the electron heat capacity depends linearly from the electron temperature ($c_e = c_{e0} T_e$, c_{e0}: electron heat capacity at room temperature), the heat capacity of the phonons is constant for temperatures higher than the Debye temperature [40]. In most cases, the system of heat equations (Eqs. (2) and (3)) can only be solved numerically. However, there are approximations to find analytical solutions within a certain range.

The values of κ_e and α depend on the temperature. According to the Drude model, the electron thermal conductivity of metals can be written as [41]

$$\kappa_e = \frac{1}{3} \upsilon^2 c_e \tau, \tag{4}$$

where υ^2 is the mean square electron speed. Both electron–electron (e-e) τ_{ee} and electron-phonon (e-ph) scatterings τ_{ep} contribute to the total scattering time $\tau = \tau_{ee} + \tau_{ep}$, where $1/\tau_{ee} = AT_e^2$ and $1/\tau_{ep} = BT_i$ [42]. The thermal conductivity then becomes

$$\kappa_e = \frac{1}{3} v_F^2 \gamma \frac{T_e}{BT_i + AT_e^2}, \tag{5}$$

where v_F is the Fermi velocity, and both A and B material dependent constants. When T_e remains smaller than the Fermi energy E_F, Eq. (5) can be approximated by

$$\kappa_e \approx \kappa_0 \frac{T_e}{T_i}, \tag{6}$$

where κ_0 is the thermal conductivity at room temperature $T_0 = 300$ K. If T_e is larger than the Fermi energy, Eq. (6) is no longer valid, and the following equation should be used [43]:

$$\kappa_e = C \frac{\left(\vartheta^2 + 0.16\right)^{5/4} \left(\vartheta^2 + 0.44\right) \vartheta}{\left(\vartheta^2 + 0.092\right)^{1/2} \left(\vartheta^2 + b\vartheta_i\right)}, \tag{7}$$

where $\vartheta = k_B T_e / E_F$, $\vartheta_i = k_B T_i / E_F$ (k_B: Boltzmann constant), and C and b are specific material parameters.

The energy intensity distribution of a line-like interference pattern (grooves) is considered

$$\Phi(x,y) = 4\Phi_0 \cos^2\left(kx \sin\frac{\theta}{2}\right) \quad \text{at } z = 0, \tag{8}$$

where Φ_0 is the laser fluence of each beam, θ the angle of incidence between the interfering beams, and $k = 2\pi/\lambda$ is the wave number at the laser wavelength λ. The temporal intensity distribution of the laser pulse follows a Gaussian distribution

$$I(x,y,t) = \frac{\Phi(x,y)}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(t-t_0)^2}{2\sigma^2}\right) \tag{9}$$