Chapter 3 Surface Functionalization by Laser-Induced Structuring

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Abstract This chapter reviews the possibilities of functionalizing solid surfaces by laser irradiation—i.e. modifying the surfaces with respect to, e.g., wettability, optical properties, friction/wear-control, corrosion resistance, electrical properties. The functionalization occurs via the modification of surface morphology by the laser-induced formation of (regular) nano- to micro-textures. We present the different approaches to generate these patterns as well as their implication for specific functions obtained by this structuring and, finally, show some examples of applications.

3.1 Introduction

Materials surfaces play a paramount role in many aspects such as the optical appearance, mechanical properties like friction and wear, the interaction with the environment like wettability, corrosion, or adhesion. Also, chemical activity, e.g. in catalysis, is strongly dependent on the surface. The modification of solid surfaces to obtain new, functional properties is of considerable impact on the quality of modern engineering products and innovation [\[1,](#page-19-0) [2\]](#page-19-1). Such modification may be achieved, on the one hand, by coating the surface with a thin film providing the desired performance [\[3–](#page-19-2)[5\]](#page-19-3). On the other hand, there is the possibility of modifying the actual surface morphology by the formation of new surface textures with feature size ranging from several hundred nanometers to the several ten microns, sometimes combining to form complex multi-scale hierarchical structures [\[6\]](#page-19-4). The patterns and their function often have their paradigm in nature and biology, with the most popular examples of the lotus leaf and its dirt-repelling hydrophobicity or of the colorful appearance of butterfly wings [\[7\]](#page-19-5). There are several techniques to achieve such surface patterning, including a combination of successive grinding, etching and chemical coating [\[8\]](#page-19-6), or lithography [\[9\]](#page-20-0), sometimes based on a template directly taken from a real

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biological antetype [\[10\]](#page-20-1). A particular method is the replication from a mold formed by, e.g., diamond-machining [\[11\]](#page-20-2). Whereas most of these procedures employ multiple processing steps, surface nano-/micro-texturing can also be achieved by either self-organized pattern formation during thin film epitaxial growth [\[12\]](#page-20-3) or by direct exposure to energetic irradiation, namely by ion, electron, or laser beams.

For charged particles beams, there are two typical ways of structuring: (a) direct-write lithography [\[13–](#page-20-4)[16\]](#page-20-5), and (b) relaxation from thermodynamic instability after ion-beam sputtering, where a large variety of regular to complex nano-patterns are attributed to a self-organized structure formation [\[17–](#page-20-6)[19\]](#page-20-7). Surface morphology modification by intense laser pulses can be, similarly, differentiated into two main lines: (a) ablative lithography of multi-beam induced interference patterns $[20]$, and (b) single beam formation of Laser-Induced Periodic Surface Structures (LIPSS) [\[21](#page-20-9)[–28\]](#page-20-10). The latter two will be discussed in detail in the present chapter. We will start by showing some typical relations between surface textures—including laser-generated ones—and functionality. Then, we explicitly consider laser patterning and present several experimental approaches for their realization, together with current models explaining the interaction.

3.2 Functionality of Textured Surfaces

More or less regular surface patterns on the sub- to several micrometer scale result in numerous surface functionalities ranging from specific coloring over modified wettability [\[29\]](#page-20-11), such as hydro- and oleo-phobicity or -philicity, special tribologic properties, wear-resistance, catalysis, modified adhesion. They can serve as templates for biological cell growth [\[30\]](#page-20-12) and can stimulate surface enhanced Raman scattering [\[31,](#page-20-13) [32\]](#page-20-14). In the following, we present some typical surface functionalities and the patterns at their basis.

3.2.1 Wettability

One of the most important surface-morphology induced functionalities is, certainly, the modification of wettability, i.e. the controlled adjustment of liquid-repellent resp. liquid-attracting properties [\[1,](#page-19-0) [7\]](#page-19-5). The correlation is bio-inspired by the surface of the lotus leaf $[7, 24, 33]$ $[7, 24, 33]$ $[7, 24, 33]$ $[7, 24, 33]$ $[7, 24, 33]$ (Fig. [3.1\)](#page-2-0) or the rose petal $[10]$.

Wettability, i.e. the capability of a solid to attract and spread or to repel a liquid on its surface, is of great importance both in daily life and, particularly, in technology. High wettability—for water called "hydrophilicity"—is required if a surface is to be covered by a thin liquid film, e.g. a lubricant or wet paint, to obtain an all-over coverage. Also the uniform spread of oil or water on a warm Teflon pan is due to this quality [\[34\]](#page-21-1), despite its "cleaning effect" (the resulting wetting film provides a smooth, separating interface between the pan and the contents). The opposite

Fig. 3.1 Super-hydrophobic lotus leaf (from [\[33\]](#page-21-0)). Left: photograph; right: SEM image of the lotus leaf surface

behavior of liquid repelling—for water called "hydrophobicity"—is important for effects like self-cleaning, e.g. of windshields, anti-icing of airplane wings, reduced corrosion, bacterial resistance.

The phenomenon of wettability is determined by the mutual interaction of the three phases: solid, liquid, and gaseous. It is, mainly, determined by attractive forces inside the liquid (cohesion) compared to interface forces between solid-liquid (adhesion) and liquid-gas. Typically, the latter are significantly weaker than the liquid internal binding, resulting in the confinement of a free liquid droplet because work against the internal binding is needed to increase the droplet surface. This effect is called surface tension; it controls the shape and size of a free liquid droplet, as described already in 1805 by Young [\[35\]](#page-21-2). At the droplet-surface interface, the competition of adhesive forces between the surface and the liquid, one the one hand, with cohesive forces within the liquid, on the other hand, is responsible for a minimization of the contact area. The shape of the droplet at the triple boundary (solid, liquid, gas) is then determined by a minimization of the total interface energy, γ [\[36\]](#page-21-3), combining surface tension (liquid–gas, LG), droplet-surface adhesion (SL) and gas-surface adhesion (SG). It is characterized by the contact angle, θ , which is >90° for liquid repulsion (hydrophobicity) and <90° for attraction (hydrophilicity). At an *ideal* surface the contact angle is determined by Young's Equation:

$$
\cos \theta = \frac{\gamma_{SG} - \gamma_{SL}}{\gamma_{LG}} \tag{1}
$$

This is, schematically, shown on the left panel in Fig. [3.2.](#page-3-0) For a *real* surface, however, we have to account for surface roughness, modifying the surface-liquid contact by, e.g., either increasing or reducing the efficient contact area, thus modifying the contact angle. These two possibilities are depicted on the center and right panels in Fig. [3.2.](#page-3-0)

It becomes immediately evident that the specific wettability is strongly influenced by the morphological structure of the surface roughness. Another important influence is due to the surface active area together with the surface charge distribution.

Fig. 3.2 Wetting states: Classical (Young) model for ideal surface (left); rough surface: Wenzel model, Cassie-Baxter model (super-hydrophobic). The wetting state is characterized by the contact angle θ

Fig. 3.3 Super-hydrophobic laser-processed steel surface

Fig. 3.4 Principle of sessile drop method for measuring contact angles: the liquid droplet sitting on the solid surface is homogenously illuminated from the back; the resulting shadowgraph is recorded by a camera

Experimentally, super-hydrophobicity, similar to the lotus leaf, can be achieved by hierarchical (multi-scale) laser-modification of stainless steel [\[37\]](#page-21-4) (cf. Fig. [3.3\)](#page-3-1).

The contact angle can be, usually, measured optically by the sessile drop method [\[38\]](#page-21-5) as shown in Fig. [3.4.](#page-3-2)

A particularly sensitive variation of the method consists in measuring the dynamic contact angle by changing the droplet volume. Then, a hysteresis of two different contact angles is obtained: advancing by increasing the droplet volume and receding by reducing the droplet volume [\[39\]](#page-21-6). Generally, the receding contact angle is smaller and the advancing contact angle is larger than or equal to the static one.

3.2.2 Color

Similar to the feature of wettability, also the connection between surface micro- /nanostructure and color is bio-inspired. The most familiar examples are the multicolored wings of butterflies [\[7,](#page-19-5) [40\]](#page-21-7) (Fig. [3.5\)](#page-4-0).

The phenomenon is based on the dispersion of white light transmitted (cf. Fig. [3.6\)](#page-5-0) or reflected by optical gratings: due to interference, light incident on a grating of periodicity (line spacing) *d* is wavelength-dispersed and the fraction of wavelength λ continues to propagate under diffraction angle α for the first diffraction order [\(2\)](#page-4-1):

$$
\sin a = \lambda/d \tag{2}
$$

For given angles of illumination and observation, thus, different values of *d* result in different colors observed. The effective values of *d* do not only depend on the physical spacing but also on the orientation of the grating with respect to the plane of incidence (Fig. [3.7a](#page-5-1)) which is clearly obvious by the multi-color directional reflection from a compact disk with its circular grating pattern (Fig. [3.7b](#page-5-1)).

Fig. 3.5 Butterfly [Robert Clark, *National Geographic*] and SEM pictures of microstructures on a butterfly wing [\[40\]](#page-21-7)

Fig. 3.6 Dispersion of white light by an optical grating. **a** Schematic (transmission); **b** Principle: incident white light (wave fronts and direction of propagation are indicated) is split and different wavelengths are diffracted into different directions; **c** Detail: for a line spacing *d*, light of wavelength λ is diffracted under angle α

Fig. 3.7 Diffraction from identical but differently oriented—with respect to the plane of incidence—gratings. **a** Schematic; **b** Dependence of the effective grating spacing d' on grating orientation; **c** Diffraction from a compact disk (CD) with annular gratings

Indeed, such multi-color diffraction effect has been produced also by Laserinduced periodic structures (LIPSS) by Dusser et al. [\[41\]](#page-21-8) (Fig. [3.8\)](#page-6-0), where they could show, in addition, that the effect can be used for hidden marking.

A particular type of structure-induced "coloring" is the formation of deep black surfaces, such as carbon black [\[42,](#page-21-9) [43\]](#page-21-10), or highly absorbing surfaces for applications in solar cells [\[44,](#page-21-11) [45\]](#page-21-12). The micro-/nano-structures responsible for that high absorption act two-fold: one the one hand, they simply increase the active surface considerably. But more important, on the other hand, is the multiple reflection and final trapping 3 Surface Functionalization by Laser-Induced Structuring 69

Fig. 3.8 Multi-color diffraction from a laser-structured surface with different LIPSS directions (from [\[41\]](#page-21-8))

Fig. 3.9 Light-trapping by complex structures. **a** Wood's Horn; **b** "Black Chrome" (from [\[44\]](#page-21-11))

inside the micro-/nano-structures like in aWood's Horn (Fig. [3.9a](#page-6-1)). Typical structures consist of multiple pores [\[45\]](#page-21-12) or multi-scale arrays of deep grooves or notches.

The creation of structural colors, notably black surfaces, by LIPSS has also been successfully reported [\[32\]](#page-20-14). As a particular application-related variant, "Black Silicon" has been produced, amongst others, by Eric Mazur's group [\[46\]](#page-21-13).

3.2.3 Field Enhancement

Whereas, so far, surface functionality mostly because of depth modulation (depression) was considered, also sharp protrusions can have an important effect. In the presence of electric fields (static or electromagnetic) such tips or tip-arrays cause

Fig. 3.10 Nanopillar arrays. **a** Ag-pillars on top of porous anodic alumina; **b** detail of the pillars in (**a**) (from [\[51\]](#page-21-14)); **c** spikes on Si (100), produced by femtosecond LIPSS (from [\[53\]](#page-21-15))

a considerable local field enhancement [\[47,](#page-21-16) [48\]](#page-21-17). This can be exploited as a very efficient, brilliant source of fast electron bunches [\[48,](#page-21-17) [49\]](#page-21-18), e.g. for applications in ultrafast electron diffraction, crystallography, and microscopy [\[50\]](#page-21-19). The strong field enhancement in a tip-array can also increase the efficiency of surface enhanced Raman scattering (SERS) considerably [\[31,](#page-20-13) [51,](#page-21-14) [52\]](#page-21-20). Again, such tip-arrays can be easily fabricated also by LIPSS [\[53\]](#page-21-15) (Fig. [3.10\)](#page-7-0).

3.2.4 Templates for Biological and Technological Films

The main biological functionality of nanostructured surfaces is related to the formation of biologically active films $[30, 54, 55]$ $[30, 54, 55]$ $[30, 54, 55]$ $[30, 54, 55]$ $[30, 54, 55]$ (Fig. [3.11\)](#page-8-0)—or, contrarily, the suppression of their growth to avoid fouling [\[56\]](#page-22-1); the nanostructures can even serve to make titanium implants antibiotic [\[57\]](#page-22-2).

An example for the formation of active films is the exploitation of ZnO nanostructures on silicon substrate as templates for the development of topography-mediated neuronal cultures [\[58\]](#page-22-3). It was demonstrated that the ZnO-templates can support neuronal cell growth and proliferation. Another effect is the stabilization and immobilization of enzymes on nano-structured surfaces [\[59,](#page-22-4) [60\]](#page-22-5). Natural nanostructures, such as diatoms with their outer shell of micro- and nano-porous silica, have been proposed as templates for bio/chemical sensors and biomimetic membranes [\[61\]](#page-22-6). Again, surface structures as templates for biofilms have also been produced by laser induced periodic surface structuring [\[62\]](#page-22-7).

Not only for bio-films, also in Materials Science, can surface textures be used as templates to increase the functionality of films grown on these surfaces. As an example, epitaxially grown self-organized semiconductor nanostructures can serve as templates for very regular nanomagnet arrays [\[12\]](#page-20-3).

Fig. 3.11 Comparison of P. aeruginosa adhesion on structured and unstructured regions. **a** Fluorescence microscopy shows the different localized adhesion on flat region (upper) and structured region (lower), with an abrupt transition. **b** and **c** are corresponding cross-sectional SEM images, showing the strong difference in attachment morphology. The aligned cells in (**c**) are false-colored to highlight their orientation. Scale bars are $10 \mu m$ in (**a**) and $1 \mu m$ in (**b**) and (**c**). (From [\[30\]](#page-20-12))

3.3 Laser Patterning

In the following, we present the main methods to use laser techniques for regular surface texturing. In one way or the other, they are all closely related to the field of laser ablation: the light of, usually, strong laser pulses is absorbed by the target material and the resulting material response ranges from (surface) melting to massive material removal. In general, one can conceive two scenarios of responsible processes for pattern formation:

- (1) Local ablation after a patterned irradiation, very similar to lithography, using electron or ion beams $[13-16]$ $[13-16]$, or local direct writing; one typical example is mask projection of excimer laser irradiation [\[63\]](#page-22-8) (a; cf. Fig. [3.12\)](#page-9-0), another approach is generating the pattern by multi-beam interference (b; Sect. [3.1\)](#page-0-0).
- (2) Surface structuring by irradiation with a single beam (laser-induced periodic surface structures; LIPSS) [\[21](#page-20-9)[–27\]](#page-20-16).

3.3.1 Multi-beam Interference and Ablation

The most obvious way to generate mask-less patterned irradiation, as required for scenario (1b), is the illumination by interference of two or more beams, hitting the sample from different directions [\[20,](#page-20-8) [64–](#page-22-9)[66\]](#page-22-10) (Fig. [3.13\)](#page-9-1).

Fig. 3.12 Patterned 90 nm thick SiOx film; laser parameters: 248 nm, 1 pulse; structure period: 4.7 µm (from [\[63\]](#page-22-8)). Fluence: **a** 180 mJ/cm2, **b** 190 mJ/cm2, **c** 200 mJ/cm2, **d** 220 mJ/cm²

A typical example of such multi-beam-interference patterning is shown in Fig. [3.14](#page-10-0) [\[66,](#page-22-10) [67\]](#page-22-11): an array of several parallel coherent beams is derived from a single laser in a diffractive optical element (DOE) and, then, focused to interfering overlap at the sample.

Fig. 3.14 Multi-beam-interference patterning. The left panel presents the principal set-up with the incident laser beam split into, here, six parallel beams by a combination of DOE and imaging lens. After spatial filtering, the beams are refocused to overlap on the sample. The right panels show micrographs of three typical patterns, obtained by interference of two, three, and six beams, respectively (from [\[67\]](#page-22-11))

3.3.2 Single-Beam Laser Induced Periodic Surface Structures (LIPSS)

The phenomenon of surface morphology modification by incidence of a single laser beam was first observed already in 1965, shortly after the realization of the first laser, by Birnbaum [\[68\]](#page-22-12). In the beginning, this "surface damage" was mostly considered as an unwanted, though inevitable, trouble in materials processing as well as for optical components used with high power lasers [\[69,](#page-22-13) [70\]](#page-22-14). It was not before two decades later, that the phenomenon was studied in its own right $[21, 71–77]$ $[21, 71–77]$ $[21, 71–77]$ $[21, 71–77]$. The next progress was the formation of sub-wavelength structures induced by ultrafast lasers [\[27,](#page-20-16) [78\]](#page-23-1), again almost two decades later, initiating still ongoing research. This time, the emphasis is no longer on the damage aspect but, instead, growing interest is focused on possible, positive applications of the generated surface patterns, as indicated above in Sect. [3.2.](#page-1-0)

Nevertheless, the first experiments of femtosecond-LIPSS were concentrated on the effect as such. The first experiments investigated the surface morphology produced on a single interaction spot with diameters ranging from a few to more than $100 \mu m$ (cf. Fig. 3.15).

The effect is observed on a wide range of solid state materials [\[22\]](#page-20-17), such as dielectric insulators [\[27\]](#page-20-16), semiconductors [\[79\]](#page-23-2), metals [\[80\]](#page-23-3), and polymers [\[77,](#page-23-0) [81\]](#page-23-4). The irradiation is always close to the macroscopic ablation threshold, and the surfacemorphology modification appeared as well after very few pulses [\[82,](#page-23-5) [83\]](#page-23-6) (or even a single pulse [\[84–](#page-23-7)[86\]](#page-23-8)) as only after many thousand pulses [\[27\]](#page-20-16).

Fig. 3.15 Typical LIPSS pattern at the bottom of a 100-µm spot on a BaF2 crystal surface, irradiated by 43,000 pulses from an amplified Ti:Sapphire Laser (800 nm wavelength; 100 fs pulse duration; 1 J/cm² fluence {90% of ablation threshold }; polarization vertical). Note that two periodic structures of different periodicity Λ (one of the order of the laser wavelength λ , the other one of about λ /10) are superimposed perpendicularly. The horizontal and vertical AFM traces, recorded in the indicated stripes, show the periodicities $\Lambda_1 \approx 690$ nm, $\Lambda_2 \approx 280$ nm. (From [\[27\]](#page-20-16))

All experiments show that the LIPSS orientation is strongly determined by the polarization of the laser beam (Fig. [3.16a](#page-12-0)–e), with a few exceptions where surface scratches or similar irregularities can override the polarization dependence (Fig. [3.16f](#page-12-0), g).

The laser-generated morphology can exhibit a variety of different appearances:

- (1) Very narrow, regular lines (for linear polarization) or spherical dots for circular polarization; cf. Fig. [3.16\)](#page-12-0) with a feature size in the 100-nm range, well below the diffraction limit;
- (2) Wider periodic, parallel lines with a spacing close to the laser wavelength;
- (3) An array of larger irregular slabs of several-µm size, separated by deep trenches (similar to a dried-out river bed);
- (4) A regular array of conical tips, with spacing and size in the μ m-range.

For convenience, the different features have been termed [\[22,](#page-20-17) [79\]](#page-23-2) "HSFL" (high spatial frequency LIPSS) resp. "LSFL" (low spatial frequency LIPSS) for types 1 resp. 2 (cf. Fig. 3.15); "groves" (type 3), and "spikes" (type 4) (cf. Fig. 3.17). In general, HSFL and LSFL are oriented perpendicular to each other, a typical example is shown in Fig. [3.15.](#page-11-0)

An crucial parameter, affecting feature type and size appears to be the total dose of absorbed energy, i.e. the laser fluence and the number of superimposed laser shots

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Fig. 3.16 Polarization dependence of LIPSS orientation. The respective polarization, circular in **a** [\[87\]](#page-23-9) and linear in **b**–**d** [\[26\]](#page-20-18) is indicated by the white arrows. **e** shows the effect of elliptical polarization [\[88\]](#page-23-10). In **f**, **g**, the influence of macroscopic scratches on the surface is shown, overriding the polarization dependence [\[89\]](#page-23-11)

Fig. 3.17 LIPSS on silicon. The three areas on the left [\[90\]](#page-23-12), showing LSFL and grooves, are from one single spot with Gaussian profile from the edge (left) to the center; the right panel shows spikes [\[91\]](#page-23-13). The scale is the same on all panels

[\[91–](#page-23-13)[93\]](#page-23-14). This can be seen in Fig. [3.17](#page-12-1) where the increasing intensity within the Gaussian spot profile results in a transition from LSFL to grooves

An important step towards an application of LIPSS structures was achieved by the discovery that it is possible to coherently modify larger areas by scanning the laser across the sample surface [\[94](#page-23-15)[–97\]](#page-23-16) (Fig. [3.18\)](#page-13-0).

In general, there are two scanning methods as shown in Fig. [3.19:](#page-13-1) (a) Keeping the target fixed in position and scanning the laser spot by means of a pair of orthogonally

Fig. 3.18 Coherently continued ripples by scanning parallel to the polarization on fused silica. Left: single track: right: coherently continued ripples in 2D by scanning several tracks with an offset of 400 nm (one spot diameter) (from [\[96\]](#page-23-17))

Fig. 3.19 Schematic of two methods for large area scanning. **a** Galvo scanning head and fixed sample, **b** fixed spot and mechanically scanning sample

moving mirrors ("scanning head"); the moving beam is, then, focused onto the target by an F-theta (telecentric) lens. (b) Keeping the laser spot fixed and moving the target, mounted on a set of precision translation stages.

Both techniques bear their individual advantages: procedure (a) allows a very high scanning speed, usually controlling the mirror motion by a galvanometric drive (galvoscanners); further, it allows a very compact and rigid set-up when using a commercial scanning head. Method (b) allows very large target areas to be covered, depending only on the translation stage size and precision; however, it is, generally, much slower and may be less compact; therefore, it is, mostly, used for laboratory application.

Typically, the full surface is covered by first writing lines along one direction (e.g. "*X*") followed by adjacent tracks, displaced in the other direction (e.g. "*Y*"). Then, there are two further options: writing the lines either along or perpendicular to the laser polarization. Generally, it turns out that scanning normal to the ripples direction, i.e., usually, along the polarization, yields more regular patterns than the other sense. Another important parameter for the pattern quality is the scanning speed, or more precisely its ratio to the repetition rate, yielding longitudinal pulse overlap and, thus, the effective number of pulses acting on one spot area. A similar role is played by the scanning pitch, i.e. the separation of adjacent tracks (or lateral pulse overlap). The importance of these parameters becomes evident when considering the dose-dependence of the generated patterns (cf. Figs. [3.15](#page-11-0) and [3.17\)](#page-12-1).

3.3.2.1 LIPSS and Functionality

As was already addressed in Sect. [3.2,](#page-1-0) most of the functional nano-structuring can be, easily, achieved by LIPSS formation. Here, we just want to compile, again, the relevant features. It should be noted that, since LIPSS functionality only involves the surface of the modified material, usually very short laser pulses in the femtoor picosecond range are used to avoid thermal effects in the target volume [\[98\]](#page-23-18). A comprehensive review on LIPSS-functionalized surfaces can be found in [\[22–](#page-20-17)[24,](#page-20-15) [32\]](#page-20-14).

A Wettability

The modification of surface wettability by LIPSS was first reported in 2006 by the FORTH group on silicon [\[99\]](#page-24-0) and by Groenendijk and Meijer [\[100\]](#page-24-1), who patterned a stainless steel surface to become super-hydrophobic (cf. Fig. [3.20\)](#page-15-0). Such surface could, subsequently, be used as a mould for plastic replicas exhibiting similar hydrophobicity.

There have been many approaches since then, showing, e.g., an improvement of hydrophobicity by chemical alkysilane post-treatment [\[102\]](#page-24-2) or the formation of hierarchical multiscale patterns [\[103\]](#page-24-3). Again, it appears that the irradiation dose plays an important role for controlling the functionality [\[37,](#page-21-4) [99\]](#page-24-0). In fact, is not only possible to make the surface (super-) hydrophobic but also hydrophilicity (superwetting) can be achieved [\[23,](#page-20-19) [32,](#page-20-14) [37\]](#page-21-4), even obtaining a surface where a water film creeps upwards against gravity [\[104,](#page-24-4) [105\]](#page-24-5) (Fig. [3.21\)](#page-16-0).

A peculiar effect, so far not fully understood, is the "aging" of processed metal surface [\[93\]](#page-23-14): even a multiscale-structured steel surface is not immediately superhydrophobic but only develops this property after longer dwelling (from hours to days) in ambient atmosphere [\[37,](#page-21-4) [93,](#page-23-14) [103\]](#page-24-3). The most probable cause is some uncontrolled chemical modification, e.g. oxidation.

B Tribology

Closely related to the topic of modified wettability is the influence of LIPSS on the tribological properties of the treated surface. The underlying idea is two-fold: on the one hand, the depressions and grooves may serve to provide a reservoir for a possible lubricant; on the other hand, the reduced surface area, consistent of smooth, narrow ridges or tips, may reduce the resulting interaction between the surface and another body, rolling or sliding on that surface. A control of friction can have important implications: for a friction clutch in power transmission between a driving and a driven shaft it should be maximized; on the other hand, for wear reduction between moving surfaces, it should be reduced.

Consequently, the first proposal for an application of LIPSS intended to reduce friction and wear in a system of rapidly rotating hard disk and sliding head [\[106\]](#page-24-6). Following experiments on diamond like carbon (DLC) [\[107,](#page-24-7) [108\]](#page-24-8), using femtosecond LIPSS, showed a reduction of both adhesive and lateral force (Fig. [3.22\)](#page-16-1).

Chen et al. succeeded in reducing the friction coefficient of a SiC seal in water lubricant by about 20% [\[109\]](#page-24-9) (Fig. [3.23\)](#page-16-2).

Significant wear reduction on silicon [\[110\]](#page-24-10) and on PEEK surface, a biocompatible thermoplastic for implanted joints, have been reported [\[111,](#page-24-11) [112\]](#page-24-12). Recently, Bonse et al. [\[113,](#page-24-13) [114\]](#page-24-14) demonstrated that not only the LIPSS texture but also the contacting materials as well as possible lubricants play an important role.

Fig. 3.21 Hydrophilicity: water droplet spreading up-hill (from [\[105\]](#page-24-5))

Fig. 3.22 Tribology: influence of LIPSS roughness on contact area (from [\[107\]](#page-24-7))

Fig. 3.23 LIPSS-reduction of friction coefficient of SiC in water lubricant (right). For comparison, the coefficient of the non-textured surface (left) is indicated by the dotted line (from [\[109\]](#page-24-9))

C Color

Already in Sect. [3.2.2,](#page-4-2) examples are given for selective surface coloring by LIPSS formation (cf. Fig. [3.8;](#page-6-0) [\[41\]](#page-21-8)). An impressive overview is presented in [\[32\]](#page-20-14). Particular interest appertains to the formation of black silicon, notably in order to optimize spectral absorption for photovoltaic applications. However, this has to be considered with care, since the structural surface modification is associated with the formation of electronic defects, significantly reducing the carrier lifetime [\[115\]](#page-24-16). Eric Mazur's group succeeded in overcoming this problem by conducting the laser processing under $SF₆$ atmosphere [\[53\]](#page-21-15) and subsequent annealing at 1200 K to remove hierarchical secondary structures [\[46\]](#page-21-13) (Fig. [3.24\)](#page-17-0).

Fig. 3.24 "Black silicon", formed by laser processing (from [\[46\]](#page-21-13))

D Field Enhancement/Surface Enhanced RAMAN Scattering

As indicated before, at sufficiently high irradiation dose the regular line-shaped LIPSS convert into an array of regular, sharp tips, capable to a considerable local electrical field enhancement. This can be exploited by using the array as a template for surface enhanced RAMAN scattering (SERS) [\[116–](#page-24-17)[118\]](#page-24-18) (Fig. [3.25\)](#page-17-1).

E Templates for Biological and Technological Films

As generally holds for nanostructured surfaces (cf. Sect. [3.2.4\)](#page-7-1), an interesting application of LIPSS patterned surface is the possibility of growing on top biological [\[119\]](#page-24-19) or technological films [\[120\]](#page-25-0) with peculiar features.

In the fields of medical and biological functionality, the main application concerns cell adhesion [\[121,](#page-25-1) [122\]](#page-25-2). There can be either a positive or, as well, a negative effect [\[62\]](#page-22-7), depending on substrate and on biomaterial. A typical application concerns the optimization of titanium implants [\[82\]](#page-23-5).

Another aspect applies to the LIPSS-template induced self-organized growth of ordered structures, both for biomaterials, e.g. directional cell spreading on titanium [\[123\]](#page-25-3), and for technological materials, e.g. vertically aligned carbon nanofiber growth [\[120\]](#page-25-0).

3.3.2.2 Lithographic Model: Laser-Plasmon Interference

The first approach to understand LIPSS formation was triggered by the apparent grating-like structures with a feature size close to the laser wavelength [\[21,](#page-20-9) [68](#page-22-12)[–76\]](#page-22-16): the pattern was assumed to result from modulated ablation [\[68\]](#page-22-12) following an illumination interference pattern between the incident wave and a surface-scattered wave [\[69,](#page-22-13) [72](#page-22-17)[–74\]](#page-22-18). Keeping the basic idea of modulated ablation due to inhomogeneous energy deposition, the model was refined by ascribing the illumination pattern to interference between the incident wave and surface plasmon-polaritons, excited by the laser field. In order to apply this model also to non-conducting targets, a laserinduced transient high conduction-electron density was considered in a Drude-model for quasi-free electrons [\[124\]](#page-25-4). For more details see [\[22\]](#page-20-17). Though this model is very successful to predict all details of periodic surface structures of the LSFL type, its validity for other structures like HSFL, grooves, spikes is only very limited. The model is practically purely based on an assumption of particular electromagnetic field distributions occurring in the interaction, an active role of the material in structure formation is neglected, so far.

3.3.2.3 Surface Dynamic Model: Self-organization and Hydrodynamics

An alternative picture of the basic LIPSS formation mechanisms, developed by our group [\[26\]](#page-20-18) has been triggered by the apparent similarity of LIPSS with patterns produced by self-organized epitaxy [\[12\]](#page-20-3) or ion beam sputtering [\[19\]](#page-20-7). Our model is closely related to the concepts developed for the latter [\[17,](#page-20-6) [18\]](#page-20-20) and assumes a much more active role of the material than the lithographic model (Sect. [3.3.2.2\)](#page-18-0). The basic idea of the model postulates that the fast energy input to the target surface generates a state of transient thermodynamic instability (a state of disorder, similar but not equal to a melt) which, then, has to rapidly relax. This could be considered as a competition between surface roughening by erosion and surface smoothing

Fig. 3.26 Irradiation dose dependence of LIPSS: Comparison between simulated structures (surface dynamic model [\[126\]](#page-25-5); panels (**a**)–(**d**)) and experimental structures (panels (**e**)–(**h**)). The dose is gradually increased from (**a**) to (**d**) respectively from (**e**) to (**h**). Unfortunately, this is only a qualitative comparison, a quantitative comparison is not possible because of unknown material parameters, such as absorptivity, energy dissipation, etc

because of diffusion and surface tension, resulting in self-organization of the surface morphology. The process is described in the framework of nonlinear dynamics and can cover, by introducing several steps of nonlinearity as a function of feedback and degree of instability (as a function of absorbed energy dose), all types of structures observed (HSFL, LSFL, grooves, spikes), as is shown in Fig. [3.26.](#page-19-7) For more details, see [\[125\]](#page-25-6).

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