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Polarization and complexity effects on ultrafast laser-induced nanoscale surface structuring

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Abstract— Ultrafast laser pulses confine a large number of photons spatiotemporally, allowing them to sculpt solid surfaces with structures smaller than 100nm. Upon multi-shot irradiation the self-organization and growth of periodic patterns arise from localized perturbations of the optical coupling on random surface nanoreliefs. This study shows how simulating engineered ultrafast laser beams interacting with rough surfaces provides unrivaled optical control for creating suitable artificial media with nanofeatures that outperform those of naturally occurring surfaces, with promised novel applications in biomedicine, nanocatalysis, or metaphotonics.

1. INTRODUCTION

The modification and structuring of materials using ultrafast femtosecond laser pulses is a mature technology that has opened up wide fields of research and numerous applications. Today, these focus on the extreme manufacture of surfaces with nanocavities, peaks, grooves or periodic surface stripes. Technological breakthroughs consist in obtaining new structures formed in synergy with the hydro-mechanical response of the material, which can have a high aspect ratio, or by shapes arranged asymmetrically by non-conventional polarization. Also, temporal and polarization pulse shaping allows the response of the excited material to be controlled along specific relaxation processes and can result in exotic shapes or symmetries.

2. METHODS

The key concept of the approach lies in the control of polarization, vorticity, duration, and transverse energy distribution along the light propagation up to the interaction with an inhomogeneous surface. Currently, numerical simulations of laser-surface interaction relies on Finite-Difference Time-Domain schemes that require to initialize the beam from a predetermined intensity distribution and given polarization states [1-2]. In this work, vectorially polarized fields or asymmetrical and twisted beams are used as boundary conditions of the Maxwell equation to generate complex-structured light fields with specially designed intensity, phase, and polarization distributions [3]. Along with the experimental achievements, we have developed a model combining three-dimensional electromagnetic and hydrodynamic approaches to deliver a comprehensive frame for understanding light-induced structuring on tens of nanometer scales [4]. The numerical calculations allow us to investigate the multipulse evolution of surface relief in a self-consistent way, evaluating both the inhomogeneous absorption due to surface roughness and the thermomechanical flows.

3. RESULTS

From a fundamental perspective, complex surface topography is driven by the regulation that originates from the constructive and destructive roles of the electromagnetic and hydrodynamic processes. During the lifetime of a transient laser-induced melting layer, we uncover that dissipative structures can emerge and continuously exchange energy and entropy with photon flux to ensure relief growth and topographical organization during liquid flow. Self-organized nanopatterns can be frozen by solidification, demonstrating the potential of ultrafast laser process for the fabrication of advanced surfaces with a topography designed at the nanoscale. Upon multi-pulse excitation, the electromagnetic roles of collective effects and inter-pulse feedback on the resulting surface topographies will be presented owing to 3D numerical approach [5]. This calculation efficiently couples electromagnetism to hydrodynamics in order to simulate the laser energy distribution, the hydrodynamic flows and the propagation of shock/rarefaction wave.

The presented results on multiphysics simulation deliver a comprehensive frame for the understanding of light-induced structuring on record scales as we elucidate that the generation of

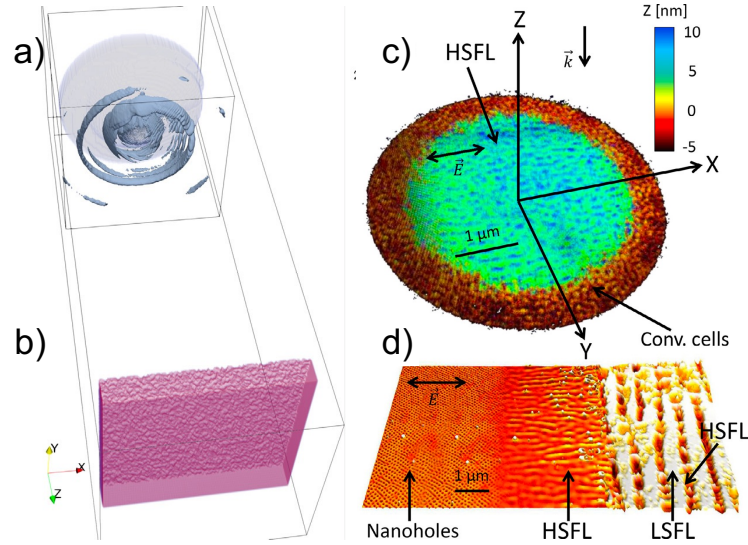


Figure 1: (a) Schematic illustration of numerical electric field distribution for ultrafast laser propagation. (b) Interaction with a rough surface. (c) Inhomogeneous absorption of the field. (d) Nanostructure growth by feedback at different number of pulses (experimental).

nanopatterns result from the interplay between non-radiative optical response on surface, liquid flows driven by surface wave excitation and thermomechanical relaxation. Any surface exposed to sufficiently intense laser irradiation tends to organize along the stationary electromagnetic fields due to the excitation of scattered and surface plasmon waves at the surface. The interference patterns constitute a light footprint that has been observed and exploited since the 70's. The scale of this patterning is mainly defined by the laser wavelength and the features can be controlled by the electric field polarization and incident angle. In addition to this light coupling, the surface self-organizes in response to random fluctuation of light absorption. This establishes complexity fingerprints sculpted on the surface. Driven by optical near fields triggering convection process in a transient thin liquid film, the structures appear at the nanoscale (< 100 nm periodic features) and carry the stigma of nonlinear dynamics.

4. CONCLUSION

Upon ultrafast multiple pulse laser irradiation, morphology-mediated feedback effects strengthen the energy deposition where the electromagnetic response is enhanced. This can be simulated by photonics computations for advanced vectorial beams, revealing complex energy distribution on a surface. The optical and then hydrodynamic response of the rough surface now offers new ways to manipulate material properties at the nanoscale as topographical structures spontaneously emerge with high potential applications in nanotechnologies.

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