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STOCHASTICITY VERSUS DETERMINISM IN LIPSS FORMATION

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ABSTRACT

Understanding the spontaneous emergence of patterns on laser-irradiated surface has been a topic of interest for many years. Brought far from equilibrium under ultrafast photoexcitation, the material exhibits topographic surfaces that reveal signatures from multiphysical coupling, interrogating on the principal mechanism that drives the organisation nature. Some laser-induced periodic surface structures reveal periods typic from electromagnetics, nonlinear optics and plasmonic origin whilst others are characteristic of fluid dynamics or even thermochemical reactions. However, upon multi-shot irradiation, liquid flows are driven by laser-induced energy gradients and patterns are unstable leading to a variety of possible states, including hybrid states of bistable patterns and chaos. Stable spatially-periodic patterns typically arise through small, localized perturbations of the underlying optical coupling on random nanoreliefs. These perturbations grow in amplitude until nonlinear saturation takes over while, at the same time, the nascent topography disturbs the optical response. Whereas the spatially uniform growth into a pattern that can be described within the classic Maxwell and Navier-Stokes equations, the propagation, spatial competition of modes and bifurcation regime require nonlinear dynamics that mimic the behavior of the excited systems near the onset of convective instabilities. One of the challenges is to develop a general and efficient model that inherit relevant symmetry and scale invariance properties and that contain the stochasticity (emergence) and nonlinear properties (dynamics) able to reproduce dissipative structures in spatially extended systems. A stochastic Swift-Hohenberg modelling is then proposed to reproduce hydrodynamic fluctuations at the convective instability threshold that has been recently demonstrated as the very nature of the laser-induced self-organized nanopatterns.

1. DETERMISTIC APPROACH

The issue of laser-induced self-organization of the surface at the nanoscale, originating in near-field light-matter coupling, carries strong scientific weight as it puts forward the question on localizing light on extreme scales [1-2]. 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 [3]. 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. Activated by transverse temperature gradients resulting from complex light coupling on the surface, we observed that the melt flow follows Marangoni surface tension forces. Destabilized by the associated rarefaction wave, we showed that a convection instability develops in thin laser-melted layers up to surface resolidification. This hydrodynamic instability, analog to the Bénard-Marangoni one, drives the matter towards self-organized convection nanocells and, being sensitive to polarization dependence, can induce thermocapillary waves by transverse surface tension gradients. These result in periodic stripes with subwavelength spacings, corresponding to high-spatial frequency LIPSS formation and also to hexagonal convection cell patterns if the polarization dependence is intentionally erased. Coupling electromagnetism to hydrodynamics enables to numerically replicate the different types of periodic structures commonly observed on metal surfaces, their precursors, origins of their periodicity and orientation, and plausible feedback mechanisms for their formation up to the formation of nanopeaks [4].

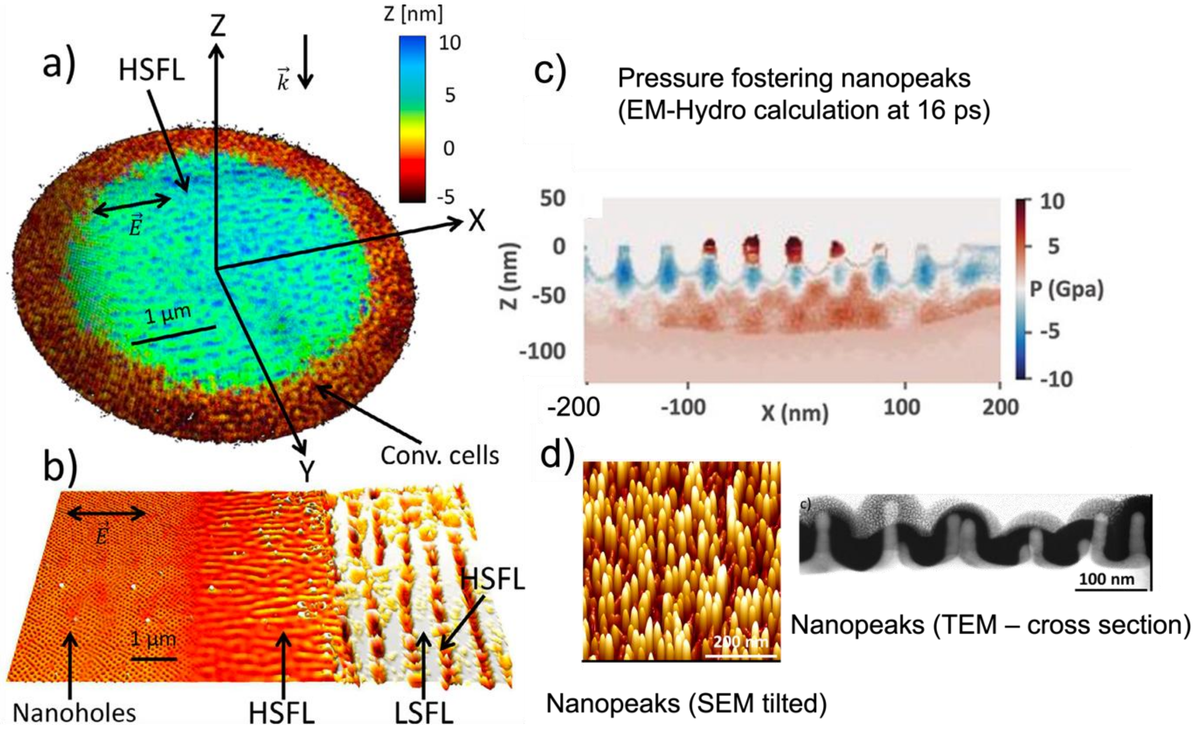

Fig. 1 Comparison between EM-hydrodynamics simulations (a-c) with experimental SEM images (b-d) for nanocavity and HSFL formation conditions (a-b) and nanopeaks generation (c-d) upon ultrafast laser irradiation (100 fs duration). A double-pulse irradiation sequence (25 pulses) with crossed linear polarization has been used to generate the structures. The delay between sub-pulse and the local fluence regulate the kind of patterns.

The results presented in Fig.1 deliver a comprehensive frame for the understanding of light-induced structuring on record scales as we elucidate that the generation of HSFL, hexagonal lattice and nanopeaks formation result from the interplay between non-radiative optical response on surface and surface-tension driven flows, leading to the development of Marangoni convection instability [4]. The optical and then thermoconvective response of a set of asperities now offers a unique process for manufacturing self-organized structures, porous or super-rough surfaces with a resolution of about ten nanometers.

2. NONLINEAR DYNAMICS APPROACH

From a formal point of view, the obtained pattern resulting from convective instability, is initiated by energetical fluctuation effects due to near fields on the surface. Technically, a noise term has been added to the surface topography fostering a strong inhomogeneous absorption that disturbs in turn the uniform response of the deterministic equations of hydrodynamics. Noise-driven unexpected symmetry of patterns and chaos offer a self-consistent framework towards realistic situations in laser-surface situations. However, to advance the understanding of complex dynamics in the presence of fluctuations at the onset of fluid instability, a nonlinear mathematical approach enabling to predict Turing-like patterns resulting to self-organization is proposed.

In convective systems literature, many works investigate self-organized features based on the Swift-Hohenberg model, especially related to the stability of stationary solution and pattern selections as well as the in influence of stochastic external noise. For dissipative dynamical systems, the existence of global attractors, namely a fully invariant and attracting pattern, has been proved.

For Rayleigh-Bénard convection, the Swift-Hohenberg model follows from a 2D projection of the governing fluid equations in the Boussinesq approximation that eliminates the dependence of the temperature, pressure, and velocity fields on the coordinate normal to the surface during the hydrodynamic instabilities.

The SH equation has been intensely investigated as a simplified model to understand the key pattern forming mechanisms and to provide insight into the dynamics of pattern formation in complicated systems. The variety of pattern-like solutions of the SH equation still constitutes an active research topic. Laser-induced pattern formation at the nanoscale can be efficiently characterized and predicted by this kind of stochastic model which is variational in time and conservative in space. Varying the model coefficients (namely a nonlinear strength and a bifurcation parameter that measures the dimensionless distance to the convection threshold in terms of the Rayleigh number) allow to reliably reproduce the nanostructures. Pattern-like solutions of the SH equation are remarkably similar to the ones that we can observe in the irradiated surfaces Scanning Electron Microscope (SEM) images, as can be seen in Fig. 2.

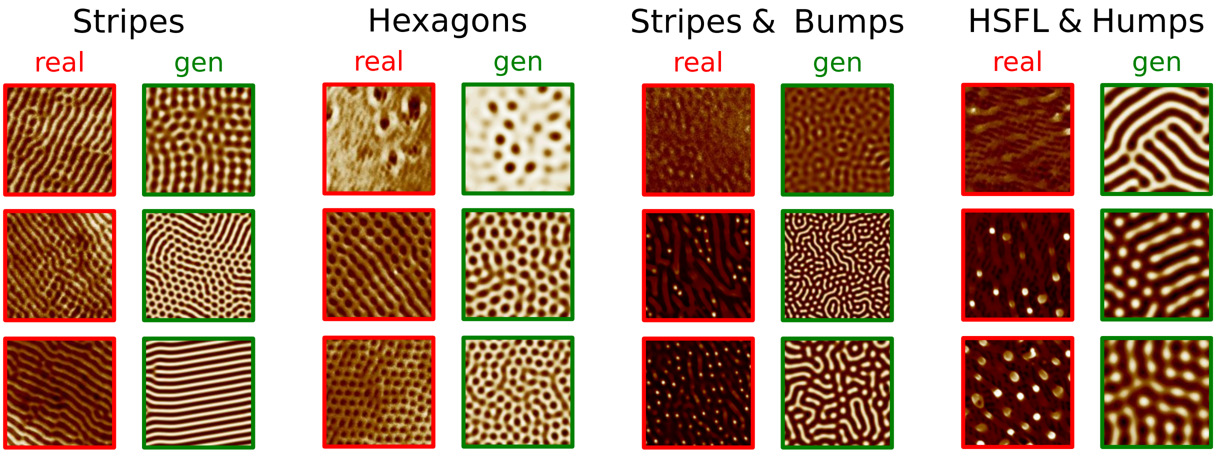

Fig. 2 Comparison between real SEM images (red) and SH-generated images (green). SH generated images are able to reproduce a variety of patterns (e.g., stripes, hexagons, bumps, HSFL, humps) and scales [5]. Since the SH model is an isotropic model, global symmetries are only apparent (e.g., oriented stripes), whereas SEM images retain some measure of global symmetry from laser polarization.

We reveal that the complexity of surface 2D patterns emergence can be learned by a deep convolutional network to connect the model coefficients to the experimental irradiation parameters, providing key process parameters to design a specific pattern [5]. Scale invariant, the model has been finally calibrated on experimental microscopy measurements evidencing that relevant timescale of convective instability process can be derived by this system dynamics modelling.

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