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# Spontaneous surface organization induced by ultra-short laser pulses

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Ultrashort laser sources (100 fs pulse duration) concentrate a large number of photons in time and space, enabling them to transform and sculpt any solid surface achieving ultimate scales of structuring down to 100 nm [1]. Upon multi-shot irradiation the self-organization and growth of periodic patterns arise from small, localized perturbations of the underlying optical coupling on random surface nanoreliefs. These perturbations grow in amplitude until nonlinear saturation takes over while, at the same time, the nascent topography disturbs the optical response by nonlinear feedback. Brought far from equilibrium under successive ultrafast laser pulse photoexcitation, the material progressively exhibits periodic pattern structure that reveal signatures of complexity. Some laser-induced surface structures have periods typical from plasmonic and nonlinear optics origin while others show symmetry breaking characteristics of fluid dynamics, interrogating on the main mechanism that drives the organization nature [2].

Upon multipulse irradiation, a dynamic self-organization established through a positive feedback process, forming 1D patterns known as laser-induced periodic surface structures (LIPSS). Some of them reach periodicities down to one hundred of nanometers. This results in periodic stripes with subwavelength spacings and polarization dependence, corresponding to high-spatial frequency LIPSS formation. If the polarization effects are intentionally erased, complex patterns as hexagonal convection cells have also been reported (see figure 1).

From the fundamental aspect, one of the challenges is to develop a general model that inherit relevant symmetry and scale invariance properties and that contain the nonlinear dynamics able to reproduce dissipative structures in spatially extended systems. A nonlinear dynamics modelling is proposed to reproduce hydrodynamic fluctuations at the onset of convective instability that we have recently demonstrated as the very nature of the laser-induced self-organized nanopatterns. I will show that the complexity of surface 2D patterns emergence can be finally learned by a deep convolutional network to connect the model coefficients to the experimental irradiation conditions, providing key laser process parameters to design a specific pattern [3].

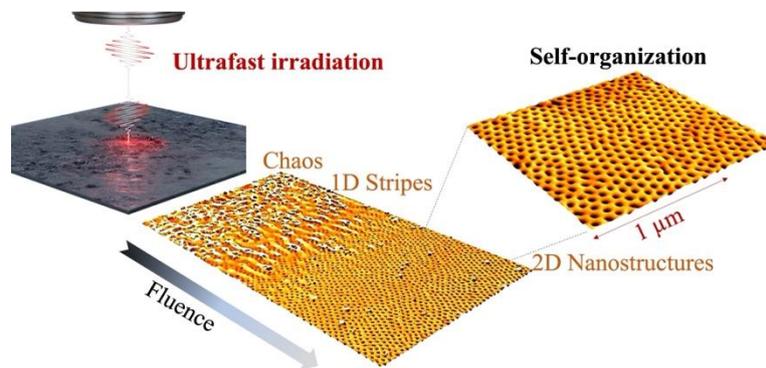


Figure 1: Crossed-polarized double pulse sequence for isotropic energy deposition. An hexagonal lattice of nanocavities of 20 nm in size and spaced by 60 nm has been performed on (100) Ni surface.

Finally, the one-step approach to fabricate nanocavities or nanopеaks with periodicities down to one-tenth of laser wavelength offers a promising way to design and engineer surface properties in optics, plasmonics, sensing and mechanics, where hierarchical nanostructures can provide spectral-sensitive responses, local field-enhancement for nanosensing, or highly efficient catalytic activities [4].

[1] R. Stoian and J.P. Colombier, "Advances in ultrafast laser structuring of materials at the nanoscale", *Nanophotonics* 9(16), 4665-4688 (2020).

[2] A. Abou Saleh, A. Rudenko, S. Reynaud, F. Pigeon, F. Garrelie, & J.P. Colombier, "Sub-100 nm 2D nanopatterning on large scale by ultrafast laser energy regulation", *Nanoscale* 12 (12), 6609 (2020).

[3] E. Brandao, J.P. Colombier, S. Duffner, R. Emonet, F. Garrelie, A. Habrard, F. Jacquenet, A. Nakhoul and M. Sebban. "Learning PDE to Model Self-Organization of Matter", *Entropy*, 24 (8), 1096 (2022).

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