Self-organized patterning and functionalization of surfaces through Nonlinear Laser Lithography

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Laser-induced pattern formation is nearly as old as the laser — the first report dates to 1965. Yet, after several decades, it suffered from poor uniformity and controllability. In 2013, we introduced *nonlinear laser lithography* (NLL), which reached unprecedented levels of uniformity (better than 0.1 nm), long-range order, perfect repeatability, and the possibility to create diverse patterns [1]. We designed the nonlinear dynamics of NLL in analogy to modelocking. Mode-locking creates preferential gain for modes that lock up in phase, leading to a coherent structure in time. We created its analogue by invoking nonlinearities in the form of positive feedback: In NLL, each laser pulse modifies the material surface, and the modified surface changes how the next pulse is scattered, creating a feedback loop (Fig. 1a). A coherent structure grows in space out of initial surface roughness. This growth is regulated by a negative feedback process, as in modelocking. The result is a periodic structure with sub-wavelength periodicity – typically ~700 nm when using 1 µm light (Fig. 1b).

NLL can be used to control not only the tribological properties [2] and wettability [3] but also to form nanostructures to orient liquid crystals [4] and even to pattern 2D materials such as graphene [5]. NLL is universally applicable to any material for which a thermally activated chemical reaction occurs, which we have demonstrated over a dozen different materials, including on flexible, bendable glass substrates. More recently, we achieved advanced control over the dynamics to enact a *programmable* sequence of symmetry breakings to create patterns of nearly unbounded complexity (Fig. 1c). An immediate application is the creation of structured color, achieved straightforwardly by diffraction from the nanostructures.

NLL is a fast, maskless, ambient-air fabrication technique. Encouraged by the preliminary indications of compatibility with graphene applications, we might begin to contemplate exploiting the material-dependent feedback mechanism of NLL together with the electro-optic and nonlinear properties of 2D materials. Such an approach may lead to functional and even dynamic adaptive 2D-material coatings on flexible surfaces.

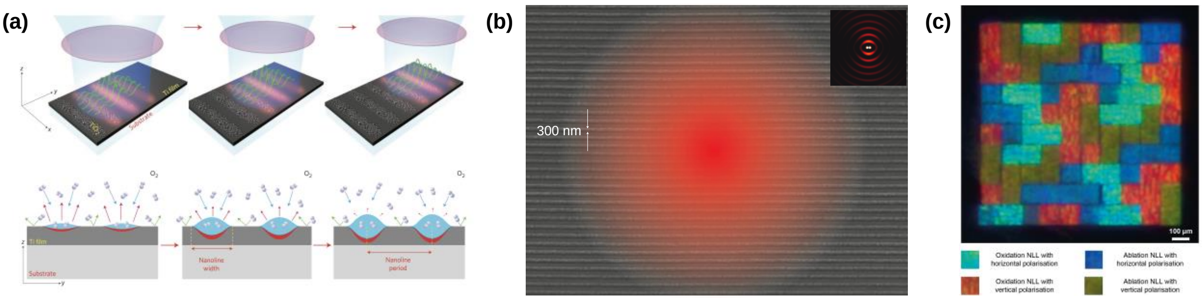

Fig. 1. (a) Top row: schematic of the NLL process is based on the surface being modified, which, in turn, changes how that surface scatters laser light as a laser beam is scanned over the surface. Bottom row: Schematic showing a cross-sectional view of the surface, depicting the deceleration of the growth process due to negative feedback. (b) A typical NLL pattern that was achieved with a 1-µm laser beam. (c) A programmable sequence of symmetry breakings allows the creation of complex patterns.

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