Subwavelength NanoPatterning via Selective Dissolution of One-**Photoisomer** <u>P. Cantu</u>,¹ B. Pollock,² T. L. Andrew,² and R. Menon^{1*}

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Top-down patterning of nanostructures is the key element that introduces functionality at the nanoscale. This is exemplified in numerous devices such as the complex geometries in nanoelectronics, exquisite control of nanoscale dimensions in integrated optics, geometries that impart photonic bandgaps in photonic crystals, and sub-wavelength control of refractive index in metamaterials. In almost all these examples, the nanopatterns are generated via scanning-electron-beam lithography (SEBL), where a focused beam of electrons is scanned over an electron-sensitive polymer resist. On the other hand, SEBL and related approaches that use electrons are extremely slow. Photons, on the contrary, can create patterns extremely fast. However, they suffer from the far-field diffraction limit, and hence, are limited to features that are larger than about one-half the wavelength [1]. Yet, efforts to address this diffraction limit have been largely based on nonlinear processes such as multi-photon absorption.

Patterning via Optical Saturable Transformations (POST) [2-4] is a novel nanofabrication technique that uniquely combines the ideas of saturating optical transitions of photochromic molecules, Figure 1, such as those used in stimulated emission-depletion (STED) microscopy, with interference lithography, which makes it a powerful tool for large-area parallel nanopatterning of deep subwavelength resolution features onto a variety of surfaces with potential extension to 2-dimensions and 3-dimensions.

The photochromic layer is originally in one homogeneous state. When this layer is exposed to a uniform illumination of λ_1 , it converts into the second isomeric state, closed-ring, Figure 1(a-c). Then the sample is exposed to a focused node at λ_2 , which converts the sample into the first isomeric state, open-ring, everywhere except in the near vicinity of the node. By controlling the exposure dose, the size of the unconverted region may be made arbitrarily small. A subsequent fixing step of one of the isomers may be selectively and irreversibly converted (locked) into a 3rd state to lock the pattern. Next, the layer is exposed uniformly to λ_1 , which converts everything except the locked region back to the original state. The sequence of steps may be repeated with a displacement of the sample relative to the optics, resulting in two locked regions whose spacing is smaller than the far-field diffraction limit. Therefore, any arbitrary geometry may be patterned in a dot-matrix fashion.

In this presentation, we will describe our recent experiments to improve the robustness of the POST process and the implications for high-throughput nanopatterning of complex geometries. We will also present simulations that demonstrate the potential of this approach.

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^[2] N.Brimhall, et al, Breaking the Far-Field Diffraction Limit in Optical Nanopatterning via Repeated Photochemical and Electrochemical Transitions in Photochromic Molecules, Physical Review Letters. 107, 205501 (2011).

^[3] P. Cantu, et al, *Subwavelength nanopattering of photochromic diaryethene films*, Applied Physics Letters. 100, 183103 (2012).

^[4] P. Catu, et al. Nanopatterning of diarylethene films via selective dissolution of one photoisomer, Applied Physics Letters. 103, 173112 (2013).



Figure 1: (a) Photochromic molecule (compound 1) used as recording medium in POST. Compound 1 exists as an open-ring isomer 10 and a closed-ring isomer 1c. (b)-(e) Sequence of steps for conventional POST. (f)-(h) Sequence of steps for dissolution-based method.



Figure 2: Linewidth vs exposure time for single exposure and development. The simulated curve is shown as a solid blue line, while the experimental data is shown using crosses. A sinusoidal illumination with period of 457nm was assumed. Inset: SEM images