Direct Imprinting of Aqueous TiO_2 Nanocrystal Dispersions for Sustainable Metasurface Fabrication

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Dielectric metasurfaces composed of subwavelength structures are of great interest due to their potential for strong light-matter interactions and wavefront manipulation. TiO_2 is chosen because it has a high refractive index and is low loss in the visible making it an excellent candidate material for ultrathin optical components. We report a room temperature, environmentally benign, water-based, single-step direct nanoimprint process to pattern metasurfaces using aqueous TiO_2 nanocrystal (NC) inks, which are free of polymer additives or non-aqueous solvents typically used in nanofabrication. We achieve large area (> 625 mm^2) prints on rigid and flexible polymeric substrates, with imprint process parameters tuned to control feature height and residual layer thickness using a soft template with 200 nm features on a 400nm pitch. Resulting imprinted metasurfaces have a high refractive index of 1.94±0.02 at 543 nm.

The TiO_2 NC metasurfaces are designed to resonate at visible wavelengths and are fabricated as two-dimensional (2D) nanopillar gratings atop waveguides. Guided mode resonances within the waveguide couple to the overlaying gratings and scatter into free space, forming high quality factor (Q), quasi-guided mode resonances (QGMs). Electric and magnetic QGM resonances are observed in the NC metasurfaces and their environmental refractive index sensitivities (S) are measured to be 69.1 nm/RIU and 70.4 nm/RIU, respectively, with a figure of merit (FOM) $= Q \times S$ >3000.

The aqueous inks used and the dense films that are printed eliminate the need for environmentally harmful chemicals and high temperature anneals that are typically used in resin-based and sol-gel based processes. This process can be readily extended to the wider NC material library. This serves as a foundation for environmentally benign, scalable, and low-cost manufacturing of optical devices on arbitrary substrates which are of interest for sensing, wearable devices, and flexible lens applications.

Reference:

[1] A. Mallavarapu[‡], C. Lawrence[‡], C. Kagan, "TiO_2 Metasurfaces with Visible Quasi-Guided Mode Resonances via Direct Imprinting of Aqueous Nanocrystal Dispersions", in review, 2023

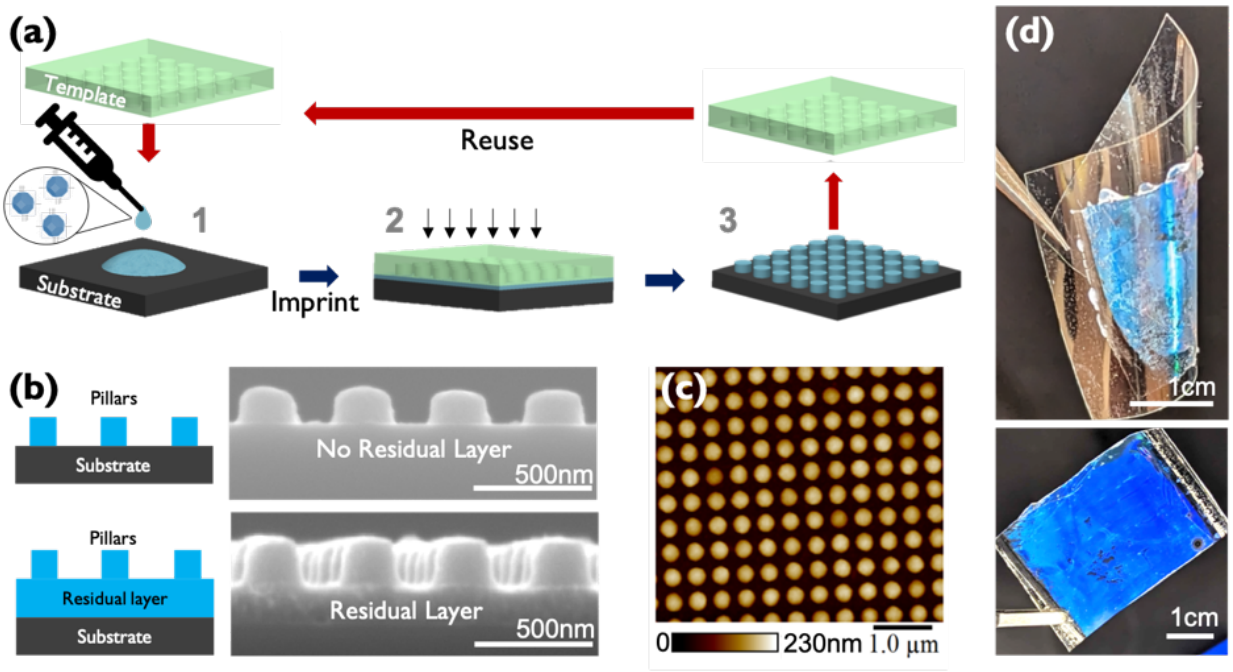

Figure 1. (a) Schematic overview of the NC imprint process: [1] aqueous TiO_2 NC dispersions are drop-cast on a substrate, [2] the template is placed onto and pressed into the dispersion as the solvent evaporates at uniform pressure and room temperature, and [3] the template is released, yielding TiO_2 NC metasurfaces. The template is reused for subsequent imprints. (b) Schematic and cross-sectional SEM images of exemplar imprinted nanopillars on a Si substrate with and without a residual NC layer. (c) AFM image of imprinted TiO_2 NC nanopillars. (d) Photographs of imprinted TiO_2 NC metasurfaces fabricated on flexible polymer and rigid glass substrates.