**Title**

Height Modulation of Nanopixel Arrays *via* Light Controlled Capillary Force Lithography

**Abstract**

Conventional nanotextures typically consist of arrays of identical nanopixels. Recently, efforts have focused on enhancing their functionalities by introducing spatial modulation to the height distribution of nanopixels for the anti-reflective surface, bactericidal effects, and structure colors. However, pre-designed, non-random height modulation holds the potential for even greater functionalities. Many applications require precise nanometric control of nanopixel height as a function of position. Only a few methods, such as grayscale photolithography, nanoimprint lithography, electron beam lithography, dip pen lithography, and two-photon polymerization lithography, can meet these stringent requirements, albeit often at a high cost. Many of these techniques involve chemical development processes, and some rely on expensive pattern generators utilizing lasers, electron beams, or ion beams. Consequently, there is a pressing need for facile and reconfigurable grayscale nanopixel printing methods that offer both high vertical and lateral resolutions without compromise.

Capillary force lithography (CFL), introduced by Suh et al. in 2001, represents a fusion of key aspects from both nanoimprint lithography and soft lithography, combining the molding of a polymer with the use of an elastomeric mold, respectively. This innovative approach preserves the precise pattern fidelity required by nanoimprint lithography while eliminating the need for the high pressures associated with traditional techniques. Furthermore, CFL simplifies the process by removing the requirement for chemical development steps, streamlining the overall procedure. These distinctive features make CFL an attractive method for producing large-area patterns with nanoscale accuracy.

However, CFL alone cannot achieve grayscale nanopixel printing, as it lacks the capability to control the polymer's capillary rise at a predetermined height with nanometric precision, let alone achieve it as a function of position at the micrometer scale. In this research, we have developed such a technique that can optically modulate the height of nanopixels in nanoscale accuracy. Recently, we have discovered that such precise height control in CFL is attainable with certain photopolymers. The key factor enabling this control was the optical pre-modification of the photopolymer's properties critical to capillary rise. This approach holds significant promise for high-resolution, two-dimensionally patterned grayscale CFL, benefiting from the easily microscale patternable nature of light. The "freeze-framing" characteristic of the CFL process also provides an accessible means to observe and analyze the dynamics of the polymeric nanocapillary effect, a phenomenon that has not been fully elucidated to date.