μAngelo: 3D Lithography Based on Thermocapillary Sculpting of Nanofilms

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Abstract

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1. Background

Many fluid systems in nature which are subject to temperature gradients tend to produce intriguing flow patterns dominated at early times by a characteristic length scale λ. Perhaps the most famous such example is the Rayleigh-Bénard instability triggered by buoyancy forces, which occurs in systems ranging in size from millimeters to kilometers and beyond. Above a critical value of the Rayleigh number, plumes of warmer fluid float upwards while cooler, heavier pockets sink downwards, thereby generating fluid cells exhibiting circulatory flow. In this limit, destabilizing buoyancy forces outweigh the stabilizing influence of viscous forces and thermal diffusion. When viewed from above, the convective cells organize into polygonal networks dominated by hexagons whose centers are spaced on average a distance λ apart, as depicted in Fig. 1.

Evaporative remnants of this instability in multicomponent systems can produce spectacular polygonal salt crust formations as seen in the largest salt flat on Earth called the Salar de Uyuni in Bolivia, as shown in Fig. 1.

Fig. 1 Salt ridges arranged in a polygonal mesh. Geometric pattern represents the remnant of flow circulation cells. (Rep. from www.elmundoverdetravel.com/zoutvlakte-tayka)

Once the dimensions of a fluid system are reduced to the micron scale, however, the surface to volume ratio increases tremendously, whereupon surface forces like thermocapillarity become predominant and trigger instabilities such as Bénard-Marangoni flow. In recent years, digital control over thermocapillary forces along an air/liquid interface has led to the development of planar microfluidic devices capable of numerous functions including droplet positioning and registration, routing, coalescence, scission and even mixing [1-2]. In this talk, we focus on even smaller systems sizes of the order of 100 nm or so, to demonstrate how patterned thermocapillary forces can be projected onto the surface of a liquid nanofilm to sculpt material in 3D. Once the driving force is removed, the resulting shapes rapidly solidify *in-situ* forming shapes highly suited to optical applications for spatial beam shaping. We coin this non-contact method of film patterning, μAngelo, and trace its origin to a long standing debate involving spontaneous nanopillar formations in ultrathin films.

2. Electrostatic,Phononic or Thermocapillary Driving Force?

For over 15 years now, researchers have struggled to explain a phenomenon whereby a flat molten polymeric nanofilm exposed to a nearby cooler substrate undergoes spontaneous formation of nanopillars, rings, spirals, chain-like forms, labyrinth and other structures separated by about 1 – 10 microns. Even after solidification, these 3D arrays exhibit ultrasmooth interfaces, which are particularly advantageous for use in optical and photonic systems. Three very different mechanisms have been proposed as the source of instability: (i) electrostatic attraction between the molten film and proximate substrate due to image charge (Chou *et al.* 2002), (ii) radiation pressure from coherent interface reflections of acoustic phonons (Schäffer *et al.* 2003), and (iii) enhancement of film fluctuations by thermocapillary forces (Troian *et al.* 2009 – 2011) [3-5]. Here we demonstrate that the observed instability is likely triggered by long wavelength thermocapillary fluctuations whose growth cannot be suppressed by capillary forces due to the excessively large thermal gradients enforced by experiment. Linear and nonlinear stability analysis of the governing thin film equation indicate there is no critical number for onset of instability and no steady state for systems which are not mass limited. Lyapunov analysis confirms that 3D formations tend to grow toward the cooler target until contact is achieved. Comparison of theoretical predictions with in-house optical microscopy and white light interferometry measurements strongly suggests that the thermocapillary mechanism is indeed predominant. Numerical studies also indicate parameter regimes displaying resonant wavelength behavior leading to uniform peak formations.

3. Fabrication of Microlens Arrays by the μAngelo Technique

To capitalize on this finding, we turn toward nonlinear amplification of film deformation by patterned masks of slender pin arrays placed in close proximity and maintained at a cooler temperature than the nanofilm. The resulting temperature distribution projected onto the film surface produces the necessary thermocapillary stress distribution for sculpting desired liquid shapes in 3D. Numerical studies based on finite element simulations are used to tune the amplitude and pitch of the resulting microlens arrays. Scanning white light interferometry of the solidified shapes also helps optimize array parameters for micro-optical applications such as Shack-Hartmann wavefront sensors [6]. We are currently exploring the use of alternative cooled mask patterns to fabricate novel components such as linear waveguides with non-rectangular cross-section. In summary, we hope that our combined studies establish the potential for a new type of 3D lithography based on spatial modulation of surface forces.

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