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Ordered high aspect ratio nanopillar formation based on electrical and thermal reflowing of prepatterned thin films

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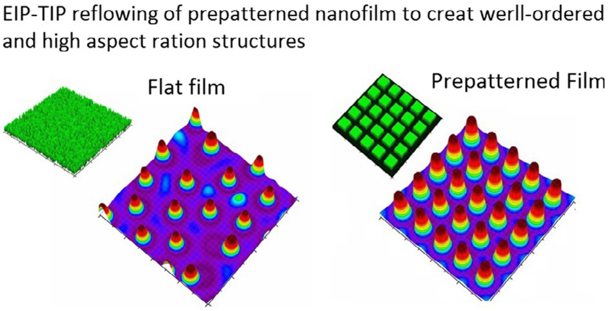
GRAPHICAL ABSTRACT



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ABSTRACT

Creating well-ordered, submicron-sized pillars have been stated as main limitation for electrically induced patterning of nanofilms (thickness <100 nm) [1]. In our previous works, it was shown that the aspect ratio of formed nanopillars was increased to about 0.35 when thermocapillary induced instabilities (Thermally Induced Patterning, TIP) is combined with electrodynamics instabilities (Electrically Induced Patterning, EIP). However, further reduction of pillar size resulted in a coarse and randomly distributed pillars [2,3]. Here, the reflowing of initially prepatterned nanofilms are examined in the EIP and combined EIP-TIP process to create a well-ordered and high aspect ratio nanopillar arrays without sacrificing the fidelity of the final structure. The long-wave approximation is used to simplify the governing equations and boundary conditions leading to a fourth order nonlinear partial differential equation called *thin film* equation that describes the spatio-temporal evolution of the interface. The mechanism of pattern reflowing is discussed for both linear (initial) and nonlinear (long-term) deformations in EIP and EIP-TIP process. The optimum initial pattern width, height and the center-to-center distance is found based on the characteristic wavelength for growth of instabilities predicted by linear stability analysis and nonlinear simulation results.

1. Introduction

Growing demand for straightforward and cost-effective lithography technique to fabricate micro-/nano structures has led to an intense research in fabrication techniques [4–7]. Making smaller structures with enhanced material properties is one main objective for this research for many applications such as opto-electronic devices, sensors and transistors, micro-/nanofluidic systems. More recently, soft lithography and most importantly contact-less patterning techniques, relies on the self-organization of a liquid (molten polymer) layers [8,9,5,10,7]. Electrically Induced

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Patterning (EIP) [11,4] and Thermally Induced Patterning (TIP) [12–15] has emerged as an inexpensive and a straightforward alternative lithography technique for mico-/nanostructing of either conducting and/or non-conducting polymers.

In the EIP, applying an electric field and the difference between the electrostatic properties of liquid film and the bounding layer (the layer which fills the gap between the liquid film and top electrode) results in a net electrostatic (ES) force applied to the interface [16]. The ES force destabilizes the liquid film whereas the viscous force and the Laplace pressure (due to interfacial tension) tend to damp the induced instabilities by the ES force. When the ES force overcomes these damping forces the instabilities grow and the interface reaches to the top electrode to bridge the gap by forming columnar structures called pillars. In the TIP process, a thin liquid film is heated from below and cooled from the top that leads to a large thermal gradient across the micro and nano-sized film. The thermocapillary (TC) force is generated along the interface as the interfacial tension decreases with increasing temperature along the interface. The non-uniformity in the interfacial tension at the interface exerts TC force tangent to the interface which, consequently, leads to a pattern evolution. In ultra-thin (nanofilms) and highly viscous films, the resulting TC instabilities have a large wavelength that leads to motion in the interface. Similarly, it is seen that the instabilities evolve until the columnar structures form and then bridge between the two plates [13–15].

Feature size, aspect ratio (height to width ratio) and their distribution (i.e either randomly distributed or well-ordered) are the main objectives in both EIP and TIP techniques. There have been tremendous efforts to improve pattern formation process, mostly in the EIP, since the initial observation of nanopillar formation in late 1990s [11,12]. The efforts in EIP can be divided into two main streams. One, is working on the electrohydrodynamic (EHD) instabilities in ultra-thin (thickness $\ll 1\ \mu\text{m}$). These are mostly focused on: lowering the pillar size to sub-micron level by various means such as enhancing the electrical properties of liquid film [17–19]; lowering the size of system (electrodes distance of 200 nm and less) and applying high electric field; lowering the interfacial tension by introducing bounding layers other than air [20,21]; and using patterned top electrode as mask to replicate features smaller than the characteristic length predicted by linear stability (LS) analysis [22–25]. There has been limitations in lowering the lateral size of pillars as electric break down of either polymer or bounding layer [26,1] which leads to imperfect pattern formation and impedes creating pillars with aspect ratios (ratio of pillar height to its width) greater than one. In the second stream the EIP process, micopillars formed using much thicker films (initial thickness of a few to hundreds microns) by exposing the film to extra high electric field [27,28]. Very recently, it was shown that electrically induced reflowing of a prepattern films results in very high aspect ratio of pillars (aspect ratio: 3–5) [29,27,30]. Since the initial pattern has a very low aspect ratio (height to width), it can be fabricated either using conventional photolithography or hot embossing [29].

In our previous works, we showed that the combination of EHD and TC instabilities will lead to much smaller sized patterns compared to the EIP and TIP formed patterns [2]. However, the presence of tangential TC forces leads to a higher tendency to merge and make larger size pillars which lowers the efficiency of EIP-TIP process [3]. In this study, for the first time, we focus on reflowing "prepatterned nanofilms" using EIP, TIP and combined EIP-TIP process to investigate the capability of this technique in minimizing the pillars size (increasing aspect ratio) and a well-ordered patterns output. The feasibility of using prepatterned nanofilms will be addressed in the following steps: First the effect of (i) initial pattern shape (cubic or spherical-cap protrusions) (ii) height, width and periodicity on the reflowing mechanism (early stages of deformation) and final formed patterns (nonlinear stages) are discussed. Second, finding a threshold value for the initial protrusion height that required to create well-ordered nanopillars. Third, the effect of applied voltage on creating well ordered and high aspect ratio pillars when using prepatterned film in the EIP-TIP process is investigated. Fourth, finding the limitation for the size of initial patterns which either leads to well ordered and high aspect ratio nanopillars or over time damped and re-organized into a coarse structure with larger sized pillars.

2. Mathematical model

A schematic of the EIP, TIP and EIP-TIP process where an ultra-thin liquid film with either intially flat or a prepatterned shape is shown in Fig. 1. Transverse electric field exposed to the film induces *Maxwell* stress normal the film interface due to mismatch of electrical properties of liquid film and the bounding fluid. The transverse thermal gradient is adjusted by heating the film from below (T_h) while keeping the top plate temperature still above glass transition temperature (T_g) but cooler than the lower substrate ($T_h > T_c > T_g$). The tangential TC stress is resulted due to thermal gradient along the interface and the resulting interfacial tension non-uniformity. Interfacial tension approximated to be a linear [31] function of temperature ($\gamma = \gamma_0 - \alpha_T(T - T_0)$) with $\alpha_T > 0$ as the surface tension gradient, γ_0 and T_0 are the reference interfacial tension and temperature. Initially, the interface is considered a flat film (unless it has been prepatterned) as the interface only affected by Brownian motion whose amplitude is negligible compared to the initial thickness of film.

The liquid film is considered as incompressible and Newtonian fluid. Mass conservation, momentum and energy balances governs the dynamics and pattern formation process. Considering the long-wave approximation, the spatio-temporal evolution of thin liquid films subjected to the transverse electric field and thermal gradient is described by the following dimensionless equation [32],

$$\frac{\partial H}{\partial \tau} + \nabla.\left(H^3 \nabla P - \frac{3H^2}{2} \nabla \Gamma\right) = 0 \qquad (1)$$

where $H = H(X, Y, \tau)$ is nondimensional interface height that is a function of lateral coordinates X, Y and time τ, and $\nabla = (\partial/\partial X, \partial/\partial Y)$. The horizontal coordinates are normalized with characteristic wave-length of $l_c(X, Y = x/l_c, y/l_c)$. The vertical coordinate, interface height and electrodes distance is scaled with film initial thickness h_0 ($Z = z/h_0, H = h/h_0$ and $D = d/h_0$) and temperature is normalized as $\theta = (T - T_c)/(T_h - T_c)$. Variable $\epsilon\ (= h_0/l_c) \ll 1$ is the dimensionless ratio of initial film thickness to the characteristic lateral length scale which confirms the validity of long-wave

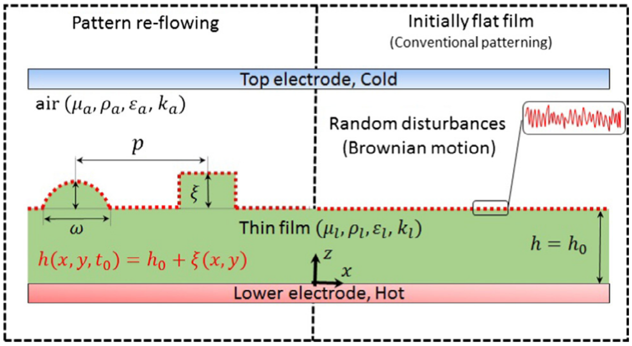

Fig. 1. A 2D schematic of EIP-TIP process. The ultra-thin prepatterned film is sandwiched between top (cold) and bottom (hot) electrodes. Initial height profile in prepatterned film is $h(x, y, t_0) = h_0 + \xi(x, y)$. p is center-to-center distance, ξ is height and w is width of protrusions.