Self-Organized Large Area Patterning of Soft Solids by Elastic Contact Lithography

Ashutosh Sharma*, Manoj Gonuguntla, Rabibrata Mukherjee# and Subash A. Subramanian

Department of Chemical Engineering, Indian Institute of Technology, Kanpur. India

**e-mail:* ashutos@iitk.ac.in

#*Present Address: Central Glass & Ceramic Research Institute, Kolkata, India*

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The surface of a soft elastomeric film (shear modulus < 10 MPa) becomes unstable and forms self-organized patterns when another flat surface (contactor) is brought to its contact proximity. The patterns, which form due to the competitions between the antagonistic effects of van der Waal's forces and restoring forces originating from the deformation of the film itself, form at a length scale ~ three times the film thickness (L ~ 3H). The morphology of the structures transforms in-situ from isolated pillars to bi-continuous labyrinths and finally to isolated holes, as the contactor is progressively brought closer. Interestingly, the wavelengths (L) of patterns, which is nearly independent of the mechanical properties of the film and the adhesive interactions between the film and the stamp (for example, silanization of the stamp fails to modify the wavelength) are also found to be invariant of the morphology of the structures.[1-5] This is clearly shown in the series images shown in figure 1, where for the same film of thickness 1.63 μm, in contact proximity of a flat contactor, the self organized structures changes from isolated pillars (separation distance between stamp and film~ 220 nm) to bi-continuous labyrinths (separation distance ~ 150 nm). On closer approach of the stamp (separation distance ~ 75 nm), the structures further re-organize to form isolated holes. This allows the creation of in situ manipulable and reconfigurable, even erasable structures by this method. The structures, which originate from the elastic deformation of the surface in contact proximity to the stamp, are transient and disappear once the stamp is withdrawn from the proximity of the film due to elastic relaxation, have been made permanent by exposure to UV irradiation before removal of the stamp.[6]

Here we propose various ways of modulating and controlling the morphology and alignment of the elastic structures, thus providing the basic principles of a potential lithographic technique-- Elastic Contact Lithography, based on the controlling, aligning

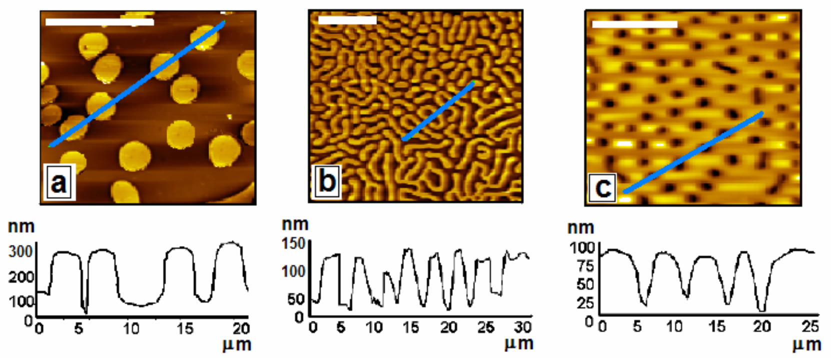

Figure 1: *AFM images of evolution of elastic contact instability patterns with a flat stamp in a* 1.63 μm *thick PDMS film at different separation distances. (a) Isolated columns (scale bar = 10μm), for a separation distance of 220 nm between stamp and film. (b) Labyrinth structure (scale bar = 25μm), when separation distance reduces to ~ 150 nm and (c) isolated cavities (scale bar = 20μm), when stamp-film separation distance reduces to 85 nm. The separation distances are calculated based on the maximum height of the structures.*

and ordering of contact instabilities of a thin soft elastomeric film. Strategies for the same include use of a patterned contactor (instead of a flat one) or a patterned substrate. The patterning thus occurs at the room temperature, in the solid-phase directly and more importantly without the application of any external pressure.[7,8]

Figure 2 shows the variety of distinct structures evolved depending on the separation distance, when a simple 1-D stamp consisting only of parallel grooves (periodicity 3 μm, stripe height 200 nm) is brought in progressive contact with a 960 nm thick soft elastic film. A *positive* replica of the stamp features (1-D parallel stripes under the stamp protrusions) is thus faithfully mirrored in the elastic deformations of the film. The height of the stripe patterns (fig. 2a) obtained is ~ 450 nm, which shows that this method can be used to create high aspect ratio structures starting with a low aspect ratio stamp. As the separation distance between the film and the stamp reduces (~ 430 nm), the raised ridges are slightly compressed, leading to the spontaneous formation of a secondary 2-D periodic structure in the form of bridges joining the parallel neighbouring stripes. This bifurcation to the 2-D instability thus results in the formation of an array of femto-litre beakers as seen in Fig. 2b. A further reduction of the inter-surface distance increasingly tends to produce a *negative* replica of the stamp as seen in fig. 2c, where the polymer ridges now shift to the spaces in-between the stamp protrusions. The height of ridges is 110 ± 3 nm which indicates that these structures are now formed by the imprint

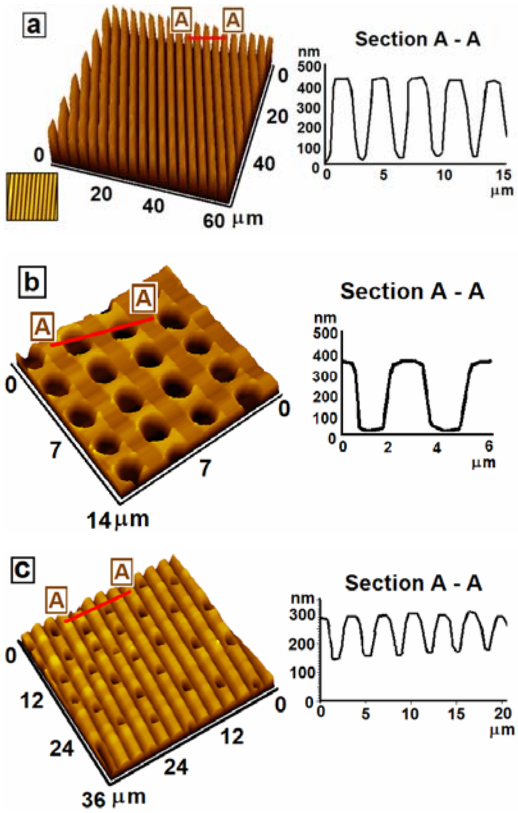

Figure 2*: Modulation and aligning of elastic instability by structures by using a patterned stamp (inset of figure 2a), for* a 960 nm *thick crosslinked PDMS film. (a) Positive replica of the stamp formed by the first adhesive contact. (b) Transition to a 2-D array of femto-liter beakers by lateral bridging between the neighboring stripes at closer approach of the stamp. (c) Greater negative replication of the stamp with remnants of cavities at still closer approach.*

of the stamp on the film. This sequence of image demonstrates the key strength of the process in obtaining patterns that are remarkably distinct from that on the stamp. Generation of patterns distinct from that on the stamp and their in-situ modulations cannot be easily achieved by other known lithographic techniques, thereby enabling *beyond the master* patterning. The other important aspect of the method is by commensuration or mismatch of the natural lengthscale of instability of the film and the stamp periodicity; it is possible to obtain variety of different structures.

Instead of using a patterned contactor, similar ordered structures are also created when a patterned or corrugated substrate (with 1-D stripes) for the film on substrate, using a flat contactor, by proper commensuration between the natural length scale of instability of the film and the substrate periodicity. Figure 3 shows the array of tiny femto-litre beakers obtained by the contact instability of a 320 nm thick PDMS film, coated on a polycarbonate substrate with stripe patterns having periodicity of 800 nm, using a flat contactor.

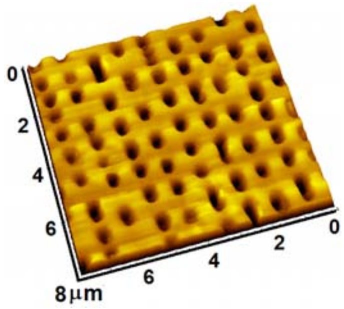

***Figure 3**: Array of femto-litre beakers obtained by the contact instability of a **320 nm** thick crosslinked PDMS film, coated on a stripe patterned polycarbonate surface, having periodicity of 800 nm.*

Based on the results, we conclude unlike the existing soft lithographic methods, which require specialized tools and setup for implementation, the proposed ***Elastic Contact Lithography*** can be used for creating variety of structures over large areas on soft solid surfaces, virtually without the aid of any specialized tools and setup, by merely modulating the separation distance between the film and the stamp. The method, apart from being a technique in which patterning is achieved directly in the solid phase, is also the *only* lithographic method that can be used to create structures beyond those imprinted on the stamp, that is, structures other than negative and positive replicas only.

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