## Underestimated impact of instabilities with nanoimprint

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With nanoimprint, instabilities are 'Janus-faced'. In most cases they result in imprint defects, e.g. when the imprint is performed in thin polymeric layers in order to achieve thin or negligible residual layers. However, instabilities have been proposed to define specific patterns in a polymeric layer with only (partial) contact to the elevated stamp structures, e.g. to shape regular pillar arrays [1].

For these reasons, instabilities have been studied in detail experimentally [2,3] and theoretically [3,4]. They develop, when small gaps between the polymeric surface and the stamp are present. The main drivers are van der Waals forces, temperature gradients and electrostatic forces that are counterbalanced by capillary forces (surface tension). The time scale for their formation depends on the viscosity of the layer. The structures formed become visible when incomplete imprints are 'freezed', either by cross-linking (UV-NIL) or by cool-down (T-NIL). Due to the formation under physical interaction, the process is often termed 'physical self-assembly'. In UV-NIL physical self-assembly is highly correlated with bubble trapping.

The figures document results obtained with polystyrene during T-NIL at 190°C. Capillary bridges (Fig. 1) may span gaps of more than a micron in height; their formation results in a strong re-arrangement of the polymeric layer as suction effects deplete the surrounding of the bridge. Even without local contact to the stamp, the polymeric surface may look as if it were 'boiling' (Fig. 2), with undulations in the µm-range and heights of up to  $\approx$  500 nm. When the stamp is patterned, the pattern results in a local modulation of the interaction forces and thus in guiding effects for the instabilities. Such a situation is shown in Fig. 3, where the stamp features lines of 400 nm in width and a period of 1.2 µm.

The re-arrangement of polymer in 'faint' contact to the stamp is of mayor impact for the imprint results obtainable. Often a thermal imprint is performed by assembling the imprint stack and then heating it to the envisaged temperature in contact (under low contact pressure, in order to assure isothermal conditions). Warping of both, substrate and stamp, then gives rise to a locally differing gap and thus strong re-arrangement of the polymer so that the final imprint does no longer proceed into a constant layer thickness but rather into a hilly polymeric landscape that hampers a uniform replication. (In fact the results shown in the figures were obtained, when the imprint was stopped before full pressure was applied.) We will show that these effects are strong even without intentionally applied temperature gradients or electric fields, experimentally as well as theoretically.

The results may ask for a re-consideration of the imprint procedure since the imprint may start with a highly non-uniform polymeric layer.

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Fig. 1: Capillary bridge formed between the stamp and the substrate. The polymer has assembled into the bridge, leaving the surrounding substrate completely dewetted. In areas without stamp contact polymeric droplets were formed, a consequence from de-wetting starting from the periphery of the capillary bridge.



Fig. 2: Appearance of the polymeric surface in non-contact areas with the stamp (small gap) after heating to 190°C. The surface looks bumpy with undulations in the micron range and heights of about 500 nm.



Fig. 3: Polymeric surface within a region of 'faint' contact to a line field (contact with elevated lines within the circular region in front, surrounded by a non-contact area). Right: Detail of transition: The cavities are partly filled in front, presumably rather by capillary suction than by forced imprint. Beyond the filled region the surface undulations are guided by the elevated lines (in between the cavities!), but with a periodicity determined by the instabilities rather than by the lines.