Nanoinmprint lithography with a focused laser beam for the fabrication of micro-/nano-hybrid patterns†

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1. Conceptual Drawing

A simple process which utilizes a 2D stamp based on UV-NIL is proposed. The main idea is to control the cured region on the micrometer scale by using a developable resist. For the conventional UV-NIL process, a collimated UV light is transmitted through the transparent stamp to cure the overall resist as shown in Fig. S1a. However, if the size of the UV light is reduced, the cured region of the resist can be also narrowed (Fig. S1b). After development, the uncured resist can then be removed and the remaining resist thus forms a coarse structure. On the top surface of the coarse structure, there are fine patterns generated by UV-NIL with the stamp. The width of the coarse structure can be determined by the size of the reduced UV light and the fine pattern on the coarse structure will be the inverted shape of the stamp pattern.

A pinhole or a narrow slit can block the UV light for the aforementioned selective curing process. However, there are limitations to minimize the size of the coarse structure since the light is diffracted from the edge of the pinhole or slit and dispersed when it reaches the resist.

If the selective curing process with UV blocking and the UV-NIL process are separated, it is possible to fabricate such hybrid patterns.18,19 However, focusing of the UV light will be the proper way to reduce the size of the coarse structure independently of the effect of the transparent stamp in the proposed concept.
2. Optical Design

For any cases of the optical design, minimization of the stamp thickness is preferred, since the spherical aberration is increased proportionally with the thickness. When handled, a hard thin stamp will be fragile and a soft thin stamp will be deformable. The thickness of the stamp needs to be balanced between the spherical aberration it will cause and the mechanical properties. Considering the fabrication and utilization of the stamp for FLB-NIL, the stamp was assumed to have 1 mm-thickness.

The optical layout of the FLB-NIL system is shown in Fig. S2. The light source is a laser diode (Sony) with the wavelength ($\lambda$) of 405 nm (near UV, h-line) and an optical power of 15 mW. The laser beam is collimated by a collimating lens and focused by an objective lens. The numerical aperture (NA) of the objective lens is 0.46. Theoretically, the laser spot size is determined by the diffraction limit and aberrations. When there are no aberrations in any of the optical components, the minimum diameter (D) of the laser spot is determined by the Airy disk as

$$D = 1.22 \frac{\lambda}{NA}.$$  \hspace{1cm} (1)

The minimum diameter of the laser spot is calculated as about 1.1 $\mu$m. In this case, as the stamp becomes thicker, the laser spot size increases due to the increased spherical aberration. The optical components should be optimally designed by considering the thickness of the stamp. Meanwhile, it is desirable for the system to have two optical modes: patterning without a stamp (DLW mode) and patterning with a stamp (FLB-NIL mode). As the DLW and FLB-NIL systems are similar and the only difference is whether a stamp is used, the fabrication result of FLB-NIL can be verified with that of DLW. Considering the fabrication and utilization of the stamp for FLB-NIL, the stamp was assumed to be 1 mm-thickness Pyrex glass. Therefore, the optical system was designed to have the same spherical
aberrations for stamps with thicknesses of 0 or 1 mm; the collimation lens was designed to minimize the laser spot size for a 0.5 mm-thickness stamp. Figure S3 shows a simulation result of the designed optical layout. The Seidel aberration coefficient W040 in waves represents the increase of the laser spot size due to the spherical aberration. When the thickness of the stamp is 0 mm or 1 mm, the absolute values of the W040 are all about 4, as shown in Fig. S3. Therefore, laser spot size can be expected to be similar for the two system modes: without a stamp and with a 1 mm-thickness stamp. As shown in Fig. S2, the reflected beam from the substrate is focused to the image sensor by the beamsplitter, quarter-wave plate, and imaging lens. The objective lens can be adjusted for the defocus error to be minimized by monitoring the image sensor signal.

3. System construction
The system was designed and constructed as shown in Fig. S4. The optical system is fixed at the upper part and the objective lens can move only in the Z direction by a vertical stage. There is a laser diode (Sony) for the light source. The laser beam is collimated by a collimating lens and focused by an objective lens. The objective lens can be adjusted for the defocus error to be minimized. The other parts of the FLB-NIL system are horizontal stages which can move the substrate in the X-Y direction. There is a vacuum chuck to fix the substrate on the top surface of the horizontal stage. The minimum resolution of the horizontal stage is 0.05 μm.

References
Figure S1 Conceptual drawing of reducing the cured region of the resist in UV-NIL. a) Conventional UV-NIL process uses a collimated UV light to cure all the resist. b) If the UV light is focused by an objective lens, the cured region of the resist will be reduced. Then, the cured resist remains after development.
**Figure S2** The optical layout of the FLB-NIL system. The laser beam is collimated by a collimating lens and then focused by an objective lens. The reflected beam reaches the imaging sensor via a quarter-wave plate and a beam splitter. The focusing error can be monitored by signal processing of the imaging sensor.
**Figure S3** Variations of the Seidel aberration coefficient W040 due to the thickness of the transparent stamp in the designed optical layout. For the DLW mode (t = 0 mm) and the FLB-NIL mode (t = 1 mm), the Seidel aberration coefficient has the same values. Thus, the size of the focused laser beam will be the same.
**Figure S4** A picture of the constructed FLB-NIL system. The upper part of the system consists of optical components to produce the focused laser beam. There is a scanning stage in the lower part to move the substrate in the X-Y directions.