## Spatial modulation of nanopattern dimensions by combining interference lithography and grayscale-patterned secondary exposure

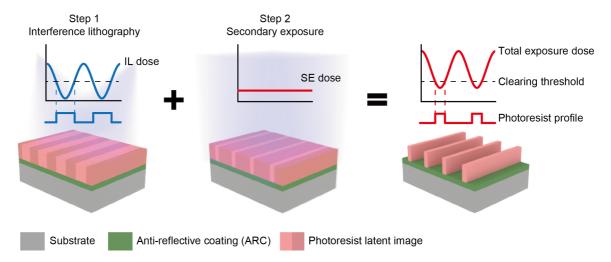
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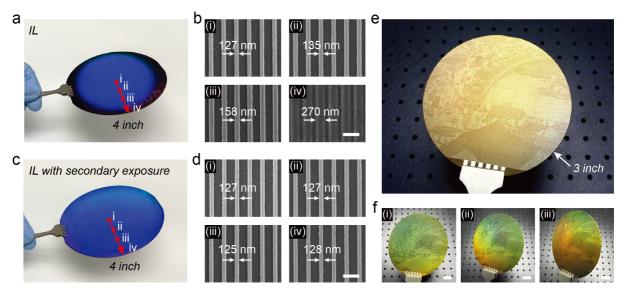
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The precise control of feature size exhibits great importance in fabricating nanodevices for emerging applications. Some applications require nanostructures with uniform feature sizes while others rely on spatially varying morphologies. However, fine manipulation of the feature size over a large area remains a substantial challenge because mainstream approaches of precise nanopatterning are based on low-throughput pixel-by-pixel processing, such as those utilizing focused beams of photons, electrons, or ions. In this work, we introduce a lithographic portfolio combining interference lithography and grayscale-patterned secondary exposure (IL-GPSE) that achieves the spatial modulation of feature size on large area. Employed after the high-throughput interference lithography (IL), a secondary exposure with patterned intensity distribution spatially modulates the dimensions of photoresist nanostructures. Based on this approach, we successfully fabricated 4-inch wafer-scale nanogratings with uniform linewidths of less than 5% variation, using grayscale-patterned secondary exposure to compensate for the linewidth difference caused by the Gaussian distribution of the interfering laser beams. Besides, we also demonstrated a wafer-scale structural painting by spatially modulating the filling ratio to achieve gradient grayscale color using secondary exposure.

**Figure 1** schematically demonstrates IL-GPSE is achieved by a combination of two exposure processes, a high-contrast IL for fabricating large-area periodic nanostructures and a secondary exposure for locally tailoring structural dimensions. The linewidth modulation can be achieved by superimposing the IL exposure dose with a patterned secondary exposure dose. **Figure 2a-d** demonstrate that IL-GPSE allows wafer-scale nanostructure patterning with improved uniformity by compensating the linewidth variation caused by the non-uniform IL exposure field using a specially designed secondary exposure intensity distribution. A 4-inch wafer patterned by uniform 400-nm-period and 125-nm-linewidth nanogratings was demonstrated, with the linewidth uniformity improved by 1100% over IL-only exposure. Figure 2e and 2f demonstrate a 3-inch grayscale painting of Chinese artistic painting *Along the River During the Qingming Festival* (Chinese name: *Qingming Shanghe Tu*) using IL-GPSE, indicating potential applications in structural-color-based cryptography or anti-counterfeiting, etc.



**Figure 1.** The schematic diagram of the IL-GPSE process. Step 1, a two-beam IL is performed on the photoresist to form periodic latent exposed regions with sinusoidal dose profile. Step 2, a secondary exposure follows the IL exposure and increases the effective dose applied on the photoresist which increases the portion of photoresist that receives above-threshold dose, and then leaves less photoresist after development.



**Figure 2.** (a) Photograph of IL-exposed 400-nm-period gratings over a 4-inch wafer with the diffracted blue light intensity gradually decreasing from the center to the edge. (b) SEM images taken in the labelled positions in (a), showing a poor control of linewidth and roughness. (c) Photograph of 400-nm-period gratings over a 4-inch wafer using secondary exposure with a designed projected grayscale pattern after IL exposure, diffracting uniform blue light. (d) SEM images recorded in the labelled positions in (c), showing the uniform linewidth and linewidth roughness. (e) A photograph of the 3-inch grayscale painting of Along the River During the Qingming Festival in photoresist patterned by IL-GPSE. (f) A dynamic structural color due to the residual diffraction at tilting angles. Scale bars, 500 nm (b, d) and 1 cm (f).