## Supplementary material

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**Quantitative analysis and modeling of line edge roughness in near-field lithography: toward high pattern quality in nanofabrication**

**S1 Feature characterization via field distributions in the photoresist**

The point-spread function (PSF) in the photoresist not only determines the resolvable feature size of the developed photoresist structures but also represents the spatial distribution of the exposure field in the photoresist [1]. Thus, the primary objective of this study is to derive an analytical formula of the photoresist PSF. For a two-dimensional metallic hole that includes a bowtie aperture, the field distributions due to the ridge aperture consist of surface plasmon polaritons (SPPs), which is a resonant mode of a flat metal dielectric structure, and quasi-spherical waves (QSWs) [2]. By solving the Helmholtz equation of an E-field in a cylindrical coordinate of the SPP, we can obtain the SPP wave as follows:

 (S1)

where *ASPP* is the SPP amplitude, is the Hankel function of the first type derived from the solution of the wave equation in a cylindrical coordinate, is the wave vector of the SPP, and  is the decay constant in the *z* direction. is an arbitrary phase of the SPP. Because the Hankel function can be approximated using a cylindrical wave, the SPP wave in the radial direction reduces to *ρ*-0.5. Simultaneously, the QSW attenuation is denoted as , where *q* varies from one to two. The evanescent mode of QSW can be expressed as

 (S2)

where is the wave vector in free space and is an arbitrary phase of the QSW. If ρ is quite small compared with the wavelength, e.g., λ/10, *q* converges to one, which implies that QSW acts as a spherical wave. Thus, the local exposure dose distribution on the photoresist surface at can be obtained from the product of the square of the E-field and exposure time

  (S3)

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where  is a possible phase delay between the QSW and SPP. Note that for aluminum at a 405-nm wavelength, and we assume that to obtain equation (S3).

In this work, a sequence of spot patterns with an increasing exposure dose was recorded to estimate the experimentally based PSF, and the developed feature sizes were measured using atomic force microscopy. To extract the decaying wave features at the lateral length without considering the effect of different photoresist sensitivities at different exposure beams , the normalized spatial distribution of exposure dose in a thin photoresist is defined as [1, 3-5]

  (S4)

where denotes the threshold dose that represents the photoresist sensitivity at the minimum lateral distance.

In the line patterns, the line-spread function (LSF) in the photoresist is denoted as which provides a direct measure of the decay constant at the line edge position. By utilizing the relationship between the PSF and LSF, the exposure distribution when a long line is exposed can be obtained by a one-dimensional integral of as

 (S5)

where is the length of the generated line patterns along the *x* scanning direction, is the number of spot patterns, and and are the scanning speed and exposure time, respectively [6]. From equation (S5), we can learn that photoresist LSF is a convolutional result of dose distribution of the PSF with a generated line pattern. Thus, the effects of the decay characteristics of the evanescent field on the line edge roughness (LER) remain.

**S2 Modeling of near-field photoresist contrast**



**Figure S1:** Schematic illustration of the photoresist contrast curve. Normalized remaining thickness of the photoresist film as a function of for a positive photoresist. is the total thickness of the photoresist film and *z* is the generated pattern depth in the photoresist film. is the threshold dose and is the clearing dose.

In near-field lithography (NFL), the photoresist contrast is not only affected by the optical absorption of the photoresist and the development process, which is similar to the far-field optical lithography, but also influenced by the attenuation behavior of the near-field [7-10]. Thus, to accurately estimate the photoresist contrast of the NFL, it is divided into two parts: far-field photoresist contrast and evanescent-field-induced photoresist contrast . In far-field optical lithography, because the depth of focus is greater than the thickness of the photoresist film, the exposure dose loss in the photoresist film can be assumed to only depend on the optical absorption of the used photoresist. Hence, the exposure dose in the photoresist film can be approximated by a simple exponential decay function, , where is the incident exposure dose on the photoresist surface at and is the decay constant determined by the optical absorption. In NFL, the exposure dose in the photoresist film needs to incorporate the exponential decay of the evanescent field along the *z* direction; thus, the exposed dose can be expressed by the following equation:

 (S6)

where is the decay constant of the evanescent field. The photoresist contrast curve is illustrated in Figure S1, and the photoresist contrast is calculated using the formula . By using the threshold dose model, i.e., , the photoresist contrast of far-field lithography can be calculated as . Then the photoresist contrast of NFL, can be expressed as

 (S7)

**S3 Extraction of near-field photoresist contrast using the thickness-versus-dose relationship**

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**Figure S2:** Estimation of the photoresist contrast in the vertical direction. (Solid line with triangles) Measured photoresist contrast curve in the far field generated using laser direct-writing lithography, (solid diamonds) measured near-field photoresist contrast curve exposed using the NFL with a plasmonic bowtie nanoaperture, and (solid line) fitted near-field photoresist contrast curve obtained from equation (S7).

Typically, the photoresist contrast curve is obtained by measuring the amount of exposure dose required to generate a photoresist feature with a given thickness, also known as the thickness-versus-dose function.9,11 Thus, to confirm the accuracy of equation (S7), we first experimentally measured the photoresist contrast curve in the vertical direction. The measured and fitted results are shown in Figure S2. The far-field photoresist contrast was measured to be *5.3*, but the measured near-field photoresist contrast varied with the distance and was much lower than that in the far-field optical lithography, i.e.,  The fitted near-field photoresist contrast curve obtained from equation (S7) agreed well with the experimental data, with a residual error of less than. The results indicated that the near-field photoresist contrastwas approximately equal to the evanescent-field-induced photoresist contrast. Moreover, the proposed analytical formula of the photoresist contrast in the NFL can potentially well estimate the photoresist contrast using the width-versus-dose relationship. By analyzing the calculation result, we can conclude that the exponential decay of the near-field can lead to the loss of photoresist contrast in the NFL.

**S4 LER modeling in NFL **

**Figure S3:** Illustration of the LER modeling in NFL. (a) LER characterization and (b) description of the relationship of the CD control and the ILS based on the photoresist LSF with different exposure doses. The photoresist LSF depicts the exposure dose distribution in the cross-section plane perpendicular to the generated line.

When the near-field probe is scanned in the *x* direction (as shown in Figure S3a) to obtain the amount of LER, the local positions of line edgesandare first measured at regular intervals, i.e.,. The line edges are defined by the LSF tails. Then, the average edges of the two sides, i.e.,and, the measured CD, and the local edge variations, i.e., and, are defined as follows:

 (S8)

whereis the local position measured at the *i*th point of the line edges, is the total measurement number of the point on the line, which is calculated as, and is the scanning length of the line patterns. Typically, LER is quantified as three times the standard deviation, i.e., , of the line edges [8, 11-13].

 (S9)

To obtain the location variation of line edge , the Taylor series expansion near the line edge is used to estimate the exposure dose distribution. For small values of , i.e., smaller than 50 nm (larger than the measured LER value), the local dose distribution can be a good approximation of the Taylor series expansion in the first order with an error of 8% (relative error < 3% with higher order). For example, the exposure dose distributionat the local position of  shown in Figure S3b can be expressed as

. (S10)

By applying the threshold dose condition to the exposure dose at, we can obtain the value ofas

 (S11)

whereis the exposure dose at the feature edge andis the local gradient of the exposure dose at the edge.  is the normalized aerial image contrast at the pattern edge position, anddenotes the exposure dose variation  of the line edge. Whereas patterning using a sufficient amount of photons leads to a clear line-pattern profile with a continuous exposure dose distribution in the theoretical point of view, the used photoresist and other uncertainties (e.g., alignment condition, stage tilt, and vibration) and the overall patterning process can be considered as a random fluctuation process. Thus, to accurately and efficiently estimate LER, the exposure fluctuation is described using the Poisson statistics by estimating the numberof the overlapped exposure spots [6]. The standard deviation of the number of exposure spots is, and the ratio of the standard deviation of the exposure doseand exposure doseis given by. The ratioyields the standard dependence observed in essentially all LER models [11-12, 14]. Thus, for some variations in exposure dose , the resulting LER can be approximated as

 (S12)

where is the normalized exposure dose and is the extracted photoresist contrast.

**S5 Experimental Section**

***Fabrication of the bowtie aperture***: A detailed description of the fabrication of the bowtie aperture used in this work was provided in our previous work [7]; thus, we only briefly describe the SEM imaging condition here. Features of the fabricated bowtie-shaped aperture were measured under the condition of a SEM (JEOL, JSM-7001F), 10-kV acceleration voltage, 79.2-μA emission current, and 9-mm working distance. With the use of SEM, the gap size between the ridges of the bowtie-shaped aperture was precisely measured within uncertainty.

***LER characterization***: Because atomic-force microscopy (AFM) has high resolution and software analysis capability, it was used to obtain accurate pattern features. The generated patterns were measured using the AFM (Park systems, XE-100) with a super sharp silicon tip (Nanosensors, SSS-NCHR, aspect ratio 4, typical tip radius ~2 nm) in non-contact mode. To enhance the measurement precision, the scan rate was adjusted to very slow (<0.3 Hz) and the scanning size was controlled to within 2 μm. After the measurement process, the acquired AFM image was sent to XEI data-analysis software to modify the measured data. The modified AFM image was then exported as a basic image for data processing to obtain the extracted value of the generated pattern features. To evaluate the LER of line patterns with high accuracy and efficiency, the AFM image analysis was performed with our own in-house software. All our code was implemented in MATLAB. As shown in Figure S3a, the sampling interval was calculated by dividing the scan size by the pixel size. The locations of the line edge were extracted by thresholding the gray-level image (shown in Figures 4C and 5B). We found that for the AFM image with good contrast, the threshold value could be selected from a large range without overly affecting the extracted line edge. The extracted line edges were then fitted to lines (including slope). The edge length was measured by taking the distance between the real edge position (the black solid line) and the fitted line (the red dotted line). The LER values represent the 3 value (see equation (S9)), where is the standard deviation. For line array patterns, each line was fitted and the final LER was averaged.

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