Stitching error reduction in electron beam lithography with *in-situ* feedback using self-developing resist

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In electron beam lithography (EBL), a large area pattern is divided into smaller writing fields, which are then stitched together by stage movement to generate the large area pattern. Precise stage movement is essential to minimize the stitching error, and this can be achieved by using laser interferometer-controlled stage. In addition, electron beam deflection must be adjusted to match the stage movement, which is referred to as "writing field alignment." To expose large area nanostructures, a large writing field must be used; otherwise, the stage movement time would be impractically long. However, writing field alignment accuracy decreases with a larger writing field owing to its low magnification. Here, the authors report that self-developing resist (for which the pattern shows up immediately after exposure, thus eliminating the need for ex-situ development) can provide *in-situ* feedback for writing field alignment accuracy, which in turn can be used to optimize the alignment. After several iterations using the exposed test pattern in nitrocellulose (self-developing) resist as feedback, the authors reproducibly achieved nearly perfect ($<$50 nm stitching error) alignment with writing field of 1 mm^2 using a Raith EBL system. © *2013 American Vacuum Society*. [http://dx.doi.org/10.1116/1.4831769]

I. INTRODUCTION

Electron beam lithography (EBL) is the most popular nanolithography method for research, prototyping, and low volume production. The EBL system has a limited area of exposure (writing field), and the large pattern is divided into many writing fields exposed side by side with stage movement between each field exposure. The deviation from perfect alignment between consecutive writing fields is called stitching error. Due to the lack of *in-situ* feedback, conventional EBL is a "blind" open-loop process where the exposed pattern is examined only after *ex-situ* resist development, which is too late for any improvement. It is thus highly desirable to detect and correct the errors *in-situ* before the lengthy exposure of the designed pattern. Here, for the first time, we propose using self-developing resist as in-situ feedback. This method can be used to detect misalignment between adjacent writing fields, as well as distorted or enlarged beam spot due to defocus and stigmation. With this closed-loop process, the beam spot can be optimized globally across an entire writing field,[1] and the stitching error can be minimized. In this paper, we will focus on the latter application and demonstrate substantial reduction of stitching error when using a very large writing field as needed for fast writing.

The *in-situ* feedback is provided by self-developing resist, for which the exposed test pattern shows up and can be examined immediately after exposure by SEM at high magnification. This is in contrast to conventional resist that requires *ex-situ* development using solvent or aqueous developer. Self-developing electron or ion-beam resists had been extensively studied in the 1980s. For instance, metal halides such as AlF_3 (which is decomposed to form volatile fluorine gas upon electron beam exposure) behave as positive self-developing resists.[2–6] Similarly, nitrocellulose is decomposed upon exposure to electron or ion beam; thus, it is also a positive self-developing resist.[7–10] However, those self-developing resists have been nearly forgotten by the EBL community after their discovery. We believe that this is because the metal halide resists suffer from extremely low sensitivity and the inability to expose arbitrary structures other than very thin lines and dot patterns since the decomposition product, metallic Al, cannot migrate far away from the directly exposed area; whereas nitrocellulose resist always leaves behind a thick nonvolatile residual layer. In fact, nitrocellulose was mainly used as an ion beam resist for which the residual layer is thinner because physical bombardment by the ion beam can help remove the nonvolatile species.[11] Though metal halides offer extremely high resolution, the film is found to be degraded by humidity after long (several weeks) exposure to air. Thus we studied nitrocellulose as an EBL resist. As expected, it behaves like a positive resist since e-beam exposure can also generate secondary electrons to decompose the resist, as an ion-beam does; and the amount of the residual layer is significant. However, a thick residual layer, though undesirable since it lowers SEM imaging contrast, is acceptable for the purpose of *in-situ* feedback.

There are roughly four types of stitching errors as summarized by Bogdanov *et al.*: shift error, field distortion (negligible when the e-beam column is well adjusted), field rotation, and deflector scale error.[12] These stitching errors can be categorized as stochastic and systematic errors. The shift error is caused by the stage movement and is stochastic, and its amplitude depends on the precision of the laser interferometer stage. The deflector scale (zoom) error and rotation error are caused by imperfect calibration of the writing

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field size and rotation relative to stage travel, respectively. They are equal at every writing field border and are thus systematic. Previously, various methods have been demonstrated to minimize the stitching error. Multiple (e.g., four) exposures each at reduced dose (e.g., 1/4×) with shifted boundaries or different writing field sizes can be used to average the stage positioning and decrease the shift error.[13–15] Spatial-phase-locked EBL, in which all writing is done with reference to a grid having long-range spatial-phase coherence that provides feedback on beam location, gives the most accurate field stitching, but with the expense of extra process steps to fabricate the grid using interference lithography.[16] Another method to eliminate stitching error is fixed-beam-moving-stage writing that is available for some EBL tools such as the Raith 150.[17] However, it is effective only for thin or wide line patterns such as a long optical waveguide.

Unlike the multiple exposure approach for minimizing the effect of *stochastic* stitching error, our method aims to reduce the *systematic* stitching error. It is relatively straightforward as it does not require the modification of exposure software or the pre-patterning of the wafer with alignment marks. In the process, the writing field is first aligned relative to stage travel, and then a test pattern is exposed in the self-developing resist near the boundary of each writing field with a minimum of two writing fields exposed next to each other. Next, the exposed pattern at the border of two consecutive writing fields is examined at high magnification, which provides feedback to adjust the writing field alignment parameters (zoom and rotation values for the Raith 150^{TWO} tool). The same procedure is repeated until an acceptable stitching error is obtained.

Obviously, this method cannot be applied to the state-of-art dedicated EBL tool for which e-beam adjustment and writing field alignment are fully automatic. However, such tools are less accessible and more costly. Our method is also not effective for small writing fields such as 100 μm where the random stage movement error (40 nm for the Raith 150^{TWO} tool) would dominate the stitching error for experienced operators. Nevertheless, it may be used as a means to confirm the accurate writing field alignment. This method is most effective for large writing fields such as 500 μm and above, where accurate writing field alignment is challenging due to the low magnification needed for a large writing field. A large writing field is essential if the tool is used to expose large patterns since the time for stage movement is inversely proportional to the *square* of the writing field size. For instance, the total stage movement time would reach 40 000 s for a pattern of 1 cm^2 at 100 μm writing field size using the Raith EBL tool.

II. EXPERIMENT

Nitrocellulose solution was purchased from Sigma-Aldrich, and further diluted with pentyl acetate (1:1 volume ratio) to obtain a film of 300 nm by spin-coating at 2000 rpm. The film was baked at 80 °C for 5 min to drive off the solvent. The resist was exposed with the Raith 150^{TWO} tool at 20 keV. An array of large squares, each 5 μm × 5 μm, was exposed with exponentially increasing doses in order to obtain the contrast curve for the resist.

For the Raith EBL system, there is a standard procedure for the writing field alignment. The laser interference stage movement accuracy is 40 nm, which sets the upper limit of writing field alignment accuracy. Basically, the alignment procedure is to calibrate the beam deflection against the stage movement that is assumed to be absolutely accurate (i.e., ignore the 40 nm random error). In the process, a feature such as a sharp corner of a dust particle is first identified, then the stage is moved by a predefined distance along a certain direction, and the beam is deflected to that position to capture an image of the identified feature. This procedure is repeated for a total of four stage movements along four directions. The locations of the feature within the four images are used to generate the transformation matrix to match the beam deflection with stage movement across the entire writing field. The basic alignment principle for other typical EBL system should be similar. The achievable alignment accuracy obviously depends on the magnification that is determined by the writing field size, as well as on the skill of the operator. After completing this standard alignment procedure, a line array pattern with 500 nm pitch was exposed in the self-developing resist near each writing field boundary at a dose of 22 nC/cm. Next, the exposed pattern was examined at high magnification (e.g., 4000×, versus 100× as determined for the writing field size of 1 mm), which revealed in a "foolproof" manner, the magnitude of the stitching error. The measured magnitudes of the stitching error along the horizontal and vertical directions were then used to correct the zoom (deflector scale) and rotation value, according to the simple formula shown in Fig. 1 caption. The steps of resist exposure, SEM measurement, and zoom/-rotation value adjustment were repeated until a satisfactory stitching error was obtained.

Once acceptable stitching was achieved, using the same zoom and rotation value, we carried out exposure of a regular resist PMMA at line dose of 2.0–3.4 nC/cm, followed by development using methylisobutylketone:isopropyl alcohol (1:3) for 40 s. Finally, 10 nm Cr was evaporated and lifted off, and the Cr pattern was examined by SEM in order to verify the effectiveness of our method.

III. RESULTS AND DISCUSSION

A. Exposure properties of nitrocellulose resist

Figure 2 shows the contrast curve of nitrocellulose exposed at 20 keV with an initial film thickness of 55 nm. As expected, a thick residual layer of 25 nm was left behind even at very high exposure doses. Therefore, nitrocellulose is not a useful electron beam resist for pattern transfer purposes; but it is acceptable for the purpose of providing *in-situ* feedback for EBL. The sensitivity, if defined as the dose for 50% remaining thickness, is approximately 3000 μC/cm^2. The sensitivity is approximately 15 times lower than PMMA (clearing dose ~200 μC/cm^2 at 20 keV), but again this is not a serious drawback for our purpose since the time to expose the line arrays

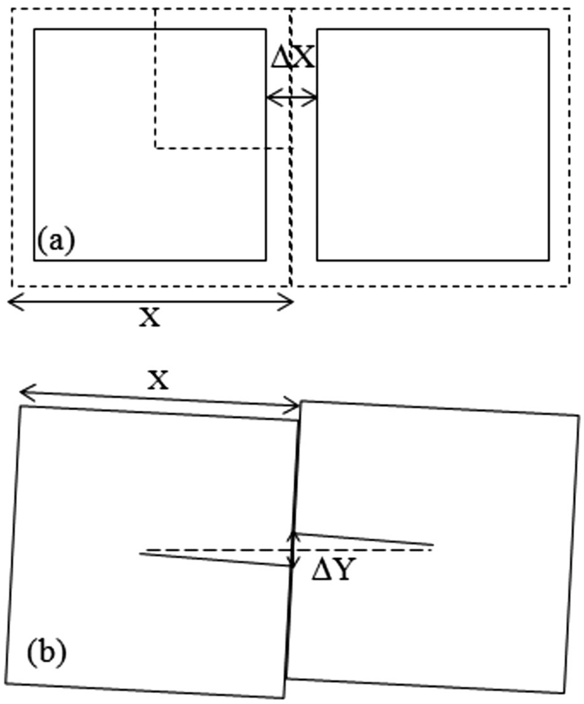

FIG. 1. Schematic showing the adjustment of zoom and rotation value. (a) If the gap or overlap between two adjacent writing fields is ΔX, then the zoom value should be adjusted by a factor of ΔX/X. (b) If misalignment along the vertical direction is ΔY, then the rotation value should be adjusted by $\Delta\theta = \tan^{-1}[(\Delta Y/2)/(X/2)] = \tan^{-1}(\Delta Y/X)$ degree.

near the writing field boundary is very short (∼10 s at 0.1 nA beam current). On the other hand, the sensitivity is approximately three orders higher than metal halide self-developing resist, for which one must design fewer and shorter lines at the writing field boundary if it were to be used for the same purpose. As for the contrast, one cannot derive a meaningful value from the contrast curve, yet clearly the nitrocellulose resist has a low contrast. For the current application, the relative location of the lines exposed at the writing field boundary is more important than their width or array periodicity, so in principle, a low contrast resist can be used to provide accurate feedback on stitching error. However, a low-contrast resist results in a sloped profile for the exposed trenches, which, combined with the low yield of secondary electrons for polymers, leads to reduced SEM imaging contrast at high magnification. This is the main issue we found with nitrocellulose resist.

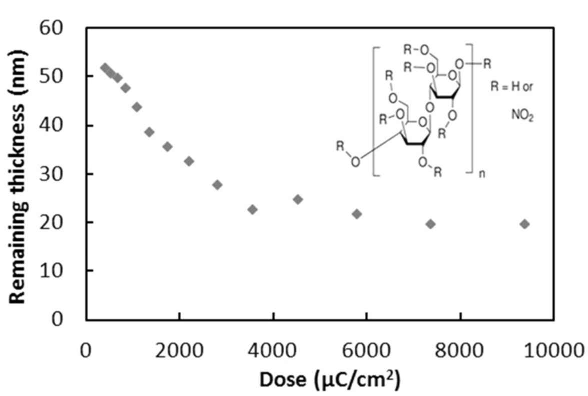

FIG. 2. Contrast curve for nitrocellulose exposed at 20 keV (without *ex-situ* development).

B. Stitching error minimization using nitrocellulose resist

Figure 3 shows the pattern design [Fig. 3(a)] and SEM images of the exposed pattern in nitrocellulose with large

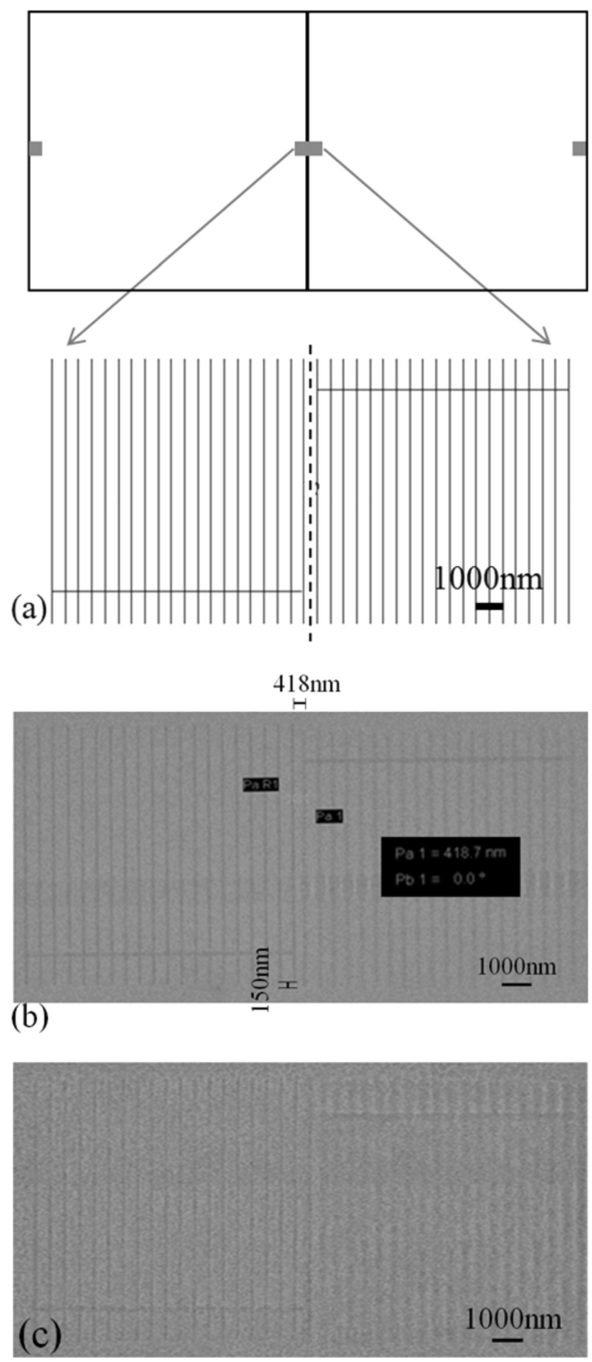

FIG. 3. (a) Pattern design at the writing field boundary; (b) SEM image of a pattern exposed in nitrocellulose at the boundary of two adjacent writing fields, showing significant misalignment of 82 nm and 150 nm along X and Y directions, respectively; (c) SEM image of the exposed pattern showing negligible (<50 nm) misalignment.

stitching error [Fig. 3(b)], and negligible stitching error after several iterations using the aforementioned procedure [Fig. 3(c)]. Here, the writing field size is 1 mm, one order higher than the typical value for high resolution exposure. The pattern consists of 20 periodic lines near the writing field edges with a pitch of 500 nm and length of 10 μm. The distance between the outmost line and the writing field edge is 250 nm, so, when perfectly stitched, the gap between the two outmost lines on the two adjacent writing fields should be 500 nm that is equal to the array pitch. In addition, a horizontal line, that is, 1 μm above/below the lower/upper end of the vertical lines is designed to connect (group) the 20 vertical periodic lines, in order to tell whether a particular vertical line belongs to the writing field at the left side or the right. This pattern design is not optimized, but is enough to serve our purpose. The SEM images [Figs. 3(b) and 3(c)] have a low contrast because nitrocellulose resist has a low contrast that leads to nonvertical profile; in order to keep the electron column condition unchanged, the images were taken at the same high acceleration voltage (20 kV) as for lithography, which gives low secondary electron yield. Imaging at higher magnification showed little improvement because the resist pattern was degraded rapidly (since nitrocellulose is self-developing resist) during the scanning. The low image contrast is a major issue for the current work. One potential solution to this issue is by coating an island film (e.g., Ag or In island film that allows decomposed component to escape) that has much higher secondary electron yield than the polymer resist. It would then be possible to detect misalignment of substantially lower than 50 nm.

Using only the standard writing field alignment procedure, the stitching is stochastic and the error can sometimes reach a high value, depending on the skill of the operator. We achieved an average stitching error (=sqrt[$(\Delta x)^2+(\Delta y)^2$]) of 543 nm with a standard deviation of 334 nm. This large stitching error is not due to the beam drift relative to the stage, since the drift for the Raith 150$^{\text{TWO}}$ system was found to be only ~1 nm/min and the total beam drift would be negligible during the exposure of the test patterns. This large error is partly due to the greatly distorted and enlarged electron beam spot at locations far away from writing field center where small imaging "windows" were opened during writing field alignment procedure. This beam spot distortion can be reduced by defocusing (manually increasing working distance) under the feedback of self-developing resist.[1] Figure 3(b) shows an error of 82 nm overlap along the horizontal direction that indicates too high a zoom value, and an error of ~150 nm shift along the vertical direction that indicates a counter clockwise rotation error. After writing field alignment optimization using the self-developing resist, we obtained nearly perfect alignment as shown in Fig. 3(c), with an error of less than 50 nm in both directions. Under this condition, the same pattern was exposed in PMMA but at a different line dose matching PMMA resist's sensitivity, followed by ex-situ development and Cr liftoff. Figure 4 shows a SEM image of Cr line array pattern across two adjacent writing fields, which again confirmed the nearly perfect alignment.

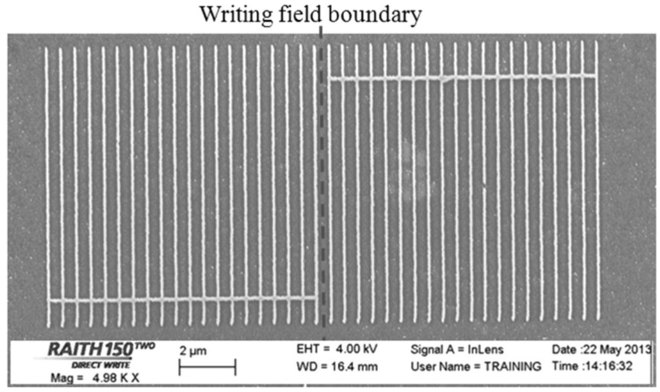

FIG. 4. SEM image of Cr line array pattern across two adjacent writing fields using optimal zoom and rotation values for writing field alignment, showing negligible stitching error.

IV. SUMMARY

We here reported that nitrocellulose, which is a self-developing resist (that is, the pattern shows up immediately after exposure, thus there is no need of *ex-situ* development), can be used very effectively to optimize writing field alignment. In the process, we first exposed a test pattern in nitrocellulose resist near the writing field boundary, and then examined the pattern at high magnification, which provided feedback on the writing field alignment accuracy. Based on such feedback, the parameters for writing field alignment (notably zoom and rotation values for the Raith 150$^{\text{TWO}}$ tool) were adjusted accordingly. After several iterations, we were able to reproducibly achieve nearly perfect (<50 nm stitching error) writing field alignment with a very large 1 mm^2 writing field size.

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