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Mapping of near field light and fabrication of complex nanopatterns by diffraction lithography

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Abstract

We report a single-step lithographic approach for precisely mapping near field light diffraction in photoresist and fabricating complex subwavelength structures. This method relies on the diffraction of UV light from opaque patterns on a photomask, and utilizes the central diffraction maximum (known as the ‘Poisson spot’ for an opaque disk) and its higher orders. By correlating pattern geometries with the resulting diffraction, we demonstrate that the near field light intensity can be quantified to high precision and is in good agreement with theory. The method is further extended to capture higher order diffraction, which is utilized to fabricate unconventional subwavelength nanostructures with three-dimensional topographies. The simplicity of this process and its capability for light mapping suggest it to be an important tool for near field optical lithography.

(Some figures may appear in colour only in the online journal)

1. Introduction

The interest in lithographic patterning of nanostructures is driven by the simple, high throughput, parallel, and wafer-scale characteristics of optical lithography. The challenge of the conventional diffraction limit \cite{1} has led to the development of alternative optical lithographies which rely on the patterning (diffraction, refraction, and interference) of incident light. These approaches employ controlled proximity \cite{2,3}, phase-shift masks \cite{4–6}, surface plasmonic effects \cite{7,8}, and fluidic media \cite{9,10}. These alternative approaches realize, in addition to the subwavelength resolution, the fabrication of unconventional two- or three-dimensional complex nanostructures, which leads to the development of novel functionalized materials, such as photonic bandgap applications \cite{11,12}. However, these lithographic methods are generally expensive, equipment intensive, and require specialized experimental apparatus, e.g., complex lens systems \cite{13,14} or multiple exposures \cite{11,15,16}, and therefore are only achievable in well-equipped research environments. Thus, there is a need for a simple, cost-effective optical lithographic method using readily available microfabrication tools, but with a capability for unconventional fabrication of subwavelength nanostructures.

The development of such approaches depends critically on the understanding/characterization of light patterns in the near field. Despite extensive theoretical work, experimental quantification/visualization of light patterns in the near field has rarely been explored due to the lack of appropriate tools for topographically mapping the light intensity. A few demonstrations relying on interference lithography require the fabrication of non-trivial phase-shift masks \cite{4,6}. Another approach based on proximity lithography \cite{17} remains...
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Figure 1. (a) and (b) show the experimental setup for lithographically producing structures on the photoresist via diffraction of light from a disk patterned on a photomask. In (c), the qualitative variation of the radius of the Poisson spot is shown for decreasing disk radius \(a\) (top, left to right: 2.5, 2, 1.75, 1.5 \(\mu\)m, \(z = 1.3 \mu m\), scale bar = 2 \(\mu m\)), and for decreasing separation \(z\) (bottom, left to right, \(z = 1.5, 1, 0.5 \mu m\)). In (d), we show the quantitative agreement between our experimental results and theoretical predictions for the radius of the Poisson spot as a function of disk radius \(a\).

qualitative due to the difficulty with precise control of the proximity. Other non-lithographical approaches, such as near field scanning optical microscopies (NSOMs), are time-consuming and limited to the characterization of small areas [18].

Here we present a simple, well-controlled method for capturing near field light patterns in photoresist by using diffraction lithography. This approach is based on a conventional hard-contact lithography without involving either complicated or expensive setups, and is compatible with all commercial mask aligners. By characterizing the resist profiles from diffracted light, we demonstrate that the light intensity in the near field can be quantified to high precision, and that the profiles agree well with the classical Fresnel diffraction theory. The approach can be further utilized to fabricate a variety of complex unconventional features with subwavelength dimensions on a large area, which can be precisely transferred to other substrates through a simple transfer method.

In this work, a collimated beam of ultraviolet (UV) light of 405 nm wavelength is scattered from opaque patterns on a transparent photomask, e.g., an opaque disk of radius \(a\). Here we intentionally introduce an accurately controlled separation \(z\) between the photomask and a photoresist-coated Si wafer. As a result, light diffracted behind the disk of separation \(z\) is selectively chosen to expose the photoresist. For an opaque disk, the most prominent feature of this light (or more precisely, any wave [19])

diffraction is a bright spot in the center of the shadow of the disk—the Poisson spot, also known as the Arago spot [20]. Theoretically, the size of the Poisson spot can be controlled via geometrical parameters, such as the separation \(z\), and the radius \(a\) [21, 22]. We note preliminary observations of the Poisson spot effect in optical lithography [23–26]. However, the difficulty with controlling process parameters (e.g., undefined mask photoresist separation and the nonlinear photoresist exposure regime) makes the quantification and the reproducibility of the results problematic, and can lead to seemingly contradictory observations. We expand beyond those qualitative observations and demonstrate the quantification of light intensity to high precision by using a conventional hard-contact optical lithography. We further demonstrate the fine-tuning of diffraction patterns, and its application to fabricate a variety of complicated structures in photoresist, with only simple opaque objects on the photomask, e.g., squares and triangles.

2. The principle of diffraction lithography

Figure 1(a) illustrates the experimental arrangement for the simple case of light diffraction behind a circular disk, and...
Figure 2. (a) An inverted AFM image of the exposed and developed photoresist. The opaque disk on the mask is 2 µm in diameter, and the separation between the mask and wafer is 1.4 µm. (b) The calculated intensity map of the diffracted light from the same opaque disk. The yellow dotted circle indicates the size of the original opaque disk. At the top are the relative light intensity profiles through the center of both images. Relative light intensity 1 is defined as the intensity of the incident light before diffraction. The scale bar is 1 µm for both the images.

Figure 1(b) shows the schematic experimental setup. The disk radius $a$, the separation $z$, and the radius $r$ of the central bright spot, the Poisson spot, are identified in the figures. The separation $z$ was accomplished by an intentionally created photoresist spacer attached to the photomask. This spacer is created by spin coating, exposing, and developing photoresist on the periphery of the patterns of the photomask. The mask and spacer can be used many times without degradation. During exposure, the mask (with the spacer attached) is brought into hard contact with the photoresist-coated wafer using a standard contact aligner, and separation $z$ between the mask and wafer is automatically realized. This setup is essentially a modified proximity lithography system, but without the expensive lithography equipment, and with a more accurate control on the separation $z$ in the near field.

In figure 1(c), the exposed and developed photoresist reveals the effect of the Poisson spot, namely an inner hole at the center of the photomask pattern. For standard hard-contact lithography with $z$ equal to zero, there would be no inner hole in the developed photoresist pattern. The radius of the hole measures the radius of the Poisson spot, the central diffraction maximum. Theoretical predictions for light diffraction from a disk show the intensity of diffracted light, measured out from an on-axis point in the disk shadow, to be proportional to the zero-order Bessel function $J_0(2πra/λ)$ under the condition that $z ≫ r$ [22]. Here, $λ$ is the wavelength of incident light, $r$ represents the distance from the center of the diffraction on the screen, and $ρ = \sqrt{a^2 + z^2}$. The Poisson spot size is defined by the radius of the first-diffraction-minimum ring on the screen, and hence the radius of the inner hole on the subsequently exposed and developed photoresist. This corresponds to the first root, or zero, of the Bessel function $J_0$, which is $J_0(2.40) = 0$ (clearly, further zeros of $J_0$ correspond to additional diffraction minima). Solving for $r$, we find $r = \frac{0.38\rho a}{a}$. This relation predicts that for a given wavelength $λ$, decreasing $a$ (with fixed $z$) results in larger $r$; and decreasing $z$ (with fixed $a$) results in smaller $r$ [22]. This prediction is qualitatively confirmed by scanning electron microscope (SEM) images of the experimental photoresist holes, shown in figure 1(c), and underscores our capacity for controlling the fabrication process through a judicious choice of $r$ and $a$.

Finally, figure 1(d) shows a quantitative agreement between our experimental results and the theoretical predictions for the radius $r$ of the Poisson spot. Error bars of the experimental data are derived from an ensemble of measurements of identical patterns on the photomask.

3. Light intensity mapping and complex nanopatterns

To characterize the intensity of diffracted light behind opaque patterns such as in figure 1, we record a topograph of an exposed and developed photoresist pattern with an atomic force microscope (AFM), as shown in figure 2. Because the experiment is carried out in a regime where the thickness of the developed (removed) photoresist is linearly proportional to the UV energy that it is exposed to [27] (for a fixed exposure time, the UV energy is proportional to the UV intensity), we can then correlate the remaining photoresist thickness to the light intensity (inversely proportional; supporting information
Figure 3. (a) Comparison of simulation versus experiment reveals the variation of the diffraction profiles obtained from the square opaque patterns on the photomask, of varying sizes. From left to right, both simulation and experiment data correspond to square patterns of dimension $4, 3.5, 3, 2.5, 2, \text{and } 1.6 \, \mu m$. (b) SEM image of an array of photoresist structures from the diffraction obtained from $2.5 \, \mu m$ square patterns. (c) SEM image of an array of photoresist structures from the diffraction from equilateral triangular patterns $3 \, \mu m$ (top) and $2.5 \, \mu m$ (bottom) in size. The scale bar is $5 \, \mu m$. $z = 3 \, \mu m$ for all cases.

S1, S2 available at stacks.iop.org/Nano/23/045301/mmedia). Here we have used a non-chemically amplified, thin $\sim 450 \, nm$ S1805 positive resist [28], an exposure of 18 s with 405 nm UV light, and a development of 30 s in MF-319 developer. Figure 2(a) shows an inverted AFM topograph of the photoresist structure (inverted for direct comparison to the light intensity), and figure 2(b) shows a simulation of a light intensity profile (with a yellow dotted circle indicating the perimeter of the disk on the photomask). The top of figure 2 shows profiles through the center of the patterns. There is excellent agreement between the recorded topograph and theory, implying that this approach is a quantitative tool for light intensity mapping. Further work allows for the correlation of optical micrograph color of the developed photoresist with the thickness, indicating a potentially simpler method for quantifying the light intensity (supporting information, S3 available at stacks.iop.org/Nano/23/045301/mmedia). The additional intensity oscillations in the simulation are due to higher order maxima/minima in the diffraction spectrum (supporting information, S4 available at stacks.iop.org/Nano/23/045301/mmedia). In the topograph, one can see both the enhanced outside ring and an indication of weak multiple higher order oscillations inside. These higher order contributions can be enhanced for appropriately designed patterns on the photomask, shown next.

It is worth mentioning that some factors such as standing waves/multiple reflections in the photoresists, and thicknesses of photoresists (e.g., depth of focus) may affect the photoresist profiling as in the case of interference lithographies. In this study, no efforts were taken to account for standing waves (since no evidence was apparent) or multiple reflections, although large effects seem improbable in the near field Fresnel zone. If such effects are problematic, antireflective coatings can be used to mitigate these effects. The concern with the depth of focus is also reduced by noting that only a small thickness of photoresist (not the entire $\sim 450 \, nm$ resist thickness) is used to record the light intensity. The topography of the photoresist profiles has a $\sim 50 \, nm$ maximum (using the experimental exposure times the development rate); figure 2 shows illustrative measured resist profiles. We also note that the resist resolution may have an effect, and the lateral or vertical resist profile resolution will be limited by the resist resolution. In figure 2, the differential development of different ‘grains’ of photoresist can be seen, with a roughness of $\sim 10 \, nm$, and the results presented here should not be considered precise below the resist resolution. Source divergence (e.g., the level of collimation of the illuminating beam) should not be an issue because of the limited depth of focus combined with the resist resolution.

The simulation in figure 2(b) uses the near field approximation of the Kirchhoff–Fresnel diffraction defined by [29]

$$E(x, y, z) = \frac{e^{ikz}}{i\lambda z} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} E(x', y', 0) e^{i\frac{2\pi i}{\lambda z} [(x-x')^2 + (y-y')^2]} \, dx' \, dy'$$

where $E(x, y, z)$ is the electric field of the electromagnetic wave at distance $z$ from the opaque pattern which is in the
x–y plane at z = 0, and for a circular disk, $E(x', y', 0) = E_0(x', y', 0)\Theta((x'^2 + y'^2) - a^2)$, where $E_0(x', y', 0)$ is the incident light intensity, and $\Theta$ is the Heaviside function. The above integral can be written as a shifted Fourier transform of the electric field amplitude at the pattern edge [30]:

$$E(x, y, z) = \frac{e^{ikz}}{4\pi z} e^{i\pi \lambda z (x'^2 + y'^2)} \int_{-\infty}^{+\infty} \left[ E(x', y', 0) e^{i\pi \lambda z (x'^2 + y'^2)} \right] \times e^{-i\frac{2\pi}{\lambda} (x'x + y'y)} \, dx' \, dy'.$$

This permits the use of a fast Fourier transform (FFT) algorithm for numerical calculation of diffraction patterns of an arbitrary 2D opaque pattern with an appropriate modification of $E(x', y', 0)$.

Unlike diffraction from circular disks resulting in a radially symmetric intensity distribution, non-circular patterns produce more complex diffraction profiles with multi-fold symmetries. Figure 3(a) shows images of the diffraction of light from square patterns of various sizes. The top row is a set of SEM images of the post-developed photoresist structures from these patterns, and the bottom row shows the corresponding images of simulated results for the same conditions used in the experiment. At large pattern size, we observe a complex structure with $C_4$ symmetry due to higher order diffraction maxima/minima. As the pattern size is reduced, we see the emergence of four intensity maxima (fourth image from the left) instead of just one. Further reduction in pattern size causes the emergence of a central maximum, similar to the case for diffraction from disks. What is striking is the large variation in complexity caused by varying pattern size with this single-step lithography approach.

Figure 3(b) shows the images of an array of photoresist structures (post-deposited with 10 nm Ti for enhancing the imaging contrast) fabricated by diffraction from 2.5 µm square patterns. The complex photoresist structures were patterned on a large area with excellent uniformity and high reproducibility. Figure 3(c) shows the images of arrays of structures fabricated with diffraction from equilateral triangular opaque patterns on the photomask, of two different sizes (top: 3 µm, bottom: 2.5 µm). We note that, like for figure 3(b), changing the size of the triangular pattern on the photomask results in very complex features involving multiple higher order diffraction maxima/minima, here with an obvious $C_3$ symmetry. The origin of the step structures near the edges of the photoresists is at present unclear. Although other alternative optical lithographies such as interference lithography are also capable of producing seemingly similar or more varied nanostructures using interference phenomena, we argue that the main advance of our approach is the very...
simple process of modulating the diffraction of incident light; therefore it is applicable to any commercially established optical lithography setup.

4. Application to pattern transfer

The above approach can be utilized to fabricate various unconventional nanostructures with subwavelength features with excellent uniformity, and therefore, potentially useful for various applications, as shown next. We first demonstrate the fabrication of vertically aligned Si hollow pillars by utilizing the robustness of the patterned photoresist rings (figure 4). The uniformity of the $\sim 0.25 \text{ mm} \times \sim 0.25 \text{ mm}$ array of transferred photoresist rings is seen in the SEM of figure 4(a), while the wafer-scale uniformity also seems to be excellent according to optical inspection. Starting from the opaque disk patterns (such as in figure 1), by adjusting parameters (smaller radius, nonlinear exposure/development regime) that emphasize the central maximum we have also produced $\sim 100 \text{ nm}$ linewidth photoresist rings (i.e., less than 25% of the wavelength), as shown in figure 4(b). Figure 4(c) shows a resultant array of vertically aligned hollow Si pillars with wall thickness $\sim 10 \text{ nm}$ produced by further reactive ion etching (supporting information, S5 available at stacks.iop.org/Nano/23/045301/mmedia), which would be difficult to achieve with other methods in a single exposure process. These hollow pillars have large surface-to-volume ratio, uniform dimensions and alignment on a large area, and inherit the single crystallographic orientation of the starting wafer along their lengths, implying a potential for Si nanotube-based energy applications such as in thermoelectrics [31] and Li ion batteries [32].

We further demonstrate the fabrication of a corrugated, flexible metal substrate with subwavelength features by transferring an array of photoresist rings (see the schematic in figure 5(a)). A photoresist ring array (figure 5(b)) on an Si wafer was deposited with Au and epoxy, and this was followed by curing and lift-off with a razor blade (steps 1–3 in figure 5(a)). Consequently, epoxy-embedded photoresist/Au patterns (step 4, figure 5(c)) or corrugated Au-only patterns result (step 5, figure 5(d)). The original subwavelength nanostructures were precisely transferred (figure 5(d)) and the Au surface is highly smooth owing to the smoothness of the original Si wafer surface [33]. Such large-area metal substrates with periodic subwavelength features could be useful for flexible electrode or surface plasmonic applications. An investigation of these applications is currently under way. The ability to produce complex three-dimensional structures in a single step would find use not just in direct pattern transfer, but also as a potential method for physical mask creation, such as in imprint lithography or nanofluidic molding.
5. Conclusion

In conclusion, we have presented a simple lithographic method for capturing light patterns in the near field using the diffraction phenomena of opaque patterns. This approach was further utilized to fabricate/transfer unconventional nanopatterns with subwavelength dimensions on a large area. The simplicity and versatility of the process suggest that this method could be readily applicable to any hard-contact lithography, therefore holding great potential for the development of near field optical lithographies.

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