Nanometer resolution mask lithography with matter waves

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Mask based pattern generation with photolithography is a crucial step in microchip production. The next generation Extreme Ultra Violet (EUV) lithography instruments with a wavelength of 13.5 nm is currently under development. In principle this allows patterning down to a few nm resolution in a single exposure. However, there are many technical challenges due to the very high energy of the photons among others. Lithography with metastable atoms has been suggested as a cost effective, less complex alternative to EUV lithography. The big advantage of atom lithography is that the kinetic energy of an atom is much smaller than that of a photon for a given wavelength. However up till now no method has been available for making masks for atom lithography that can produce arbitrary, high resolution patterns. Here we present a solution to this problem. We show that it is possible to make masks based on binary holography that can generate arbitrary patterns down to a few nm resolution using a state of the art metastable helium source. We compare the flux of this source with that of an established EUV source (ASML, NXE:3100) and show that patterns can potentially be produced at comparable speeds. Finally we present an extension of the grid based holography method for a grid of hexagonally shaped subcells. Our method can be used with any wave beam, including other matter wave beams such as helium ions or electrons.

I. INTRODUCTION

In standard photolithography, the resolution is determined by the wavelength of the light: the smaller the wavelength, the higher the resolution. The present industrial photolithography standard is the immersion scanner using a 193 nm light source. Following standard diffraction theory (abbe resolution criterion) this light source gives a maximum resolution in air of 95 nm. This is increased by using off-axis illumination and a high refractive immersion medium, hence the name [1]. Furthermore in modern chip production the patterns are generated by subtle use of underexposure, overdevelopment and multiple exposures, so that patterns with a resolution of around 20 nm can be created. Lithography methods for higher resolution exist, i.e. electron beam lithography which is used to make the masks for photolithography. However, these are all serial lithography techniques and much slower than mask based lithography. The industry is currently implementing the next generation of lithography devices, extreme-ultraviolet lithography (EUV) based on a 13.5 nm-wavelength light source, which is expected to be able to produce patterns with a resolution better than 10 nm in single exposures [1]. The photon energy of this source is 91.84 eV. Atom lithography has been suggested as an alternative to EUV lithography. The de Broglie wavelength of an atom is much smaller than the wavelength of photons for a given energy. High intensity atom beams with narrow velocity distributions can be created by an expansion from a high pressure reservoir through a nozzle followed by selection of the central beam with a conically shaped aperture, which prevents backstreaming into the beam. This aperture is typically referred to as the skimmer [2]. For helium atoms at energies between 0.02 meV (corresponding to a liquid nitrogen cooled beam) and 0.06 meV (room temperature beam), the corresponding wavelengths are between 0.1 and 0.05 nm. This makes atom beams, in principle, a very attractive candidate for high resolution pattern generation. One approach in atom lithography is to use a beam of metastable atoms for the pattern generation. When a metastable atom hits the substrate, it decays, and the energy of the metastable state is transferred to the substrate [3–5]. In their famous paper from 1995 Whitesides et al. demonstrate pattern generation in a thiol-based resist using a beam of metastable argon atoms manipulated by a lightfield mask. Since then numerous groups have experimented with atom lithography using either metastable noble gas atoms and patterning in resist or direct deposition of atoms on substrates.

In most of these experiments the atomic beams were manipulated either by light or electrostatic fields [3–8]. The reason for this is that atoms at low energies, as they typically are in these beams, do not penetrate any substrates. Furthermore the metastable atoms decay when they impinge on a surface. Therefore it is not possible to use masks made on substrates as it is done in photolithography. This limited the patterns that could be made in a mask configuration so far to essentially stripes and dots. Experiments have also been done focusing atom beams with lenses [9–17]. This can be used for serial writing of arbitrary patterns. However, for mass scale production serial writing is not a suitable method.

In 1996 Fujita et al. [18] used a different approach. Instead of using light or electrostatic fields, they made a solid mask consisting of a distribution of uniformly sized holes, etched in a silicon nitride membrane. The hole dis-
tribution was calculated using the theory of grid based binary holography (GBH) developed by Lohmann and Paris \[19\], and later Onoe and Kaneko \[20\]. GBH imposes the limitation on the binary holograms that the openings are all of the same size and positioned on specific positions of a rectangular grid. That is to say, the holes are not only uniformly sized, they are also placed at a regular minimum spacing. The hole distribution is an approximated Fourier transform of the final, desired pattern; Murphy and Gallagher \[21\] extended the binary holography technique of Lohmann and Paris to work also for a hexagonal grid. Originally, the binary holography method was developed to create holograms for electromagnetic waves using a computer, and the procedure is often referred to as computer-generated holography in the literature. Because of the de Broglie wavelength associated with a matter wave, the method also works for atom beams. It may be necessary to include a correction caused by the van der Waal interaction between the mask material and the atoms. However, as shown in a range of experiments \[22\] the only effect will be a slightly smaller effective hole size which can easily be corrected for. We do not discuss this further in this paper.

Because the phase of the atoms when they arrive on the image plane (target plane) is not important, only the intensity is, many different hole distributions can create the same intensity pattern. In a recent publication it was shown how it is possible to vary the number of open holes in a mask over a large range, without changing the final pattern \[23\]. Coverage differences of up to 83 % were demonstrated.

Up till now one major problem with the binary holography method has been that it is based on monochromatic, plane incident and outgoing waves. This means that it cannot be used to make patterns with high spatial resolution without introducing a lens that draws the Fraunhofer diffraction pattern in from infinity. Some work has been done on atomic lenses as mentioned earlier but no lenses with the required precision presently exist. Furthermore real atom sources are not perfectly plane waves and they are not perfectly monochromatic. The monochromacy and spatial coherence of an atom beam is determined by the velocity distribution (wavelength distribution) and extension of the source \[15\]. The ultimate coherent beam would seem to be a Bose-Einstein condensate (BEC). Recently, Zeilinger and co-workers \[24\] generated a beam of BEC metastable helium atoms. However, standard Fraunhofer diffraction theory does not apply for a BEC \[25\]. Furthermore, the de Broglie wavelength of a dropping BEC of helium is very large, about 30 nm after a drop of 0.5 m. For high resolution lithography, one wants to use small wavelengths. One can think of experimental ways to get around this, for example, by moving the mask relative to the BEC so that the BEC wavelength relative to the mask gets smaller, but considerable amendments would have to be made to the theory we present here. It would also be very challenging to make a high flux BEC source.

Recently, a beam of metastable helium atoms with a very narrow wavelength distribution $\lambda/\Delta\lambda = 200$ was produced using a pulsed source \[26\]. In this paper we show that it is possible to make binary masks which can be used to create patterns with nm resolution using an atom sources with this wavelength distribution and no lenses. The lithography setup is shown in Fig. 1. We start out with a target pattern at a specific distance from the mask. Then we compute a field at the mask position that would result in the desired target pattern. The phase of the atom wave when it arrives at the pattern plane is not important for the pattern generation. Therefore there are several possible fields that will create the same pattern, as discussed above. We describe a way to calculate a desired mask field. Then we describe how a mask can be designed that approximate the desired field. The last step is numerical verification of the generated mask.

The main justification for a mask method, is the ability for fast large scale production. Therefore we finish the paper with an estimation of what writing speed (throughput) can be achieved with a state of the art metastable atom source and compare it to that of an established EUV source (ASML, NXE:3300).

**II. THEORY**

![FIG. 1: Overview of the lithography setup: The source, $\psi_0$ is incident at an angle $\theta_t$ on the mask, placed a distance $d$ from the target plane.](image)

The lithography system that we consider is presented in Fig. 1. It consists of a source, a mask that will diffract the incident wave originating at the source, and a target plane, where the diffraction pattern will be displayed. In lithography one aims at creating in the target plane, typically with high resolution, a predefined pattern — the target pattern.

The first step consists of calculating the field at the back plane of the mask (seen relative to the source). This field will be referred to as the mask field in the following. It has the property that when this field is propagated to the target plane and the corresponding intensity distribution is calculated, the target pattern is obtained. To calculate the mask field one therefore
has to back-propagate the field from the target plane to
the mask plane. The target field in the target plane is
taken as the square root of the target pattern, the
desired intensity distribution on the screen, multiplied
by an arbitrarily chosen phase function. By changing
the phase function, different masks are obtained that in
principle should give rise to the same target (intensity)
pattern. The back-propagation of the target field can be
performed in a number of different ways; which approach
is the most appropriate depends on the structure of the
target pattern and the mask-screen separation. Here our
goal is to create high-resolution masks (<10 nm) that can
be used in realistic lithographic setups based on neutral
atom beams. We therefore focus on geometries for which
the source, mask and screen are relatively close together,
that is to say the total system dimension is less than 2 m.
Under this assumption the back-propagation can be per-
formed by the use of (near-field) Fresnel propagation
\[27\].

In the second step the calculated mask field is used as
the starting point for generating the mask. The structure
of the mask is designed so that when an incident field
that originates at the source passes through it, the field
just behind the mask approximately equals the mask field
calculated during the first step.

The third and final step of the design process we pro-
pose involves evaluation of the performance of the gen-
erated mask to verify that it functions as intended. Nu-
merical simulations are used for this purpose. Adequate
results can be obtained by the use of the Fresnel prop-
agation approach similar to what was used under step
one to calculate the mask field. However, in contrast to
what was done under step one, we now forward-propagate
the incident field from the source, through the mask and
onto the screen, where the corresponding intensity dis-
tribution is compared to the target pattern the design
started from.

In the following subsections we will detail each of the
steps of the mask design and evaluation process.

A. Mask field calculation — Fresnel propagation

To calculate the mask field that we want our inci-
dent field to approximate after passing through the holo-
graphic mask, we need a way to propagate the desired
target field backwards from the screen to the mask. An
accurate approximation for describing near-field prop-
agation of scalar fields is the Fresnel diffraction integral
\[27\]. Let \(\psi(r')|z'|\) denote the scalar field in the
mask plane at \(z'\) that we defined by the constant \(z'\) \(\in\mathbb{R}\)
where \(r' = r'_x + z'\hat{z}\) is an arbitrary point in this plane,
\(r'_x = (x', y', 0)\), and a caret on a vector indicates that it
is a unit vector. Similarly, \(\psi(r'_x|z')\) represents the field
in the target plane defined by constant \(z' \in \mathbb{R}\) \([z' > z]\]
where \(r' = r'_0 + z'\hat{z}\) and \(r'_0 = (x', y', 0)\). These two scalar
fields are related by the Fresnel diffraction integral, which
for the plane parallel geometry assumed here, takes the form
\[27\]:

\[
\psi(r_x|z) = \frac{\exp(ik d)}{\lambda d} \int d^2 r'_y \left\{ \psi(r'_0|z') \exp\left(\frac{i k}{2 z'} r'_y^2\right) \right\} e^{-i\frac{\lambda}{2 d} r_x r'_0}, \tag{1}
\]

where \(d = z - z'\) is the mask-screen separation, \(k = 2\pi/\lambda\)
is the wave number of the beam of wavelength \(\lambda\), and the
integration is assumed to extend over the entire
target plane. Equation (1) states that the scalar field in
the mask plane, \(\psi(r_x|z)\), can be obtained from the
Fourier transform of the function \(\psi(r'_0|z') \exp(ik r'_0^2/2z')\),
that involves the target field \(\psi(r'_0|z')\), by first evaluat-
ing the Fourier transform of this function for wave vector
\(K_0 = k r'_0/d\) and multiply the result by a known prefac-
tor. The theoretical foundation of Fresnel propagation is
based on Eq. (1).

When performing Fresnel propagation, high numerical
performance can be achieved thanks to the use of the fast
Fourier transform (FFT). To this end, we first discretize
the spatial coordinates of the mask and target planes. A
flexible way of doing this has been previously described
by Muffoletto et al. \[25\]. This method allows for the cal-
culation of Fresnel propagation between two areas on
parallel planes that have the same number of discretiza-
tion points, but can be scaled and shifted freely within
the valid region of the Fresnel approximation.

The method starts by discretizing the in-plane spatial
coordinates \(r_x = (x, y, 0)\) of the mask plane
\[
x_m = x_0 + m \Delta x, \quad 0 \leq m \leq M - 1 \tag{2a}
\]
\[
y_n = y_0 + n \Delta y, \quad 0 \leq n \leq N - 1 \tag{2b}
\]
and \(r'_x = (x', y', 0)\) of the target plane
\[
x'_p = x'_0 + p \Delta x', \quad 0 \leq p \leq P - 1 \tag{2c}
\]
\[
y'_q = y'_0 + q \Delta y', \quad 0 \leq q \leq Q - 1 \tag{2d}
\]
Here \(M, N, P\) and \(Q\) are all positive integers; \(x_0, y_0\) and
\(x'_0, y'_0\) are known offset parameters; and \(\Delta x, \Delta y\) and
\(\Delta x', \Delta y'\) are the discretization intervals in the mask and target
planes, respectively. By substituting the results (2) into
Eq. (1) and defining
\[
U(m, n) \equiv \psi(x_0 + m \Delta x, y_0 + n \Delta y|z) \tag{3}
\]
\[
u(p, q) \equiv \psi(x'_0 + p \Delta x', y'_0 + q \Delta y'|z'), \tag{4}
\]
one obtains the discretized version of the Fresnel dif-
fracton integral (1) that we write in the form
This equation has the structure of a scaled two-dimensional discrete Fourier transform of the function inside the curly brackets, with the scaling parameters \( s = \Delta x' \Delta x k / d \) and \( t = \Delta y' \Delta y k / d \). For a specific selection of discrete Fourier transform, but we want to be able to select the two coordinate systems freely. Muffoletto et al. \[28\] describes a technique for evaluating scaled discrete Fourier transforms of this kind by taking advantage of results due to Bailey and Swartztrauber \[29\]. This technique is based on rewriting Eq. (5) as a discrete convolution, which can be computed efficiently by performing three FFTs.

The only major restriction coming from evaluating the Fresnel diffraction integral by the method of Muffoletto et al. \[28\] is the requirement that the number of discrete elements in the mask plane and in the target plane must be the same, that is, \( M = P \) and \( M = Q \). However, this limitation can be overcome at the cost of having to perform several Fresnel diffraction steps and shifting either the input or the output region, and tiling the results as described in \[28\].

\[ U(m, n) = \frac{e^{ikd}}{\lambda d} \sum_{p=0}^{P-1} \sum_{q=0}^{Q-1} \left\{ u(p, q) e^{i\pi \left( x_p^2 + y_q^2 \right)} e^{-i \frac{k}{2} \left( x_p x_0 + y_q y_0 \right)} \right\} e^{-i \frac{k}{2} \left( \Delta x' \Delta x pm + \Delta y' \Delta y qn \right)} \]  

\[ (5) \]

\[ \psi(r'\|z) = \frac{d}{\lambda} \int_{\Sigma} d^2 r' \frac{\exp(ikr)}{r^2} \psi(r'\|z') \]  

\[ (6) \]

B. Mask generation – grid based holography

In the preceding subsection we outlined how to calculate the mask field that corresponds to a given target field. Here we will detail how a binary holography mask can be constructed, given a mask field, so that just after an incident beam passes through it, the resulting field will approximately equal the mask field.

Following Lohmann and Paris, and Onoe and Kaneko, a rectangular grid of sampling points is used to cover the mask field with rectangular, non-overlapping areas of equal size (cells). Each of these cells is then divided into a number of rectangular subcells. Some of these subcells are opened so that a scalar field can be transmitted through this open region. The area and locations of the open regions inside the cell determine the amplitude and phase of the field that passes through the cell. The total area of the open regions determine the magnitude of the field associated with that cell. The phase of the field changes along one of the axes of the cell, so that the positions of the openings along this direction allow one to modify the phase of the field propagating from the cell towards the screen in a plane spanned by this direction and the normal vector to the mask plane. Using this method one approximates the sampled value of the mask field by a field propagating towards the screen in a given direction \[19\] \[23\].

Murphy and Gallagher \[21\] extended the binary holography technique by placing the rectangular cells on a hexagonal grid. This means that every other row of cells is shifted. To facilitate the generation of masks based on a hexagonal grid of holes, we have extended the grid based holography method to work with a hexagonal grid of hexagonally shaped subcells. This extension is described in detail in Appendix \[A\].

C. System evaluation – Huygens-Fresnel diffraction integral

After constructing a realization of the mask, one can simulate the target pattern on the screen that the mask gives rise to by using the Fresnel diffraction integral, Eq. (1), as was done to find the mask field. Alternatively a more rigorous approach based on the Huygens-Fresnel diffraction integral can be used \[27\]:

\[ \psi(r'\|z) = \frac{d}{\lambda} \int_{\Sigma} d^2 r' \frac{\exp(ikr)}{r^2} \psi(r'\|z') \]  

\[ (6) \]

with the distance between the points in the source and target plane defined as

\[ r = \sqrt{(r' - r)^2 + d^2} \]  

\[ (7) \]

Here we propagate a field of source points \( \psi(r'\|z') \) to a target point \( r\|z \) and \( \Sigma \) denotes the surface to integrate over, in our case the holes in the mask.

The integral in Eq. (6) is performed for each point in the target pattern by integrating over the area of each hole of the mask using an adaptive integration scheme until a certain convergence criteria has been achieved.

In the simulations we model the supersonic helium source by using an adapted version of the virtual source model introduced by Beijerinck and Verster \[30\]. Recently, the same source model was used successfully to describe experimental measurements of the scattering of a beam of helium atoms from a photonic crystal structure \[31\]. The virtual source model is based on the idea that after an initial region behind the nozzle where the atoms collide, they will eventually reach a free flow regime at a distance from the nozzle referred to as the
quitting surface. When this happens, the individual trajectories can be traced back to a plane that is perpendicular to the mean direction of travel and where the width of the spatial distribution function of the trajectories is at a minimum [30] [32].

We consider the incident beam as an incoherent and weighted superposition of spherical waves or point sources, located approximately in the skimmer plane. The weight (or amplitude) used in the superposition will be taken to follow a Gaussian function whose width, \( \sigma \), mimics the half width of the skimmer. Mathematically, the incident field at position \( \mathbf{r}' \) can be written

\[
\psi_{\text{inc}}(\mathbf{r}') = \int \mathrm{d}^2 \mathbf{r}_\parallel^* e^{-\frac{r'^2}{2\sigma^2}} e^{i \mathbf{k} \cdot (\mathbf{r}' - \mathbf{r}_||^*)} e^{i \phi(\mathbf{r}_\parallel^*)}, \tag{8}
\]

where \( \mathbf{r}_\parallel^* \) denotes a position in the skimmer plane, with the center of the skimmer opening at the origin. The integral in Eq. (8) should be performed over the entire skimmer plane. However, numerically we introduce a cut-off after the Gaussian factor becomes small (after a few standard deviations). In Eq. (8), \( \phi(\mathbf{r}_\parallel^*) \) represents a random phase function associated with the spherical wave source at \( \mathbf{r}_\parallel^* \). We assume that the random phase function should be an uncorrelated stochastic variable that is uniformly distributed on the interval \([0, 2\pi]\). The amplitude of the wave has been set to one in Eq. (8). To perform numerical simulations using the virtual source as an incident field, we must average the calculated diffraction patterns on the screen over an ensemble of realizations of the random phase function.

III. RESULTS AND DISCUSSION

![FIG. 2: Target pattern used. The widths of the lines vary from 10 nm down to 5 nm.](image)

To illustrate the versatility of the theoretical methodology outlined in the previous sections, we now present simulation results with different source and mask parameters. For all simulations we use the same standard test pattern as target pattern, seen in Fig. 2. The pattern consists of a series of corner lines with decreasing width and spacing from 10 nm down to 5 nm. For the simulations the pattern was resolved into 512 \( \times \) 512 pixels and placed at a fixed distance, \( z = 40 \mu\text{m} \) from the mask. For the beam we use an average wavelength: 0.1 \( \mu\text{m} \), which is typical for a helium beam as discussed in the introduction. We use two different source configurations: i) plane wave, that is to say, an ideal, monochromatic, perfectly coherent source and ii) a realistic source configuration as described in section II.C with source-mask distance 1.5284 m and skimmer diameter 400 \( \mu\text{m} \). A wavelength distribution of \( \lambda / \Delta \lambda = 200 \) showed no appreciable impact on the results and was left out in the simulations presented here.

Masks can be made in two different ways: Holes can be "drilled" through a solid membrane as was originally done by Fujita et al. [19]. Alternatively one can use a natural membrane and fill all of the undesired holes. This has the additional advantage of high precision because the position of the holes is built into the material. We have chosen our mask test parameters to reflect these two approaches. We have chosen one rectangular and one hexagonal mask. Both with a hole to hole distance or periodicity of 0.9 nm. This distance is chosen because it represents a natural limit of what one can imagine as possible hole density based on a natural porous material. 0.9 nm is roughly the periodicity of beryl. Beryl is a silicate with a channel structure that allows individual atoms and water to be trapped in the channels. These trapped atoms were recently imaged for the first time using atomic resolution transmission electron microscopy [39]. In principle quartz has similar channels, with a thinner wall structure, which would allow for an even smaller periodicity. However, the thin wall means that quartz tends to turn amorphous when prepared for thin membrane experiments. Holes of the order of 1 nm have recently been fabricated, using helium ion beam lithography, which justifies the rectangular mask [34].

Most self-organized porous membranes, including beryl, have a hexagonal structure, hence the hexagonal mask.

The masks were designed using the grid-based subdivision method, with a minimal open hole configuration [22] to save computation time. The same masks were used for plane wave and realistic source.

The rectangular mask was designed to have an array of 512 \( \times \) 512 cells, with 4 \( \times \) 4 subcells, corresponding to a maximum 2048 \( \times \) 2048 hole openings. The hexagonal mask was made on a grid with 718 \( \times \) 718 subcells. The mask was made from a hexagonally sampled mask field with 4 \( \times \) 3 subdivisions in a cell using the method described in Appendix A.

Fig. 3 shows the simulation result for the plane wave
FIG. 3: (a) Logarithm of the normalized intensity in simulation results for a rectangular mask using a plane wave source. The mask had a periodicity of 0.9 nm, and the distance between the mask and target plane was \( z = 40 \mu m \). The desired target pattern is superimposed on the resulting target pattern, showing that the reproduction is true to size to within 2 nm. (b) A 100 nm \( \times \) 100 nm cutout of the center of the mask.

FIG. 4: (a) Logarithm of the normalized intensity in simulation results for a rectangular mask using a realistic source. The mask had a periodicity of 0.9 nm, and the distance between the mask and target plane was \( z = 40 \mu m \). The desired target pattern is superimposed on the resulting target pattern, showing that the reproduction is true to size to within 3 nm. (b) A 100 nm \( \times \) 100 nm cutout of the center of the mask.

source with mask to target plane distance. Note that the desired target pattern is superimposed on the actual target pattern, to illustrate how well the pattern generation works. We see that the target pattern is reproduced true to size to a precision of around 2 nm. The contrast is 0.92.

Fig. 4 show the simulation result for the realistic source with the desired target pattern superimposed as before. The target pattern is now reproduced true to size to a precision of around 3 nm. The contrast is 0.82.

Finally Fig. 5 shows the last set of results using the realistic source with a hexagonal mask. As expected the pattern is reproduced just as well as with the rectangular mask. The only difference is a slightly lower contrast of 0.77, which we contribute to the fact that the hexagonal mask had a smaller number of sub-cells in total.

IV. THROUGHPUT ESTIMATION

A high wafer throughput is a necessary requirement for chip mass production. It is therefore important to
FIG. 5: (a) Logarithm of the normalized intensity in simulation results for a hexagonal mask using a realistic source. The mask had a distance between neighboring holes of 0.9 nm, and the distance between the mask and target plane was 40 µm. The desired target pattern is superimposed on the resulting target pattern, showing that the reproduction is true to size to within 3 nm. (b) A 100 nm × 100 nm cutout of the center of the mask.

consider what writing speed we can hope to achieve with a mask-based atom lithography device and compare it to what is possible with EUV. We have chosen here for comparison the NXE:3100 EUV tool from ASML. We have used the numbers publicly available at ASML’s websites.

It is not trivial to make a suitable comparison. We have decided simply to compare the photon flux (number of photons per second) and the atom flux (number of atoms per second). It is important to emphasize that this does not take into consideration resist performance, which is a very crucial factor. At the moment very little work has been done on resist development for metastable atoms.

We first calculate the flux for the EUV source. The power is 10 W. The energy of one EUV photon is 91.84 eV = 1.47 × 10⁻¹⁷ J. Thus we get the flux 7 × 10¹⁷ photons/s.

The atom flux can be calculated on the basis of the numbers in reference [26]. For a beam with optimum, narrow velocity distribution, the experimentally measured flux from the source is 3 × 10¹⁶ atoms for a 20 µs pulse. The metastable discharge works with an efficiency of 1 × 10⁻⁴ giving 3 × 10¹² atoms per pulse or 1.5 × 10¹⁷ atoms/s during the pulse.

In this perspective the atom source is comparable in efficiency to the EUV source (1.5 × 10¹⁷ atoms/s versus 7 × 10¹⁷ photons/s). However, in practice the EUV source is superior, since the valve in the pulsed source can currently only operate at 300 Hz. Also, newer EUV instruments with up to 25 times more power than what is listed here are currently under development (NXE:3400B). Still, these simple calculations illustrate the promising potential of the metastable atom sources for lithography.

V. CONCLUSIONS AND OUTLOOK

In this paper we show how nm-resolution mask-based lithography of arbitrary patterns can be carried out using realistic masks and atom sources. In addition we extend the binary holography method to hexagonal cells and subcells, which makes it easier to calculate masks using self-organised porous membranes as mask substrates.

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Appendix A: Binary holograms on hexagonal grids

Murphy and Gallagher [21] extended the binary holography technique of Lohmann and Paris by placing the cells on a hexagonally sampled grid. The cells themselves are constructed as before, but their center points are placed on a hexagonal grid. This means that the cells are rectangular and that every other row of cells is shifted, something that must be taken into account when finding the correct location of each opening. An illustration of the setup can be seen in Fig. 6. The proposed use of hexagonally sampled fields would theoretically make more accurate holograms because of the higher degree of symmetry available when representing circularly sym-
metric functions. Computationally there are also potential advantages, first from a reduction in the amount of data stored and second from a reduction in the amount of work required for propagating the field between hexagonally sampled regions. Fourier methods of field propagation can be extended to work as efficiently on hexagonal grids as on rectangular grids using hexagonal fast Fourier-transforms (HFFT) \[36\].

![Image](image.png)

**FIG. 6:** The Murphy and Gallagher approach: rectangular cells on a hexagonally sampled grid.

![Image](image.png)

**FIG. 7:** New approach: Hexagonal subcells on a hexagonal grid, with two possible sampling schemes: a) Rectangular sampling and b) Hexagonal sampling.

Most self-assembled porous membranes have a hexagonal structure, and we are therefore concerned with adapting binary holography to work with a hexagonal lattice of holes. In this appendix we will discuss the adaptation of the grid based holography method to work with subcells placed on a hexagonal grid. This can be done in a number of ways, but here we are going to focus on two methods. The first method works by filling out a rectangular grid of cells using hexagonal subcells [Fig. 7(a)], while the other fills out a hexagonal grid of cells using hexagonal subcells [Fig. 7(b)].

We will first discuss performing grid based holography with hexagonal subcells by starting from a rectangularly sampled grid. Let us consider a hexagonal grid with one edge of the hexagon parallel with the x-axis. The full width, \(w\), of such a hexagon is related to its height, \(h\), by

\[
w = \frac{2}{\sqrt{3}} h.
\]  

(A1)

Every other column of the grid will be shifted along the y-axis a half height \(h/2\). On such a grid we can create cells with \(m \times n\) subcells, to subdivide a rectangularly sampled grid with

\[
\Delta x = \frac{3}{4} wm, \quad \Delta y = hn.
\]  

(A2)

(A3)

In such a cell not all of the subcells will be aligned on the same rectangular grid, and if we assume a phase change along the y-axis when constructing the hologram this must be taken into account. If we only have a phase change along the x-axis, the contribution from each column will be the same. When performing such an approximation some of the subcells will overlap with the neighboring sample point. This is a natural extension of the ordinary method of grid based holography, but it sets some limitations on the possible rectangular grids that can be used for sampling. An example of such a discretization scheme can be seen in Fig. 7(a), with \(m = 4, n = 4\).

It is also possible to adapt the method of grid based holography to hexagonally sampled mask fields, as can be seen illustrated in Fig. 7(b). In the illustrated case we have also created rectangular cells of \(m \times n\) subcells, but now the numbers \(m\) and \(n\) cannot be selected freely. The cells must have the correct proportions if they are to tile hexagonally, and be of the same shape and orientation. This is only possible for even numbers of \(m\). If \(m\) is odd, adjacent cells have to be flipped upside-down, something that slightly moves their center. In the figure we have \(m = 4, n = 3\) which is one of the valid configurations. In this configuration, the repeating height is \(H = 2nh = 6h\), and the repeating width is \(W = (6/4)mw = 6w\), and we see that both the width and the height is equally scaled.

Similarly to previous schemes, the next step is to assume a phase difference along the x-axis of \(2\pi\) across every cell, and then select in which columns to open subcells and how many subcells to open. The axis of phase change is shown below each figure in Fig. 7.

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