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Fabrication of microsieves with sub-micron pore size by laser interference lithography

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Abstract
Laser interference lithography is a low-cost method for the exposure of large surfaces with regular patterns. Using this method, microsieves with a pore size of 65 nm and a pitch of 200 nm have been fabricated. The pores are formed by inverting a square array of photoresist posts with a chromium lift-off process and by subsequent reactive-ion etching using the chromium as an etch mask. The method has wider process latitude than direct formation of holes in the resist layer and the chromium mask allows for etching of pores with vertical sidewalls.

1. Introduction

Microsieves are a promising innovation in microfiltration technology. Their unique properties offer new filtration possibilities such as, for instance, accurate separation of particles by size. The well-defined and uniformly distributed pores are made with silicon micromachining technology [1, 2]. Microsieves usually consist of a micro-perforated silicon nitride membrane that is attached to a macro-perforated silicon support. The membrane is perforated using photolitography and reactive-ion etching.

Due to diffraction of the UV light the smallest possible pore size is about 1 μm for contact-mask lithography. However, many filtration applications require pore sizes below 1 μm. Using a wafer stepper we have obtained 0.5 μm pores (see figure 1), but for sterile filtration (removal of micro-organisms with membranes) a pore size below 0.22 μm is required, so that even the smallest known bacteria (Pseudomonas diminuta) can be retained.

Such small pores may be obtained using a wafer stepper and deep UV light or electron-beam patterning. However, both techniques are rather time consuming and require expensive machinery.

Another method for obtaining very small pores is laser interference lithography [3–6]. With this method, a feature size smaller than the wavelength of the used light source can be obtained. It may be considered as a low-cost method for the exposure of large areas. Despite these advantages, it has not been widely used in the semiconductor industry, as it can only be applied for the creation of regular patterns. For microsieve production however, such regular patterns are perfectly suitable. Previously we have reported the fabrication of 260 nm pores by direct formation of holes in the resist layer [7]. In this paper, we present an alternative method, in which the interference pattern is inverted, which offers significantly wider process latitude.

2. Theory

When two beams of coherent light interfere, a pattern of parallel fringes will appear. These fringes can be used for the exposure of a photosensitive layer. Figure 2 gives a schematic illustration of this method of exposure.

The depth-of-focus of this method is dependent on the coherence length of the light and can be of the order of metres or more, compared to microns for conventional optical lithography systems. As a result, the demands on substrate flatness and wafer positioning are not critical.

If the light intensity of each beam is I_0, the irradiance on the surface is given by:

$$I = 4I_0 \sin^2\left(\frac{\pi x}{\Lambda_x}\right) \tag{1}$$

with Λ_x the fringe period in the x-direction (see figure 2):

$$\Lambda_x = \frac{\lambda_{\mathrm{UV}}}{2\sin\theta}. \tag{2}$$

Here, λ_{UV} is the wavelength of the laser light in the medium that surrounds the substrate (usually air) and θ is the half-angle between the two beams. The smallest period that can

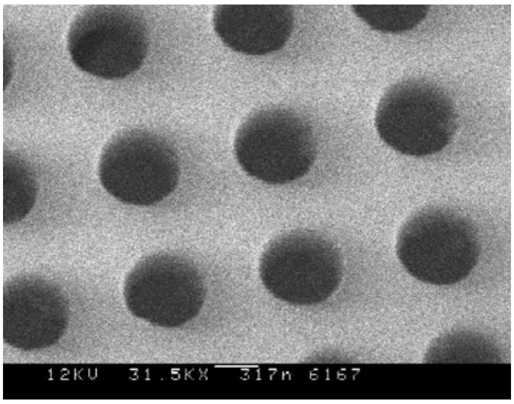

Figure 1. The surface of a microsieve with 0.5 μm pores, obtained with a wafer stepper.

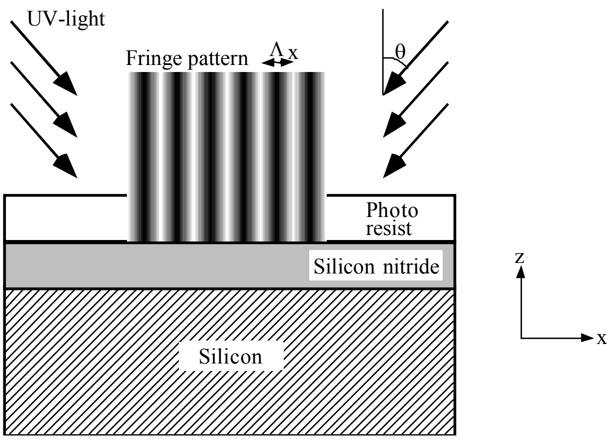

Figure 2. Interference pattern created by two coherent beams.

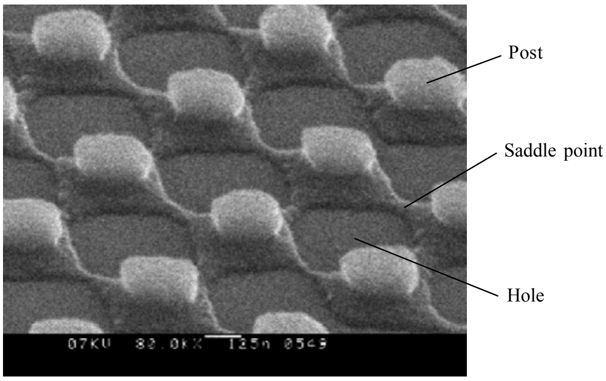

Figure 3. A photoresist layer after a double exposure and development.

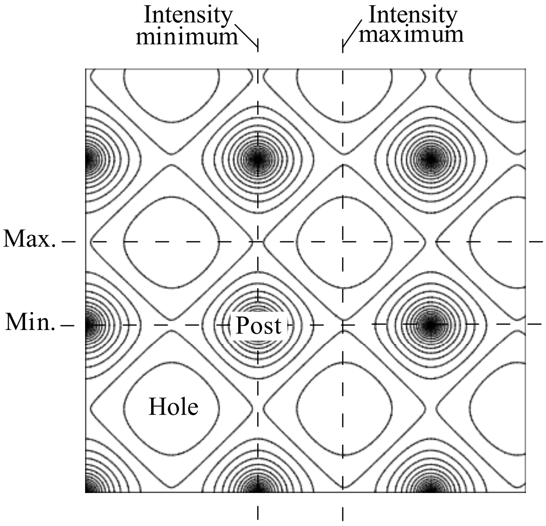

Figure 4. Contour plot of the received dose of UV light in photoresist exposed twice to a fringe pattern with an intermediate rotation over 90°.

theoretically be obtained occurs for $\theta = 90°$ and is equal to $\lambda_{UV}/2$.

For the fabrication of microsieves an array of holes is needed. Such an array can be obtained by a double exposure with an intermediate rotation over an angle α. For $\alpha = 90°$ the array is square and for $\alpha = 60°$ it is hexagonal [8]. After the second exposure, the photoresist layer is developed, where the sum of the two exposure doses determines whether it dissolves in the developer. For a positive resist, the areas that receive a dose above a certain threshold dose will completely dissolve. For a certain (short) exposure time, only the areas where two intensity maxima overlapped will have received a total dose that exceeds the threshold value. These areas will dissolve during development and an array of holes will appear in the resist layer. An SEM micrograph of such holes is shown in figure 3.

The picture shows that the resist between the holes forms so-called 'saddle points', due to the overlap of a minimum and a maximum. For increasing exposure times these saddle points will also dissolve. The result is then an array of posts on the places where two intensity minima overlapped. The transition from holes to posts occurs quite rapidly, as the difference in received dose between the centre of a hole and a saddle point is only a factor of 2. The formation of posts is less critical. In theory the posts will never disappear for increasing exposure times, as the received dose in the centre of a post is always zero. The exposure process for a double-exposed resist layer for $\alpha = 90°$ is explained in figure 4.

The contour lines in figure 4 are iso-dose lines and have been plotted on a logarithmic scale. This implies that, going from a bright region towards a dark region, the difference between two contour lines represents a decrease in received dose by a constant factor (in figure 4 this factor is $\sqrt{2}$). Suppose the first contour in a bright region (where a hole is formed) indicates the threshold dose. The photoresist within this contour line will dissolve and an array of holes will appear. If the exposure time is increased by a factor of $\sqrt{2}$, the next (square-shaped) contour line will indicate the threshold dose. An array of large square holes will appear that are almost interconnected (the result will be similar to the picture in figure 3). Another increase in exposure time leads to the next contour line, which represents the contour of a post. A further increase in time leads to smaller posts, but the line density shows that the change in size of the posts is less dependent on a relative increase in dose than the change in size of the holes. In other words, the process latitude for the formation of posts is wider than for the formation of holes. This is important for the uniformity of the array as, in practice, the laser light varies

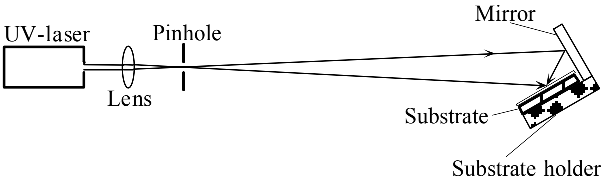

Figure 5. Interference lithography set-up (Lloyd's mirror configuration) for the creation of regular sub-micron structures.

in intensity over the surface due to the Gaussian profile, the distance to the source, imperfect filtering, drift of the beam and laser noise. In order to overcome the limitations of the hole-formation process, the post-formation process can be used in combination with an image reversal method. Decker *et al* [9] have used exposure doses for the creation of posts, but applied before development a base-catalysed method to make the exposed areas insoluble. A subsequent flood exposure with a UV lamp made the previously unexposed parts soluble. The result was an array of holes in photoresist produced with the wide process latitude of the post-creation method.

Pattern transfer from a photoresist mask into a silicon nitride layer usually gives rise to tapered walls due to lateral etching of the mask. As tapered pores affect the filtration performance of membranes, we have developed an alternative process. We create the posts and invert the pattern with a chromium lift-off process, where 15 nm of chromium is evaporated onto the posts, which are then removed in an ultrasonic acetone bath. The remaining chromium forms a perforated layer. This layer serves as an etch mask for plasma etching. Pattern transfer into the silicon nitride improves in comparison with a photoresist mask, as the plasma hardly attacks the chromium.

During exposure, the laser light partly reflects from the substrate and interferes with the incoming light. This causes the creation of an interference pattern in vertical direction. The period Λ_z of this pattern is given by:

$$\Lambda_z = \frac{\lambda}{2n_{\rm res}\cos\theta_{\rm res}} \qquad (3)$$

where $n_{\rm res}$ is the refractive index of the photoresist layer and $\theta_{\rm res}$ the angle of incidence in the resist. As a result of this vertical pattern, the posts will have a rippled sidewall, which makes the lift-off process more effective.

3. Experimental set-up

We built two well-known exposure systems. The easiest system is known as 'Lloyd's mirror configuration' (see figure 5) [10].

Part of an incoming plane wave reflects on the mirror and interferes with the undisturbed part of the wave to form an interference pattern (grating) on the substrate surface. To produce the plane wave, TE polarized light of an argon laser with wavelength of $\lambda = 351.1$ nm is spatially filtered and expanded by focusing it onto a pinhole. For large θ (small fringe periods) the system works satisfactorily, but for small θ the image of the mirror on the substrate becomes so small that most of the substrate is not exposed to the interference pattern. An increase in mirror size is expensive, as the demands on smoothness and flatness are high. However, the set-up is very useful for research purposes, as it is simple and θ can be changed easily by tilting the substrate holder.

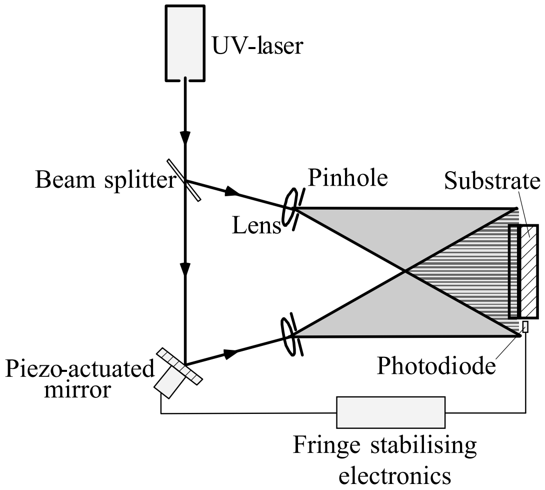

Figure 6. Interference lithography set-up for the exposure of large areas.

For the exposure of large surfaces, we built a second set-up (see figure 6). In this set-up the laser beam is split, after which both beams are expanded separately.

As both beams travel along separate paths, the effects of vibration and air turbulence can easily disturb them. Therefore, a fringe-locking system is necessary. Fringes are detected by a photodiode, after which the signal is used by fringe stabilizing electronics to actuate a piezo element. Using a similar set-up, Spallas *et al* [11] reported the fabrication of uniform photoresist posts on a 50×50 cm^2 glass substrate. The set-up is less sensitive to dust particles than Lloyd's mirror configuration, as there are no mirrors after filtering of the beams. However, it is time consuming to change θ, because this requires movement of a spatial filter after which the beam has to be aligned on the pinhole again.

4. Fabrication

A silicon wafer was coated with a 0.5 μm thick silicon-rich nitride layer to obtain a low-stress membrane [12]. On top of this a 250 nm thick, positive photoresist layer (1 part Shipley 1805 diluted with 1 part Microposit thinner) was spun at 4000 rotations per minute. Using Lloyd's mirror configuration with $\theta = 20.55°$ a post pattern with period $\Lambda_x = 500$ nm was obtained. Various exposure times led to posts of different diameters. The smallest posts obtained are shown in figure 7. A further increase in exposure time led to posts falling over. The rippled sidewalls show about two periods of the vertical interference pattern in the 250 nm thick layer. Calculation with equation (3) and $n_{\rm res} = 1.7$ gives a period of 106 nm.

After chromium lift-off, the pattern is transferred into the silicon nitride membrane, using plasma etching with a CHF_3/O_2 mixture. Figure 8 shows that the lift-off process works well, even for posts that have fallen over.

The holes in the membrane are significantly larger than the 100 nm holes in the chromium mask. Apparently, an aspect ratio of 5:1 is not possible with our etch recipe. However,

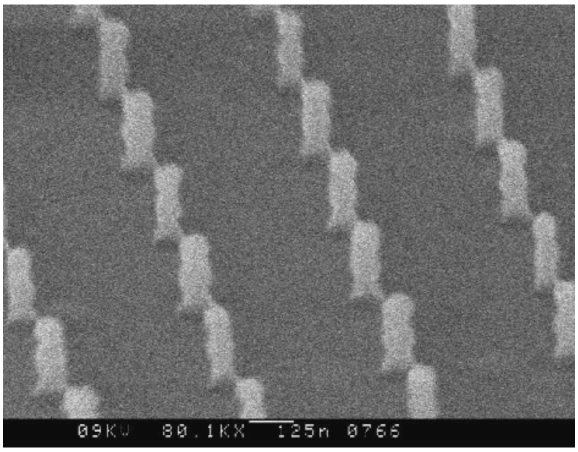

Figure 7. SEM micrograph of 80 nm wide posts with rippled sidewalls caused by the vertical interference pattern.

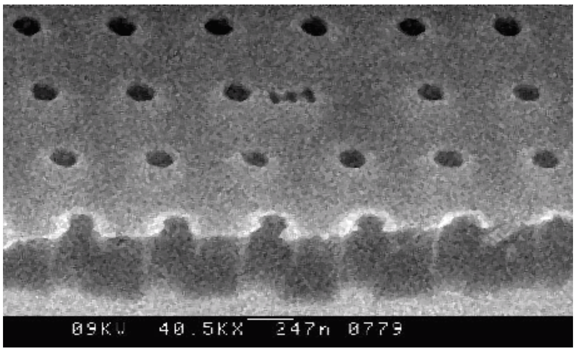

Figure 8. SEM micrograph of membrane after plasma etching through the holes in the chromium layer. The curious hole in the centre is the image of a post that has fallen over prior to the lift-off process. The rippled sidewall of the image is clearly visible.

microsieves are usually made with a pore size larger than the membrane thickness in order to obtain a small flow resistance and a membrane that is easy to clean. Etching with an aspect ratio of 1:1 would be sufficient for such membranes.

In order to make 100 nm pores, a membrane thickness of about 100 nm is needed. If the grating period is set at 200 nm, θ has to be adjusted to 61.37°. Such a large angle gives a higher reflection from the resist/membrane surface. Moreover, the 100 nm thick silicon nitride membrane absorbs less UV light than the 500 nm thick membrane used in figures 7 and 8, which increases the reflection from the membrane/silicon surface. The increased reflection causes a stronger vertical interference pattern, which hinders the formation of posts. The 'waists' of the posts get so thin that the tops fall off. Only the 'foot' formed by the first intensity minimum near the surface remains. Figure 9 shows that the lift-off process on such a foot is problematic.

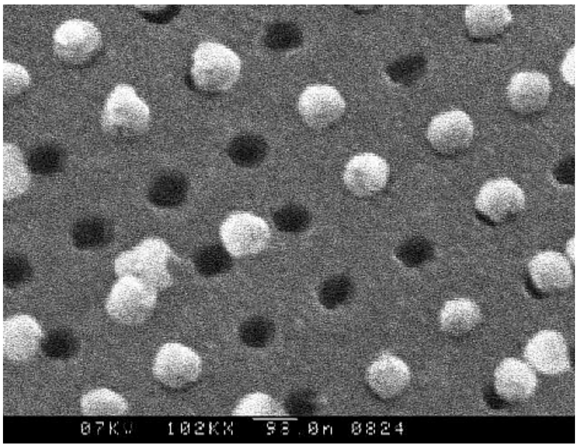

Figure 9. Unsuccessful lift-off due to badly formed posts caused by reflection of the UV light on a 120 nm thick membrane.

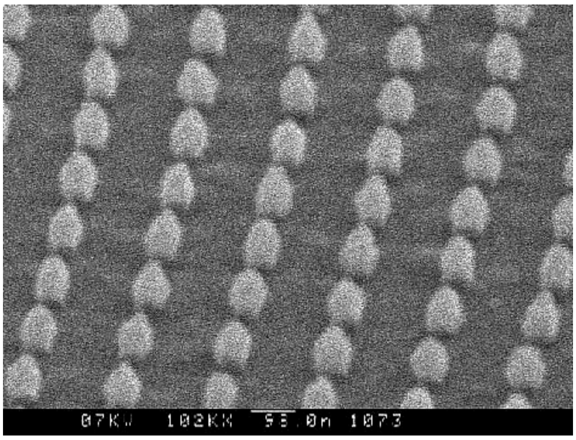

Figure 10. SEM micrograph of posts created on a membrane with an anti-reflection thickness (100 nm).

The intensity minimum near the resist/membrane surface is caused by the 180° phase shift that arises during reflection on this surface. The incoming and reflected beams are always in anti-phase at the surface. In order to reduce the total reflection, the reflected light from the silicon/membrane surface should have the opposite phase of the light reflected from the resist/membrane surface. Reflection measurements show that this is the case for a membrane thickness around 100 nm. We covered such a membrane with a 160 nm thick photoresist layer (1:2 resist/thinner mixture at 5000 rotations per minute) and repeated the exposure conditions used in figure 9. The SEM micrograph in figure 10 shows that the resulting posts have a relatively thin foot, which implies that there has been an intensity *maximum* near the surface. The lift-off process on such small-footed posts is fairly easy. The vertical period for the posts in figure 10 is 121 nm according to equation (3). As the resist thickness is only 160 nm, the posts do not show the ripple as seen in figure 7.

After lift-off and plasma etching a regularly perforated membrane is obtained. The diameter of the pores in the chromium is approximately 65 nm, but the pores in silicon nitride membrane are approximately 100 nm wide due to underetching. Depending on the application the chromium layer can be left on the surface or be removed by wet etching. An SEM micrograph of the etched membrane with the chromium layer on the surface is shown in figure 11.

With our double-beam set-up we successfully fabricated microsieves using 3-inch wafers. A quality check shows that sometimes a pore is missing due to removal of a post during the development process. For filtration purposes this is not a problem. Dust particles form a more serious problem. During exposure a dust particle keeps part of the resist in its shade.

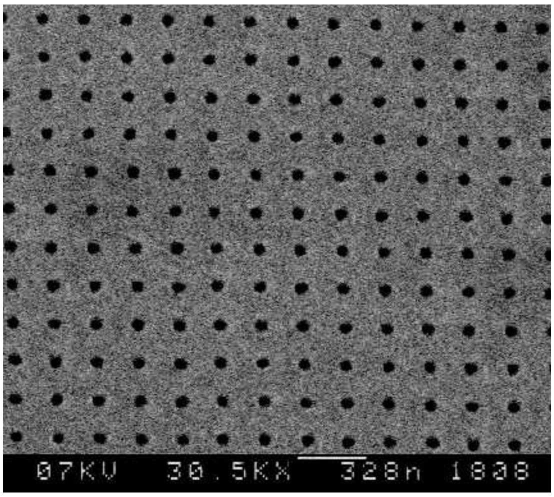

Figure 11. Overview of the pore pattern in an etched membrane. The chromium layer has not been removed.

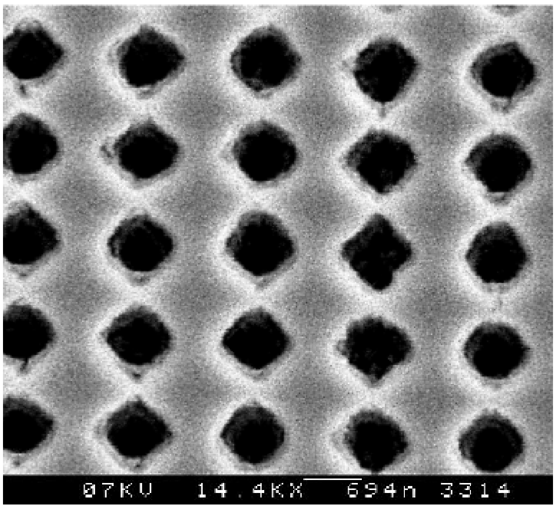

Figure 12. SEM micrograph of a high-porosity microsieve made with the lift-off method. The small process latitude causes irregular results.

This part will not be removed during development and might form a large hole after lift-off. However, a successful lift-off on such a spot is unlikely, as it will probably not have suitable sidewalls (i.e. with a vertical or negative slope). This is confirmed by the fact that we could not find any pores larger than the desired pore size, despite the fact the exposure was not carried out in a clean room environment. The homogeneity of the pore diameter over a larger area is dependent on the solid angle of the expanded laser beam(s) and the distance of the substrate to the pinhole.

The method of laser interference lithography is less suitable for the production of microsieves with high porosities. Creation of large pores gives irregular results due to the rapid transition from holes to posts. An example of an attempt to produce a high-porosity membrane with the lift-off process is given in figure 12. This SEM micrograph shows the irregular square pores that arise after plasma etching.

5. Conclusions

Microsieves with pore sizes down to 65 nm were fabricated using double-exposure laser interference lithography. The pores are obtained with an inverse process, as the direct process of hole formation in photoresist has narrow process latitude. An array of posts is transferred into an array of holes by evaporating chromium onto the posts, followed by a lift-off in acetone. The resulting patterned chromium layer is used as an etch mask for plasma etching of the silicon nitride membrane. The method is fairly robust, as the lift-off process exploits the rippled sidewalls of the posts to allow the acetone to dissolve the photoresist. Underexposed areas (underneath dust particles) do not have such rippled sidewalls and will therefore in most cases not lead to a large hole. And if a post is removed during development due to a poor adhesion, this will only lead to the absence of a pore.

Acknowledgment

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