Holographic lithography for continuous-relief diffractive optical elements

1 Introduction

Diffractive optical elements (DOEs) offer new design flexibility for classical optics as well as for integrated microoptical systems. In these elements, light is diffracted at laterally periodic structures. Their potential lies in the fact that any phase profile can be fabricated as a DOE [1]. Economically advantageous is the ability to fabricate compact and lightweight optics by a low cost replication technique like hot embossing. Therefore, a master of the element needs to be fabricated using the methods of microtechnology, i.e. micro-lithography and etching techniques.

As the efficiency of DOEs increases with the number of phase levels, i.e. surface steps, the fabrication of elements with multilevel or continuous-relief profiles is desirable. Multi-level diffractive structures require multi-mask processing, which implies significant effort in aligning the masks accurately. Alternatively, grey-scale illumination can be applied for the fabrication of continuous phase profile elements, as it is possible in holographic lithography through interferometric illumination. In this maskless approach, the superposition of at least two coherent wavefronts (object and reference) leads to an intensity pattern, which is used for the illumination of a photoresist layer. Thus, the continuous phase profiles can be fabricated with a single exposure.

The two main challenges of this approach are the generation of an arbitrary grey-scale illumination pattern and the precise control of the resist response to that illumination. The main focus of our work lied in the investigation of the performance of the positive resist “Allresist SX AR-P 3500/6” [2] under different processing conditions, i.e. the pre-bake and development. We investigated the response curves of the resist-developer-system and evaluated its suitability for analog lithography.
2 Positive Photoresists

The resist under investigation is a so called DQN positive photoresist. This type of resist basically is composed of three components: a base resin, a photoactive compound (commonly called inhibitor) and solvents. The base resin is soluble in aqueous alkaline developers but the presence of the photoactive compound strongly inhibits the dissolution. The exposure of the resist to light of a specific spectrum converts the inhibitor into a carboxylic acid and thus increases locally the solubility of the film. This photochemical reaction can quantitatively be expressed using the change in the local inhibitor concentration $M$ which is the fraction of inhibitor remaining after exposure relative to that before [3]. The development rate $R$ (thickness of removed film per time) is only a function of $M$ so that the determination of the $R(M)$-curve is essential.

$M$ is dependent on the exposure energy $E$ but not necessarily proportional. A proportionality can be assumed and it is sufficient to work with the $R(E)$-relation if the following effects are negligible:

- multiple reflections at the interfaces and
- bleaching of the resist during exposure.

The requirements can be fulfilled working with glass substrates and by index matching between substrate and resist. Furthermore, the wavelength of the illumination source has to be in the visible blue-violet. In such cases, the absorption is very poor and the irradiance is low, requiring long exposure times to saturate the material. Thus, changes in the intensity pattern during exposure because of bleaching can be neglected and the total exposure energy can be found directly by multiplying the irradiance by the exposure time [4].

The $R(E)$-curve represents the response of the photoresist film to uniform light exposure. This curve can be experimentally measured [5] or calculated using Mack’s model [6].

3 From grey-scale illumination to the resist profile

When a grey-scale illumination is applied to a resist layer, the developed surface profile is not a linear projection of the intensity pattern. As it is depicted in Fig. 1 the resulting profile depends on the shape of the $R(E)$-curve. It defines the resist performance and its characteristics depend in addition to the exposure wavelength and the energy regime, on both the resist processing and composition. Hereby the processing parameters provide the best opportunity to influence actively the resist performance if the wavelength
and the energy regime are given by the illumination source. A typical flowchart for the resist processing is shown in Fig. 2. The critical steps are the prebake, which is a temperature step immediately after coating the wafer with resist, and the development.

Fig. 1: Schematic description of the process of profile forming.

Fig. 2: Flowchart for the resist processing.
4  Suitability for the recording of continuous-relief structures

A resist, which should be suitable to form analog profiles, has to meet the following demands [3]:

- no surface inhibition,
- no dark development,
- no bleaching, and
- a linear $R(E)$-curve.

4.1 Surface inhibition and dark development

Surface inhibition means that the surface of the resist is inhibited in its solubility so that the bulk regions develop faster than the surface itself. Figure 3 shows this effect of underetching.

![Fig. 3: SEM photograph of a developed resist layer. Parts of the resist surface have remained and the effect of underetching due to surface inhibition can be seen.](image)

Dark development occurs if the photoactive compound is partly destroyed or converted without any exposure. It must be noticed that the development rate of the unexposed resist never equals zero, but normally it is negligibly small compared to the rate of exposed regions.

4.2 Bleaching

The term “bleaching” dates from the pioneering work of Dill et al. [5]. Bleaching means the decreasing absorbance of a DQN-resist layer during exposure due to the conversion of photoactive compound into carboxylic acid. The absorption coefficient $\alpha$ of a DQN-resist depends on the so called Dill’s $A$ and $B$ parameters and the inhibitor concentration $M$.

$$\alpha = AM(x,t) + B \quad \text{eq. 1}$$
As $M$ decreases from a starting value of 1 during exposure, $\alpha$ decreases correspondingly. A transmission measurement with a simple optical setup [5] can be used to determine Dill’s parameters and estimate the bleaching behavior.

5 Experimental results

We investigated the resist “Allresist SX AR-P 3500/6” with respect to its suitability for the recording of continuous-relief profiles. We used the developer “Allresist AR 300-26”. Neither surface inhibition nor dark development were observed during the experiments. To evaluate the bleaching behavior we determined the Dill’s $A$ and $B$ parameter. We found out that for this resist $B$ is approximately one order of magnitude greater than $A$ so that the absorption coefficient is primarily dependant on $B$ and does not change significantly over time. Furthermore, the exposure steps were carried out at room temperature employing an Ar ion laser (maximal power 25 mW, adjustable) running at a wavelength of 457.9 nm (visible blue). Thus due to long illumination times and the use of glass substrates bleaching and multiple reflections are negligible and it was sufficient to determine the $R(E)$-curves instead of the $R(M)$-curves.

5.1 Resist characterization: $R(E)$-curves

Our work then focused on the mapping of the $R(E)$-curve under different prebake und development conditions. Therefore, we illuminated resist layers of about 4.2 µm thickness with an uniform intensity over different time segments resulting in different exposure energies. After developing the wafers by immersion for 30 s we measured the removed resist thickness with a profilometer. The determined curves are shown in Fig. 4 and Fig. 5.

As one can see from the $R(E)$-curves a threshold energy must be exceeded before a photochemical reaction occurs. If this energy is not reached at all positions of the exposure pattern, plateaus of undeveloped resist are formed. Furthermore, it is essential to work in the linear range of the $R(E)$-curve to generate analog profiles. Compared to standard photoresists optimized for the recording of binary structures the dissolution rate of the evaluated resist system raises more slowly with increasing exposure energy. Thus, the energy range where a linear $R(E)$-curve can be assumed is sufficiently broad for a precise control of the surface profile.
5.2 Recording of analog phase gratings

Based on the $R(E)$-curves we chose the processing and illumination parameters to transfer first analog structures into the resist (prebake: 20 min, 105°C, convection oven;...
developer concentration: 2:1. We generated a sinusoidal intensity pattern using a Michelson Interferometer (Fig. 7). Hereby the intensity pattern of fringes is generated by the coherent superposition of two plane wave fronts. Its intensity distribution \( I \) can be calculated with respect to the local phase difference \( \delta \) using the following equation:

\[
I = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos \delta.
\]  
\textbf{eq. 2}

Through modification of the ratio \( I_1/I_2 \) of the intensities in both arms of the interferometer the maximal and minimal intensity of the fringes (\( I_{\text{max}} \) and \( I_{\text{min}} \)) can be set. If \( I_1 \) equals \( I_2 \), the highest contrast is reached (see Fig. 6 and Tab. 1).

**Fig. 6: The variation of fringes with respect to the ratio \( I_1/I_2 \).**

**Fig. 7: Michelson interferometer.**

<table>
<thead>
<tr>
<th>( I_1/I_2 )</th>
<th>( I_{\text{max}} )</th>
<th>( I_{\text{min}} )</th>
</tr>
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<tbody>
<tr>
<td>1</td>
<td>4( I_1 )</td>
<td>0</td>
</tr>
<tr>
<td>0.5</td>
<td>2.91( I_1 )</td>
<td>0.09( I_1 )</td>
</tr>
<tr>
<td>0.1</td>
<td>1.73( I_1 )</td>
<td>0.47( I_1 )</td>
</tr>
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For the given experimental setup, there are two different methods to assure the threshold energy to be exceeded:

1. contrast reduction so that the minimal intensity in the fringes increases and the threshold energy can be exceeded on the entire surface
2. exposure in two steps: background exposure to exceed the threshold followed by the pattern exposure

We applied both methods to transfer sinusoidal profiles into the resist. By choosing the illumination parameters, we succeeded to work in the linear range of the curve. The profilometer scan of a resulting profile is shown in Fig. 8.

Finally, we investigated the diffraction behavior of the recorded gratings. In Fig. 9 one can see the highly symmetrical distribution of optical power within the diffraction orders of the sinusoidal phase grating that mirrors its symmetric shape.
6 Conclusions

We showed that the photoresist “SX AR-P 3500/6” is suitable to generate continuous-relief profiles. It meets the required demands if working in a linear range of the $R(E)$-curves is achieved.

We investigated the influence of the prebake and development conditions on the $R(E)$-curves. Increasing developer concentrations primarily shift the threshold energy towards lower values. The prebake mode mainly changes the slope of the curves. An overview how the different effects influence the curve is given in Fig. 10.
Based on the determined $R(E)$-curves we chose the processing and illumination parameters and succeeded in the first recording of sinusoidal resist profiles. Future works focus on recording arbitrary intensity patterns for the fabrication of diffractive optical elements. Therefore, a previously generated grey-scale pattern, e.g. a computer generated hologram (CGH), will be transformed into a phase profile requiring only a single analog photoresist processing step.

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References:


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