Sub-50 nm patterning by immersion interference lithography using a Littrow prism as a Lloyd’s interferometer

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We present a simple setup that combines immersion lithography with a Lloyd’s mirror interferometer. Aiming for smaller structure sizes, we have replaced the usual Lloyd’s interferometer by a triangular Littrow prism with one metal-coated side, which acts as a mirror. Because of the higher refractive index of the prism, the wavelength and, thus, the attainable structure sizes, are decreased significantly. Using a laser with a wavelength of 244 nm, we could produce line patterns with a period of less than 100 nm and a width of 45 nm. The introduced setup retains all the advantages of a Lloyd’s mirror interferometer, in particular the flexibility in periodicity. © 2010 Optical Society of America

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Laser interference lithography (LIL) is a powerful tool that is frequently used for the fabrication of nanostructures [1]. Simple line gratings, but also more complex two- or even three-dimensional photosensitive patterns, can be created [2,3]. Laser interference lithography is an attractive method to reproducibly and efficiently produce large-area periodic patterns [4]. These can then be used, e.g., as templates for nanowire growth [5], nanowire etching [6,7], or solar cell optimization [8].

Smaller structures offer the opportunity to investigate new physical phenomena and allow for the fabrication of devices with a higher packing density [1]. The structure size obtainable by laser interference lithography is proportional to the wavelength of the laser but also depends on the refractive index of the surrounding medium. Interference in a prism instead of in air reduces the obtainable structure size by the refractive index of the prism.

Using different setups [9–13], immersion interference lithography has been used to produce patterns with periodicities below 50 nm. However, such setups are designed to produce patterns with a certain, fixed periodicity. Thus, to vary the periodicity, optical components have to be exchanged and/or the optical setup has to be realigned, which can be delicate and time consuming. In contrast, changing the pattern periodicity in a Lloyd’s mirror interferometer setup is fast and simple and requires no realignment. In this Letter, we combine the principle of a Lloyd’s mirror interferometer with immersion lithography by using a Littrow prism as a Lloyd’s interferometer and water as the immersion liquid to prevent total internal reflection. As for a standard Lloyd’s interferometer, the periodicity of the obtained patterns can be varied by adjusting the angle between incident laser light and the sample. Using a laser with a wavelength of 244 nm and a prism with a refractive index of ~1.5, patterns with a periodicity below 100 nm and a width of 45 nm were fabricated.

A standard Lloyd’s mirror interferometer is shown in Fig. 1(a). It consists of a sample holder and a mirror perpendicular to the sample. The direct incident light and the light reflected from the mirror reach the sample at the same angle, interfere, and create a sinusoidal intensity distribution across the sample. The distance between the intensity maxima, the pitch or periodicity \( p_{\text{LIL}} \), is given by [14]

\[
p_{\text{LIL}} = \frac{\lambda}{2n_{\text{air}} \sin \theta_{\text{air}}}, \tag{1}
\]

where \( n_{\text{air}} \) is the refractive index of air and \( \theta_{\text{air}} \) is the angle (in air) between the incoming laser light and the sample normal. Because of Snell’s law \( \eta_{\text{PR}} \sin \theta_{\text{PR}} = n_{\text{air}} \sin \theta_{\text{air}} \), refraction at the air/photoresist interface does not change the periodicity. For evaluation of Eq. (1), one can therefore use the refractive index of air and the angle (in air) between the laser and the substrate normal. Both are experimentally easily accessible. Experimentally, \( p_{\text{LIL}} \) can easily be varied by rotating the Lloyd’s interferometer before exposure.

In Fig. 1(b) we present an immersion Lloyd’s mirror interferometer, composed of a Littrow prism and water as the immersion liquid. The expanded laser beam is directed onto the prism, which has a right-angled base and is positioned with its short side parallel to the sample. The side perpendicular to the sample has a metal coating and replaces the mirror of the standard setup. To ensure optimal contrast [1,14], the beam should be centered at the angle between the mirror and the short side of the prism. The resulting periodicity in the immersion LIL (ILIL) setup shown in Fig. 1(b) is reduced due to the refraction of the light in the prism and given by

\[
p_{\text{ILIL}} = \frac{\lambda}{2n_{\text{prism}} \sin \theta}, \tag{2}
\]

and, thus, it is a factor \( n_{\text{prism}}/n_{\text{air}} \) smaller than in the standard setup shown in Fig. 1(a). 30°–60°–90° prisms, so-called Littrow prisms (22 mm × 37.9 mm × 22 mm), made from fused silica with an Al coating (\( \lambda/10 \) quality) on the long leg were purchased from Altechna Co. Ltd., Lithuania. These Littrow prisms, used, e.g., for wavelength selection in laser cavities, are standard components and are, therefore, inexpensive (~$100). Note that the prism does not have to be a 30°–60°–90° prism; it just has to be
rectangular. The chosen prism has the advantage, however, that it minimizes undesired reflection at the prism/air interface for large \( \theta \) and \( \theta' \), and, thus, for small periodicities.

Total internal reflection occurs at the prism/air interface for \( \sin \theta = n_{\text{air}}/n_{\text{prism}} \). Because we are interested in small periodicities, i.e., large \( \theta \), this is highly undesirable, but can potentially be circumvented if a substance with a higher refractive index is situated between photoresist and prism. We used deionized water as the immersion liquid to decrease reflection at the short side of the prism. While the short side of the prism was rinsed with deionized water, the samples were carefully pressed against the prism. Because of surface tension, a water film wets the prism and the sample, separating them.

The laser used for illumination is a frequency-doubled argon-ion laser with \( \lambda = 244 \) nm (85-SHG, Cambridge Lasers, USA) and a typical output power of 3 mW. The laser is used in the TEM\(_{00}\) mode and the light is TE polarized. TE polarization is desired for most applications, as it ensures optimal contrast \([10,15]\). The light is directed into a spatial filter, consisting of a focusing lens and a 10 \( \mu \)m diameter pinhole. The distance between the spatial filter and the sample holder is around 1 m. The resist requires an exposure dose of \( \sim 50 \) mJ cm\(^{-2}\) and, with typical exposure times of 2 to 3 min, the intensity at the resist is of the order of 2 Wm\(^{-2}\). The refractive index of the prism at 244 nm is calculated from the Sellmeier equation \([16]\) to \( n_{\text{prism}} = 1.51 \), while the refractive index of water is \( n_{\text{H}_2\text{O}} = 1.38 \) \([17]\); thus the critical angle for total internal reflection is \( \theta_{\text{crit}} \approx 66^\circ \). It is possible to use liquids with higher refractive index \([12,17]\) to increase the critical angle further. We did not pursue this approach mainly because the reduction in periodicity is only very moderate for angles \( \theta \) larger than \( 65^\circ \).

The angle \( \theta \) in Eq. \((2)\) is the angle between substrate normal and laser light inside the prism, which is not directly experimentally accessible. For our geometry, Snell’s law can be rewritten as

\[
\frac{\sin(60^\circ - \theta')}{\sin(60^\circ - \theta)} = \frac{n_{\text{Prism}}}{n_{\text{air}}},
\]

where \( \theta' \) is the angle in air, which is easily controlled experimentally.

A negative tone diazonaphthoquinone-based resist (AR-N 4240, mixed with diluter AR 300-12 at ratio 1:4) was spin coated onto cleaned silicon wafers at 4000 rpm (30 s), with adhesion improved by primer AR 300-80 (2000 rpm, 30 s). This was followed by a prebake at 85 °C for 2 min. After these steps, the photoresist thickness was 100 nm. After exposure, the sample was post-baked at 85 °C for 30 min and developed in AR 300-475 for 30 s. All photochemicals were obtained from Allresist GmbH.

Figure 2(a) shows a large-area SEM image of the photoresist after exposure and development. The line pattern is clearly visible and is regular and uniform. From the higher magnification image shown in Fig. 2(b), a periodicity of 90 ± 2 nm can be deduced. This fits well to the expected result of Eq. \((2)\): \( \rho_{\text{LIL}} = 93 ± 4 \) nm at \( \theta' = \theta = 60^\circ \). The width of the resist stripes is 43 ± 4 nm and shows some roughness, presumably because a standard photoresist with a spatial resolution of \( \sim 50 \) nm was used. Depending on optical constants and angles, a fraction of the laser light is reflected from the resist–substrate interface. Using an antireflection coating beneath the resist layer could suppress these reflections and, thereby, further improve the quality of the resist profile \([14]\). Coherence loss could, in principle, also be a reason for poor pattern quality, as one requirement for good imaging in a Lloyd’s mirror interferometer is good temporal and spatial coherence. However, the temporal coherence length of the employed

![Fig. 1](image1.png)

![Fig. 2](image2.png)
The laser is ~10 m and, thus, orders of magnitude larger than typical beam path differences in the setup. Regarding the spatial coherence, we can conservatively estimate from the Van Cittert–Zernike theorem [18] that the pattern quality should not be affected significantly for a patterned area width below 5 mm.

Resist patterns obtained by lithography are usually transferred into other materials, e.g., by etching or by vaporization and lift-off. Figure 2(c) shows a metal stripe pattern after evaporation of 15 nm Ag and subsequent lift-off with acetone. This yielded a metal stripe pattern with a stripe width of 47 ± 6 nm.

To prove the versatility of the proposed setup, we have conducted exposures at different θ. The resulting periodicities, shown in Fig. 3, were deduced from SEM images. The solid curve represents the theoretical curve according to Eq. (2). This can be compared to the periodicity of a standard Lloyd’s mirror interferometer (dashed curve). Structures with (86 ± 4) nm ≤ p ≤ (132 ± 5) nm have been successfully fabricated. The measured periodicities follow the theoretical curve, albeit being systematically about 5% too small. Possible error sources include incorrect measurement of θ, inaccuracies in the analysis of the SEM images, or a refractive index of the prism differing from the assumed n_{prism} = 1.51.

The presented results have been obtained with a laser with λ = 244 nm. The presented immersion Lloyd’s interferometer concept is, of course, applicable to lasers with different, in particular, smaller wavelengths, provided the laser source meets the coherence requirements for the targeted field sizes and throughputs.

In summary, we have shown a simple and inexpensive modification of a Lloyd’s mirror interferometer. Using a laser with λ = 244 nm, a fused silica coupling prism, and water as the immersion liquid to avoid total reflection, line patterns with sub-100 nm periodicity and 45 nm width were fabricated. The proposed setup extends the range of periodicities that are accessible with a Lloyd’s mirror interferometer significantly, while retaining its advantages like ease of alignment and flexibility in periodicity.

References