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# Photolithographic periodic patterning of gold using azobenzene-functionalized polymers

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ABSTRACT

Metal micro- and nanostructures for optical and electronic applications are typically fabricated by means of interferometric optical or electron-beam lithographies using conventional photo- or electron-beam resists. In this work, we report on fabrication of periodic nanostructures of gold by exploiting photoinduced surface-relief gratings in an azobenzene-functionalized polymer film as masks for reactive ion etching of the metal. The proposed technique provides a convenient, fast and flexible alternative to photoresist-based lithography for fabricating metal nanostructures of large surface area. Owing to the fact that the azo-polymer is sensitive to the polarization rather than the intensity modulation of the exposing light, the technique is particularly suitable for patterning highly reflective surfaces.
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## 1. Introduction

Periodic arrays of metallic micro- and nanostructures are widely used in optics and electronics. Among the optical applications we mention diffraction gratings, nanofabricated wire-grid polarizers, spectral filters, plasmonic and photonic waveguides, substrates for surface enhanced fluorescence and Raman spectroscopy, and optical metamaterials. Periodically structured metal films are usually fabricated by means of optical or electron-beam lithography which include creation of a resist pattern and lift-off metallization [1–3]. Electron-beam lithography is able to provide structures with high accuracy, but it is expensive and very time-consuming and, therefore, not quite suitable for fabrication of large-area arrays [3,4]. The same can be said about focused ion-beam milling [4,5].

One of the simplest and fastest fabrication methods that allow creating large-area periodic arrays of micro- and nanostructures is optical interference lithography [6–8]. The achievable dimensions of the structures and the fabrication precision are mostly limited by the illumination wavelength, intensity contrast of the interference pattern and the contrast of the photoresist. Interference lithography is typically used for obtaining periodically patterned metal films by initially creating a photoresist mask and then applying metallization of the sample and subsequent etching of the photoresist lift-off underlayer. Such a lift-off based approach, however, cannot be exploited to pattern bulk-metal surfaces, since for such surfaces the mask can be created only above the metal. In principle, dry etching could be used to pattern the metal through such a mask. However, when using photoresist as a mask material, the highly reflecting metal surface causes a standing-wave intensity modulation of the exposing light to appear across the photoresist thickness even for rather thin 100 nm layers. This makes it difficult to control the final geometry of the mask [6,9]. Still, some reports on successful metal-film patterning by using this approach can be found in the literature [10,11]. To reduce the optical reflection and the resulting formation of the unwanted standing optical waves, a sublayer of a strongly light-absorbing material is often used beneath the photoresist [1,6,8]. The introduction of this layer, however, complicates the fabrication process and leads to difficulties in obtaining accurate nanopatterns.

In this work, we introduce a very simple approach to fabricate periodically nanostructured metal films and bulk metal surfaces by means of optical interference lithography using a technique, in which the photoresist is replaced with an azobenzene-functionalized polymer (azo-polymer) film [12–14]. In contrast to photoresists, this material is sensitive to *light polarization modulation*. In particular, it can be processed under ordinary lighting conditions without any degradation of its properties. When two interfering p-polarized (or counter-circularly polarized) optical waves are incident on the azo-polymer film, the resulting periodic modulation of the field polarization causes the material in the film to macroscopically move under the interference pattern and form a surface relief grating (SRG) [12]. We have observed that the SRG is formed equally well on transparent, absorbing and reflecting substrates, owing to the polarization sensitivity of the material. Therefore, such an SRG can be used as a mask also for etching metals without need in a light-absorbing sublayer. In addition, neither wet development nor pre- and post-baking of the mask are required. The mask will stay unchanged for months, if not for years, under ordinary storage conditions even in ambient light. Moreover, if one would need to further pattern the mask, it will be possible to do so

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by exposing the film once again to interfering laser light. We also emphasize that azo-polymers are sensitive to a wider spectral range of the exposing light and are more tolerant to overexposure than conventional photoresists [13,15,16]. The latter property makes it easy to obtain more sophisticated mask patterns by applying several independent exposures onto the same azo-polymer film.

In the following, we begin by presenting a theoretical explanation of the concept of using azo-polymer films instead of photoresist (Section 2). Then we introduce our fabrication technique (Section 3). In Section 4, we give some examples of particular applications of the technique and, in Section 5, summarize our results.

## 2. Theoretical consideration of the exposure conditions

We start by calculating the intensity and polarization distributions of light in an azo-polymer film on gold. To match the experimental conditions, we assume that an SRG is inscribed in an azo-polymer film with two interfering p-polarized waves and that the incident angles of the waves are equal to $\theta = \pm 35$ deg. The wavelength of the light ($\lambda$) is 488 nm. At this wavelength, the complex-valued refractive index of the polymer is $n_{\mathrm{pol}} = 1.78 + 0.20i$ and that of gold is $n_{\mathrm{Au}} = 1.14 + 1.84i$ [17–19]. For a 110 nm thick layer of the polymer on the surface of gold, the calculated intensity distribution of the interference fringes is shown in Fig. 1(a). The calculations were done by using the standard electromagnetic boundary conditions [20]. The fringes have a period of 430 nm along the film. The field intensity is seen to be significantly modulated also along the surface normal. In addition, the in-plane intensity modulation considerably decreases towards the material's interfaces with air ($z = 0$) and gold ($z = 110$ nm). The $z$-dependences of the intensity maximum and minimum in the pattern are shown by the red and blue curves, respectively, in Fig. 1(b). The uneven intensity modulation inside the film could make the fabrication of a desired etching mask a challenging task, if the exposure of the film would follow the intensity distribution, as in the case of an ordinary photoresist.

As has been mentioned before, SRGs are formed in an azo-polymer layer under the action of a polarization interference pattern. The material usually escapes from the regions where the light is polarized perpendicularly to the polarization interference fringes [12]. Fig. 1(c) shows the amplitude distribution of the x-polarized component ($E_x$) in the pattern of Fig. 1(a). It is seen that between the fringes the value of $E_x$ reaches exact zero throughout the whole thickness of the film. During the exposure, the polymer will move towards the locations of low $|E_x|$ and an SRG will be formed. The $z$-polarized component, depicted in Fig. 1(d), does not essentially contribute to the mass transport of the polymer along the surface. This component, however, is the one that makes the intensity minima across the film thickness in the pattern of Fig. 1(a) deviate from zero.

While in the experiments performed in this work the incident angles of the waves were kept at $\theta \approx \pm 35$ deg, it is of interest to

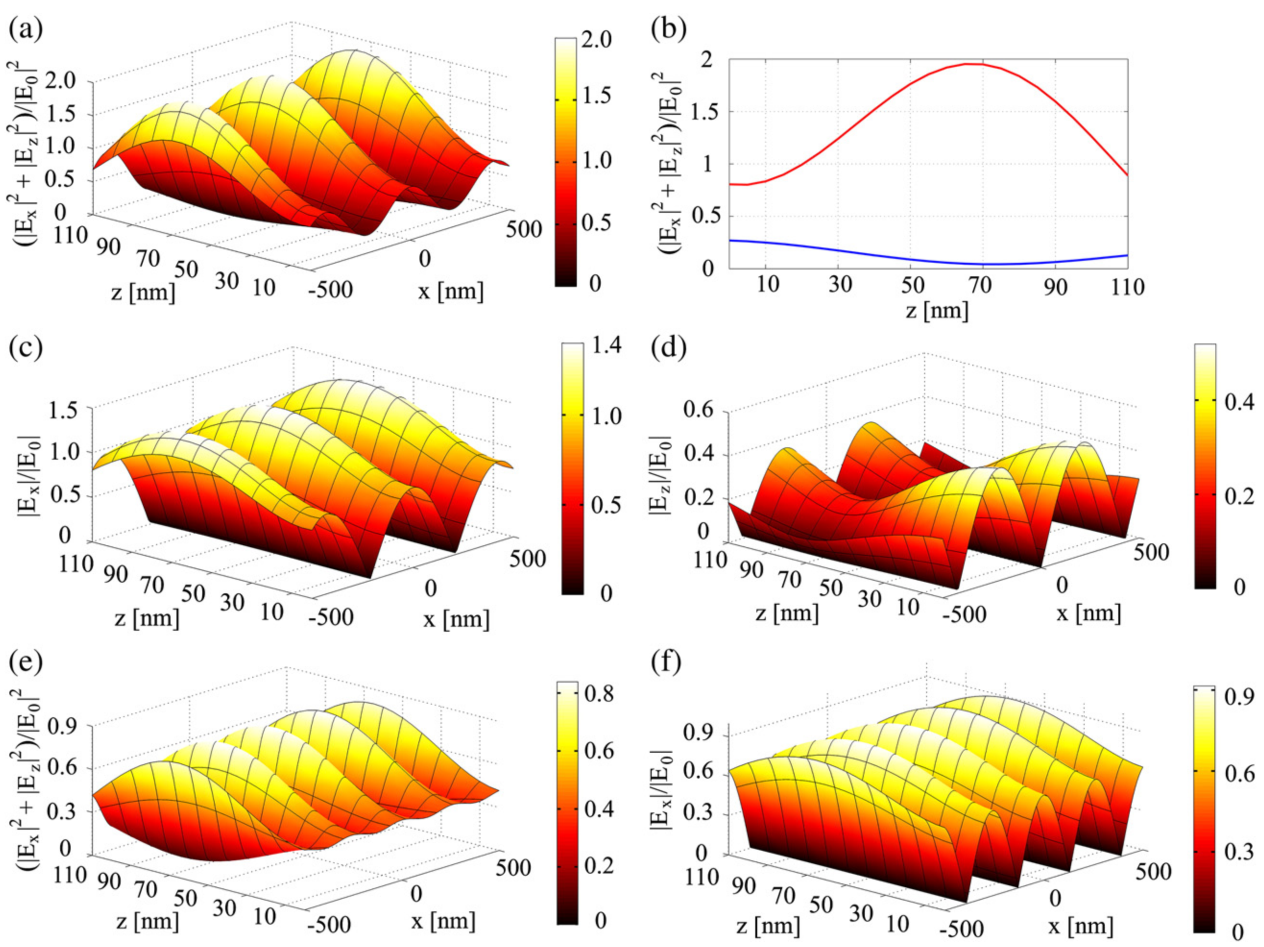


**Fig. 1.** Intensity and amplitude distributions in a 110 nm thick azo-polymer film on gold under illumination by two optical plane waves with amplitudes $E_0$ [$\lambda = 488$ nm; $\theta = \pm 35°$ for figures (a) – (d) and $\theta = \pm 70°$ for figures (e) and (f)]. (a) Intensity distribution of the interference fringes. (b) Intensity as a function of distance $z$ from the film surface along a fringe (blue) and in between two neighboring fringes (red). Cases (c) and (d) show the amplitude distributions of the $x$- and $z$-polarized components of the laser light, respectively. The plots in (c) and (d) correspond to the plot in (a). Case (e) illustrates the intensity distribution at $\theta = \pm 70°$ and (f) shows the corresponding amplitude distribution of the $x$-polarized field component.

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