Resonant metamaterials for contrast enhancement in optical lithography

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Abstract: The transmission through ultra-thin metal films is noticeable and thus limits their potential for the formation of lithographic masks. By sub-wavelength patterning of a metal film with a post structure, a resonant metamaterial is formed, which can effectively suppress the transmission. Measurements as well as calculations identify the width of the metal islands as a critical geometrical feature. Hence, the extraordinarily low transmission effect can be explained by the resonant response of single scatterers known as Localized Surface Plasmon Resonances (LSPR). A potential application of this suppressed transmission effect to thin metal masks in optical lithography is experimentally investigated.

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1. Introduction

Optical lithography is constantly being challenged by decreasing structure sizes, requiring improved fabrication procedures and masks. The utilization of thin metal masks allows for downsizing the lateral structure dimensions by keeping up the low aspect ratio which guarantees a sufficient mechanical stability of the absorber patterns. Furthermore, these thin absorbers provide better performance for off-axis illumination, which is frequently used in advanced lithographic systems [1]. However, this approach results in higher transmission through the nominally intransparent areas of the mask, and therefore a decrease of contrast and resolution is inevitable. In this context, suppressed transmission by subwavelength patterning can open new prospects. The inverse effect of extraordinarily high transmission in periodic sub-wavelength structures - pioneered by Ebbesen [2] - has already drawn a lot of attention as well as stimulated application proposals [3, 4]. Inspired by this work, also the optical response of single building blocks has been investigated [5, 6]. Recently, the effect of reduced transmission of 2D structured ultra-thin metal films has been reported theoretically [7, 8] and experimentally [9, 10]. It is generally agreed that the effect is based on plasmonic excitations. In an earlier publication [8] theoretical simulations predict that the extraordinarily low transmission of the considered ultra-thin metamaterial consisting of post-like building blocks merely depends on the width $w$ of the posts (see Fig. 1). In contrast, the position of the resonance is only marginally dependent on the pitch $p$ of the structure, which is known to play an important role for the excitation of propagating surface plasmons [2]. Hence, it is more likely that a localized surface plasmon (LSP) is excited in each individual post, thus acting as an antenna [11]. This statement is consistent with findings for sparse gold post structures with constant fill factor $= w/p = 0.5$ located on a 150 nm thick ITO (indium tin oxide) layer [12].

In this paper we report on the fabrication and investigation of a structure with suppressed transmission consisting of an ensemble of resonant nanostructures. We investigate how far its optical response can be treated as that of an ensemble of isolated resonant dipoles and whether such structures can form building blocks of lithographic masks. On the basis of a simplified model we furthermore discuss the theoretical limits of transmission suppression by a metamaterial of this kind. By evaluating angular dependent transmission data we quantify the weak coupling between the posts and discuss the resonance splitting and the appearance of new modes for oblique incidence.

2. Fabrication and measurement setup

Following the design proposed in [8] ultra-thin Ag layers were deposited on a BK7 substrate via magnetron sputtering. First, a 1 - 2 nm adhesion layer of chromium (Cr) was deposited before a Ag layer of 30 nm was sputtered. The samples were then patterned via Focused Ion Beam (FIB) milling with various geometrical parameter combinations of width $w$ and pitch $p$. The structure design is schematically shown in Fig. 1, together with a Scanning Electron Microscope (SEM) image of a representative sample.
Measurements were performed with a homebuilt scanning setup [13] illustrated in Fig. 1. We used a commercial tunable light source (Koheras SuperK Extreme with a SpectraK Dual spectral filter). The laser light is polarized along one of the axes of the structure geometry (x- or y-polarized). Due to the fabrication method of FIB milling, the total sizes of the samples are limited to 5 μm × 5 μm. Thus, the incoming light is focused on the sample by an objective (NA = 0.9). To minimize the angular divergence of the beam and to allow for a comparison with plane-wave simulations the back aperture of the objective was not completely illuminated thus realizing an effective numerical aperture of NA_{eff,FWHM} = 0.28 corresponding to a focal spot with a diameter of approximately 1 μm for λ = 633 nm. The transmitted light is collected by a second microscope objective below the sample (oil immersion, NA = 1.3). The reflection can be recorded simultaneously due to the inserted beam splitter in front of the focussing objective. The confocal beam alignment stays fixed during the measurements whereas the sample is x-y-scanned in the focal plane by a 3D piezostage.

3. Measurement results

Measurement results were compared with simulations based on a rigorous coupled wave analysis [8]. To quantify the effect of suppressed transmission we introduce the relative transmission $T_{rel}$ defined by

$$T_{rel} = \frac{T_{Bulk} - T}{T_{Bulk} + T},$$

where $T_{Bulk}$ is the transmission of the unpatterned and $T$ that of the structured film. A $T_{rel}$ value of +1 corresponds to zero transmission through the structured film. This relative transmission was determined for various samples and for a fixed wavelength of 633 nm. In the left part Fig. 2 the experimental results are displayed as color-coded squares on top of theoretically predicted values. These results demonstrate that the transmission can indeed be suppressed in agreement with the results of electromagnetic simulations. As the post shape strongly influences the resonance spectrum, small fluctuations in fabrication as well as rounding of the edges lead to slight discrepancies between experimental and theoretical results. A spectral scan displayed in the right part of Fig. 2 illustrates a broad resonance, which is characterized by a dip in transmission and a corresponding increase of $T_{rel}$. Obviously the suppression of transmission is not only an absorptive feature, but also accompanied by a resonant enhancement of reflection. Both features indicate a LSP resonance. To further clarify, whether suppressed transmission is a collective effect or caused by individual posts we varied the pitch of the periodic structure.
Fig. 2. (a) Comparison of theoretical results and measurements of $T_{\text{rel}}$ calculated with Eq. (1) for a wavelength of 633 nm. The underlying colored plot shows the results of a simulation [8] whereas squares indicate measured values of $T_{\text{rel}}$ (same color code) for a 30 nm thick structure with the respective lateral geometrical parameters (measurement errors $s_{\text{pitch}} = 10$ nm, $s_{\text{width}} = 10$ nm). 
(b) Measured transmission $T$ and reflection $R$ for a post structure and an unstructured Ag film. Assuming scattering into the zeroth diffraction order only, the absorption was calculated as $A=1-T-R$. Also indicated in the plot is the resulting spectral resonance curve of $T_{\text{rel}}$.

Fig. 3. Measured resonance curves for different structures of 30 nm thickness and with (a) constant width and (b) constant pitch for x-polarization. For the perpendicular polarization the resonance positions slightly differ due to fabrication discrepancies, but their dependence on the width also holds.
and the width of the individual posts separately (see Fig. 3). The graphs clearly show a strong
dependence of the resonance position on the width of the metal posts as it would be expected
for an antenna-like interaction. In contrast, the interaction between different elements seems to
play a minor role. Only for small element spacing we observe a change of the spectral shape
of the resonance. Furthermore, the strength of the transmission suppression changes with the
amount of surface coverage.

4. Model

In order to better understand the suppression of transmission by the excitation of localized
dipole antennas, we introduce a highly simplified model. We neglect the influence of the sub-
strate and restrict our model to monochromatic excitation with a plane wave propagating normal
to the structure in z-direction, \( \vec{E}_{\text{in}} e^{ikz} \) with the wavenumber \( k = \omega / c \). As the pitch of the structure is sub-wavelength no diffracted orders appear, and the field \( \vec{E}(z) \) can be described by a
one-dimensional Helmholtz equation as

\[
\left[ \frac{\partial^2}{\partial z^2} + k^2 \right] \vec{E}(z) = -\mu_0 \omega^2 \vec{P}(z),
\]

where \( \vec{P}(z) \) is the polarization density induced in the metamaterial. Since the thickness \( d \) of the
metallic structure is extremely small compared to the wavelength \( (d \ll \lambda) \), we represent the
total response of its polarization \( \vec{P}(z) \) by a delta distribution
\( \vec{P}(z) \approx \vec{P}_0 \delta(z) \). The resulting
electric field

\[
\vec{E}(z) = \vec{E}_{\text{in}} e^{ikz} - \frac{\mu_0 \omega^2 d}{2ik} \vec{P}_0 e^{ik|z|}
\]

consists of the incident light and the radiation emitted by the metamaterial layer. As the
polarization is driven by the incident field, an effective susceptibility can be defined by
\( \vec{P}_0 = \varepsilon_0 \chi_{\text{eff}} \vec{E}_{\text{in}} \). Because this susceptibility is determined by a single resonance of the met-
metallic posts, it can be approximated as

\[
\chi_{\text{eff}}(\omega) \approx \chi_0 \frac{\tau}{\omega - i\tau},
\]

where \( \chi_0 \) defines the strength of the interaction, \( \tau \) the linewidth and \( \omega_0 \) the resonance fre-
quency. At resonance \( \chi_{\text{eff}} \) is purely imaginary, resulting in destructive interference between the
incident and radiated field. Consequently the total field transmitted in forward direction \( \propto e^{ikz} \)
is reduced, whereas the back-reflected field \( \propto e^{-ikz} \) is at its maximum. As energy must be con-
served we find upper limits for the strength of this effective susceptibility. In particular reflec-
tion cannot exceed 1 resulting in vanishing transmission \( T = 0 \). Based on the derived relation
for the total electric field (Eq. (3)) this leads to the following condition at resonance

\[
E_{\text{in}} \geq \frac{\mu_0 \omega_0^2 d}{2ik} \vec{P}_0,
\]

which sets an upper bound to the strength of the susceptibility as

\[
\chi_0 \leq \frac{2}{k(\omega_0)d} = \frac{\lambda}{\pi d}.
\]

An effective susceptibility, which satisfies this upper limit, ensures a total cancellation of trans-
mission at resonance. This can only be reached if the losses of the individual resonant elements
are solely radiative. The amount of residual transmission therefore quantifies the strength of non-radiative decay channels. Interestingly, Eq. (6) does not depend on the properties of the particular metamaterial but defines a general property of optically resonant layers with sub-wavelength thickness. The results displayed in Fig. 3 illustrate that we come close to this limit if the surface coverage is high.

Of course the above model cannot account for the complete optical response as it regards the posts as simple dipoles only and neglects all mutual interaction. In the following we will probe for transverse coupling by studying the response of our samples at oblique incidence. Any spectral shift of the resonance position will indicate collective contributions to the response.

5. Angular dispersion

To probe the angular dispersion of the resonance the optical setup was modified. A long working distance objective was used to focus the light on the tilted sample. The transmission photodiode was fixed directly underneath the sample. The air-gap was filled with index matching fluid. The sample-photodiode system was then rotated relative to the incoming focused beam. In Fig. 4 respective measurement results are presented. For comparison 3D numerical simulations of the post structure with the Finite Difference Frequency Domain (FDFD) method [14] were conducted in a larger parameter range not accessible in the experiment. We find good agreement between the measured and numerically calculated spectra. Deviations in the total value of the relative transmission can be attributed to variations in the width of the posts, the grain size of the sputtered Ag film, and the used Cr adhesion layer. To allow for qualitative comparison of the experimental and theoretical dispersion results the colorcodes for the measured $T_{\text{rel}}$ plot are scaled to the maximum value.

Grating assisted coupling to propagating surface plasmon polaritons is known to be highly dependent on the angle of incidence. In contrast, our calculations and measurements indicate nearly no angular dispersion of the resonance. Also the calculations, which include a larger wavelength range than the measurements indicate no further resonances in the visible and near infrared spectral region. The weak resonance splitting observed for large angles and TE (transversal electric) polarization indicates the occurrence of a new mode excited in addition to a simple electric dipole mode. Nevertheless, the simple model of dipole excitation of the whole post holds true for the TM (transversal magnetic) excitations at all angles. Further calculations indicate electric quadrupole as well as magnetic dipole resonances due to phase differences in excitation on the individual posts in the TE case. In summary, also the angular dependent response together with the results from Fig. 3 clearly demonstrate the dominance of localized excitations in the individual building blocks.

6. Possible application

The model of localized excitations of individual building blocks suggests that already a limited number of metamaterial structures in the vicinity of larger openings improves the contrast of a lithographic mask. As a proof of concept, a series of mask structures (three square openings) was fabricated. According to the design steps shown in Fig. 5, an increasing number of lateral post layers with resonant feature sizes were applied around single mask structures. Figure 5 displays line scans in transmission through the center of the mask features performed with the scanning setup described above at a wavelength of 574 nm. Using a resonant post structure around the square openings with $T_{\text{rel}} = 0.63$ enhances the contrast by a factor of 1.4 by probing with a beam diameter of approximately 1 $\mu$m. Additionally, the posts structures cause no noticeable shape distortions of the mask features, as it was already predicted by numerical simulations in [8]. Already a single post layer improves the scan image. Hence, no large-area metamaterial is required to reduce transmission.
Fig. 4. (a) Schematic drawing of the oblique incidence investigations for an array of posts with pitch $p = 215$ nm and width $w = 160$ nm. (b) The numerical results are illustrated in a dispersion diagram. $T_{rel}$ is plotted versus the wavevector $k = \frac{2\pi}{\lambda}$ and its x-component $k_x = \frac{2\pi}{\lambda}\sin\theta$ in the plane of the posts structure ($\theta = 0^\circ$ to $75^\circ$, $\lambda = 300$ to 1200 nm). The marked area indicates the experimentally accessible parameter range. (c) Oblique incidence measurements of $T_{rel}$ for the corresponding structure ($\theta = 0^\circ$ to $60^\circ$ in steps of $10^\circ$, $\lambda = 470$ to 720 nm). The colorcodes for $T_{rel}$ are scaled relative to the maximum measured values.

So far, the suppressed transmission effect has been investigated in the visible spectral range. Advanced optical lithography tools for semiconductor fabrication operate at a wavelength of 193 nm. The identification of appropriate materials and geometries for this wavelength is very challenging. On the other hand, special lithography techniques for other areas of micro- and nanofabrication such as mask aligner lithography [15], or absorbance modulation photolithography [16] use wavelengths from 300 up to 800 nm. The exploitation of the investigated metamaterials could open new possibilities for the design of lithographic masks for these lithographic techniques.
Fig. 5. Transmission signals measured along cross-sections of mask structures. (left) Schematic drawing of a series of mask structures with an increasing number of posts \((w = 120 \text{ nm}, p = 190 \text{ nm})\) supporting a square opening \(830 \times 830 \text{ nm}^2\). (middle) SEM pictures of mask structures (indicated scale bar: 1 \(\mu\text{m}\)) (right) Transmission signals along cross-sections of three square openings in a row; the corresponding contrast values \(C = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}}\) are indicated in the individual plots.

7. Conclusion

In summary, we have demonstrated that specially designed sub-wavelength structures show suppressed transmission. The measured dependences on geometrical feature sizes and the linked spectral features of transmission, reflection and absorption support the description via a local plasmonic effect. Placed in the vicinity of larger features, for example contact holes, the sub-wavelength arrays are able to enhance the contrast. This scheme could open new possibilities for the design of lithographic masks.

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