How Holes Can Obscure the View: Suppressed Transmission through an Ultrathin Metal Film by a Subwavelength Hole Array

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If a metal film, thick enough to be totally opaque, is perforated by tiny subwavelength holes in an orderly fashion, the transmission will be enhanced extraordinarily [T. W. Ebbesen, *et al.* Nature (London) **391**, 667 (1998)]. Here, we investigate the transmission through an ultrathin semitransparent Au film with a square array of subwavelength holes and observe the opposite behavior: less light is transmitted through the pierced metal compared to the closed film.

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The observation that the presence of a subwavelength hole array (SWHA) in a thick opaque metal film can lead to an extraordinary optical transmission [1] has triggered a huge number of publications over the past decade [2–7]. Although agreement exists that surface plasmon polaritons play a crucial role, there is still an ongoing debate on the physics leading to a transmission exceeding the value predicted by Bethe [8] by orders of magnitude. For thick opaque films, the transmission through SWHAs increases with decreasing film thickness down to about 100 nm, where it is believed to saturate [9]. For ultrathin semitransparent films, Spevak *et al.* recently predicted a resonantly suppressed transmission through one-dimensional periodically modulated films [10]. Although SWHAs in semitransparent metal films allow for a quantitative measurement of the transmission by using the unperforated film as reference, to our knowledge, there are no investigations of SWHAs in ultrathin films with a thickness comparable to the skin depth of the metal. In this study, we measured a two-dimensional hole array in a semitransparent Au film and found an unexpected behavior: less light is transmitted through the perforated film compared to the closed metal film.

For our studies, we prepared a 2×2 cm^2 square hole array with a periodicity of $P = 300$ nm and a hole diameter of $d = 200$ nm in a $t = 20$ nm thick gold layer on glass, fabricated by means of optical interference lithography. As substrate BK7 glass was used. Prior to the Au metallization, the substrate was coated with 2 nm chromium as adhesion layer. Afterwards, the sample was spin coated with negative photoresist, illuminated, and developed. The gold film was then textured via argon ion dry etching. Details of the preparation procedure are described in [11]. Figure 1 shows a scanning electron microscopy image together with the scheme of the specimen. This structure was characterized by transmission measurements as well as by spectroscopic ellipsometry at various angles of incidence and different azimuth orientations in the energy range 0.6 to 4.6 eV (4400 to 37 000 cm^{-1}; 0.27 to 2.3 μm). Both ellipsometry as well as transmission measurements were performed by a Woollam VASE rotating analyzer spectroscopic ellipsometer. At normal incidence, the size of the illuminating beam is about 2.2 mm, which expands to 12.7 mm along the x axis for angles of incidence of 80°. As reference for all transmission measurements the bare glass substrate was used.

First, the pristine Au film was characterized by spectroscopic ellipsometry [Fig. 2(a)]. The obtained dielectric constant $\hat{\varepsilon}_m = \varepsilon_{1m}(\omega) + i\varepsilon_{2m}(\omega)$ resembles the literature bulk value [12] and perfectly describes the transmission measurements. Then, a spectrum transmitted through the array of holes was recorded under normal incidence [Fig. 2(b)]. In contrast to expectations, additional holes do not increase the transmission, but the inverse effect is observed: the transmission is reduced. The spectra of the pristine Au film and the perforated film differ by an additional strong absorption at about 1.96 eV. The spectrum of the hole array can be perfectly modeled by using one Lorentz oscillator and slightly modified dielectric constants for Au [fit in Fig. 2(b)]. The slight change of the optical properties of the Au film most likely arise from the

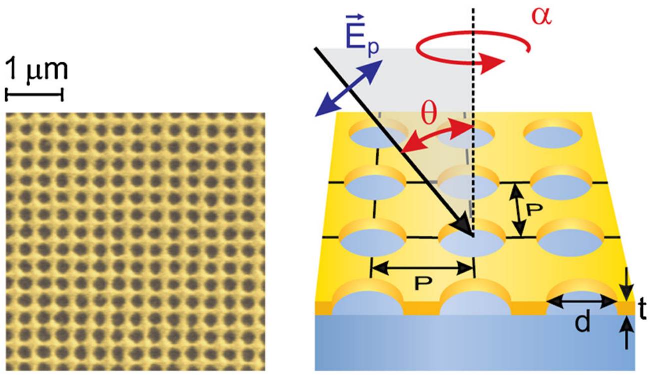

FIG. 1 (color online). Scanning electron microscope image and scheme of the investigated hole array. Parameters: period $P = 300$ nm; hole diameter $d = 200$ nm; Au film thickness $t = 20$ nm. The light is polarized with the plane of incidence (p polarization); the angle of incidence is denoted by θ, the azimuth angle by α.

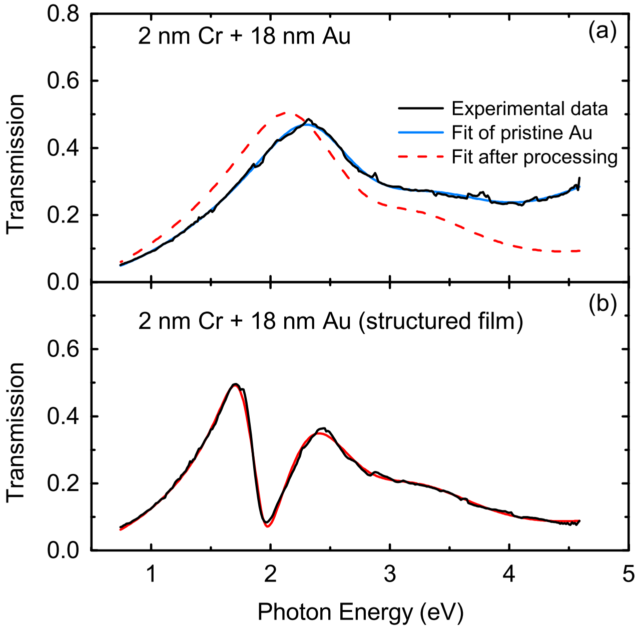

FIG. 2 (color online). Transmission spectra of (a) the pristine Au film and (b) the hole array at normal incidence. The transmission through the array can be perfectly fitted by using slightly modified Au dielectric constants and one additional Lorentz oscillator. The dashed line in panel (a) corresponds to the transmission calculated for a closed metal film with dielectric properties identical to the Au of the perforated film.

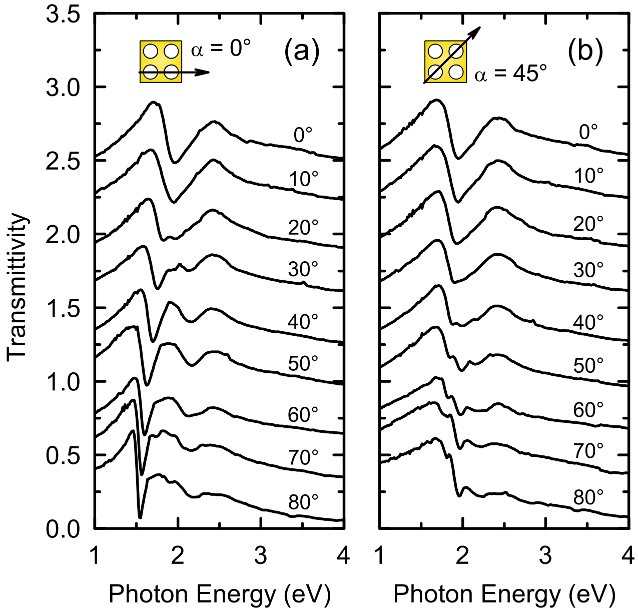

FIG. 3 (color online). Transmission spectra of the hole array measured for varying angles of incidence θ with p polarization (a) along the holes; (b) under 45°. In both graphs, the curves have been plotted with subsequent vertical offsets of 0.3.

lithographic process. To illustrate this effect, a transmission spectrum of a hypothetic closed Au film possessing these modified optical constants is additionally displayed in Fig. 2(a).

Before we discuss the physical origin of the strong absorption, let us look at the anisotropy of the SWHA. Under normal incidence, the spectrum is independent of the azimuth orientation of the sample, i.e., it does not show any in-plane anisotropy as expected for a square array. However, with increasing angle of incidence, the pronounced absorption peak at 1.96 eV strongly shifts to lower energies for the plane of incidence along the holes, i.e., for an azimuth angle $\alpha = 0°$ or $90°$ [Fig. 3(a)], whereas for $\alpha = 45°$ only a small shift is observed [Fig. 3(b)]. Furthermore, for both orientations additional absorption peaks occur at oblique incidence, less dominant but clearly visible. This behavior excludes a simple out-of-plane anisotropy as this would affect all azimuth orientations in the same way; it clearly evidences that the optical properties cannot be described by some effective optical constants or a dielectric tensor. The observations can only be understood by considering $\vec{k}$-dependent optics [13].

The periodic structure of the SWHA enables light to couple to surface plasmon polaritons via reciprocal wave vectors [14]. For thick films, plasmons can be excited via a two-dimensional array when the following conditions are fulfilled:

$$|\vec{k}_{\mathrm{SP}}| = \frac{\omega}{c}\sqrt{\frac{\hat{\varepsilon}(\omega)_m \varepsilon_d}{\hat{\varepsilon}(\omega)_m + \varepsilon_d}} \tag{1}$$

$$\vec{k}_{\mathrm{SP}} = \vec{k}_x \pm m\vec{G}_x \pm n\vec{G}_y \tag{2}$$

where $\vec{k}_{\mathrm{SP}}$ is the surface plasmon wave vector, $\vec{k}_x = \vec{k}_0 \sin\theta$ with $k_0 = \omega/c$ the component of the incident photon's wave vector projected onto the plane of the grating, and $|\vec{G}_x| = |\vec{G}_y| = 2\pi/P$ are the reciprocal lattice vectors of the square array with m and n being integers. In this case, plasmons are excited on both interfaces of the metal film if the above conditions are fulfilled for the respective dielectric constants ε_d.

For decreasing film thicknesses, the surface plasmon polariton modes guided by the two interfaces couple due to the overlap of their fields inside the metal. Starting from Maxwell's equations and taking into account all boundary conditions, the following equation can be derived for these coupled modes [15,16]:

$$\tanh\{S_2 t\} = -\frac{\hat{\varepsilon}_m S_2(\varepsilon_{d1} S_3 + \varepsilon_{d2} S_1)}{\varepsilon_{d1}\varepsilon_{d2} S_2^2 + \hat{\varepsilon}_m^2 S_1 S_3}. \tag{3}$$

The values S_1, S_2, and S_3 are defined by the relations

$$S_1^2 = k_x^2 - \varepsilon_{d1} k_0^2, \qquad S_2^2 = k_x^2 - \hat{\varepsilon}_m k_0^2,$$
$$S_3^2 = k_x^2 - \varepsilon_{d2} k_0^2.$$

In our sample $\varepsilon_{d1} = 1$ (air) and $\varepsilon_{d2} = 2.25$ (glass) and therefore $\varepsilon_{d1} < \varepsilon_{d2}$. For this strongly asymmetric geometry and for very thin films, Eq. (3) yields only one strongly

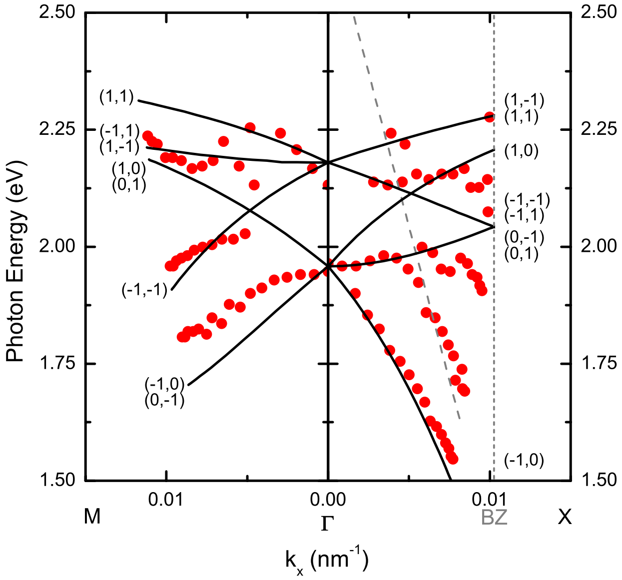

FIG. 4 (color online). $\vec{k}$-dependent plasmon energies from Eq. (3) (black lines) and obtained minima from Fig. 3 (red dots). The different branches correspond to different pairs of values (m, n) as indicated.

damped (antisymmetric) short range surface plasmon mode [16]. The calculated energies of this plasmon are plotted in Fig. 4 (black lines) as a function of the in-plane wave vector along the Γ-X direction (along the holes) as well as along the Γ-M direction (45° to the holes) in reciprocal space. The different branches correspond to different (m, n) integers as indicated in the figure; the Brillouin zone boundary (BZ) is given by $\pi/P = 0.01\ \mathrm{nm}^{-1}$. The dashed line corresponds to Wood's anomaly that occurs when a diffracted order becomes tangent to the grating plane [17]. It has to be stressed that for the calculations only, the structure parameters P and t and the measured dielectric function $\hat{\varepsilon}_m(\omega)$ of the pristine Au film were used, without any free parameters. The minima obtained from the $\theta(\vec{k})$-dependent spectra (Fig. 3) are added as red dots in Fig. 4. For the $(-1, 0)$ branch (Γ-X direction), which describes the pronounced minimum starting at 1.96 eV, the match between measurement and calculation is almost perfect. The agreement for the $(-1, 0)$, $(0, -1)$ branch (Γ-M direction) is less pronounced but still describes the overall behavior remarkably well. In general, the minima are relatively broad and therefore the exact positions are hard to define particularly at small $\vec{k}$ values. That might be one reason why the agreement for the higher branches is less convincing. Nevertheless, Fig. 4 gives clear evidence that for the SWHA investigated here the excitation of a short range surface plasmon leads to a strong absorption only and thus suppresses the transmission. There is no region in the whole spectra under investigation where the transmission of the hole array exceeds that of the pristine metal film. In other words, punching holes in a metal film, does not give you a better view.

On both counts this is an unexpected result: In the framework of findings on thick SWHAs one would expect that more light is transmitted through the perforated than through the closed metal film. But also from prior results on very thin closed metal films one would expect an increased transmission. Very thin films with a strongly asymmetric geometry ($\varepsilon_{d1} < \varepsilon_{d2}$) were studied by several authors using attenuated total reflection techniques [18,19]. With this method they also found that only strongly damped short range surface plasmons can be excited. Contrary to our results, on these rough metal films nonradiative surface plasmons decay into photons leading to light transmission through an otherwise more or less opaque metal film.

A nice descriptive explanation for the extraordinary transmission through SWHAs was given by Genet *et al.* [20]. They developed a Fano analysis of the related scattering problem by distinguishing two interfering contributions: light directly scattered by the hole array (transmission continuum) and light scattered via the resonant excitation of surface plasmons. The Fano-type profile of the observed spectral features is then mainly given by the dimensionless parameter q [21]:

$$q = \frac{2\delta}{\Gamma} \tag{4}$$

where Γ is the linewidth and the parameter δ defines the ratio between the resonant transition amplitude and the direct transition amplitude. For SWHAs in thick films, the fraction of light resonantly scattered by surface plasmons and that nonresonantly by the hole array (direct transmission) is approximately in the same order of magnitude, and the linewidth is relatively small leading to q values of the order of 20 (Fano resonance) [22]. Reducing the film thickness to the range of the skin depth adds a third contribution, the direct transmission through the metal film. This not only increases the nonresonant transmission by roughly 2 orders of magnitude, but it also leads to a much larger linewidth Γ due to the excitation of strongly damped short range plasmons. As a consequence, $q \longrightarrow 0$, or in other words the Fano resonance degenerate to a Lorentzian absorption peak as observed in our experiments. This also explains why we observe our plasmons exactly at the positions calculated by Eq. (3) and not red shifted as in thick SWHAs.

The films investigated in this work (200 nm holes and a periodicity of 300 nm) cover roughly half of the surface area and are therefore comparable to evaporated "amorphous" films close to the percolation threshold. In those films, a dielectric anomaly occurs leading to a transmission higher than that of the bare substrate [23,24]. On this background the role of surface roughness, periodic surface modulations and the presence of holes in very thin metal films, seems to be still not clear. For their description, the altered effective dielectric properties of ultrathin metal films had to be taken into account as well. Whether the observed reduced transmission is simply an evanescent diffracted order effect or whether it is associated with hole resonances can only be answered by modelling the detailed structure, for example, with rigorous coupled wave analysis.

In summary, we have demonstrated that the transmission through ultrathin metal films is considerably suppressed when regularly perforated by subwavelength holes. This behavior can be qualitatively explained by a Fano-type analysis of the two contributing scattering processes: for ultrathin films, the light nonresonantly scattered increases by orders of magnitude compared to the resonantly scattered light by surface plasmons. This causes a strong absorption instead of an enhancement in the transmission spectrum. The observed energies of the plasmons agree well with our calculations when the strong coupling between the two interfaces is taken into account.

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