Gap plasmon-based phase-amplitude metasurfaces: material constraints

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Abstract: Gap surface plasmon (GSP) based metasurfaces, consisting of a subwavelength thin dielectric spacer sandwiched between a metal film and an array of metal nanobricks, have in recent years attracted considerable attention due to the ease of fabrication and the possibility to control both the phase and amplitude of the reflected light. In this work, we numerically investigate the influence of metal properties on the performance of GSP-based metasurfaces, considering in detail (at the wavelength $\lambda = 800\,\text{nm}$) the typical plasmonic metal - gold, the alternative plasmonic material - titanium nitride, and the ideal metal (i.e., perfect electric conductor). We demonstrate that the plasmonic properties of non-ideal metals, in addition to the possibility to engineer the amplitude of the reflected light, also lead to a wider range of reflection phase control for relatively small unit cell sizes of $\sim \lambda / 3$ as compared to the metasurfaces using the ideal metal. Moreover, titanium nitride is found to represent a viable alternative (to gold) material that promises less stringent requirements when designing amplitude and phase-gradient GSP-based metasurfaces.

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References and links
1. Introduction

Contemporary with the advance in nanofabrication technology in the last few decades, the optics community has come to realize the possibility to fully control light at the nanoscale using man-made materials, the so-called metamaterials [1]. Several groundbreaking applications have been suggested and verified, such as super-resolution imaging [2] and invisibility cloaks [3, 4], but the transfer of metamaterials to commercially oriented products seems hindered by difficulties in fabrication and, particularly at optical wavelengths, too high losses associated with the usage of metals.

The first issue of time-consuming and cumbersome fabrication, related to the three-dimensional nature of true metamaterials, can in many ways be circumvented by utilizing the planar analog, the so-called metasurfaces. These surfaces are subwavelength thin and typically only consists of one or few layers of nanostructure constituents, hereby making them particularly suitable for planar fabrication techniques, while full control of light is preserved by proper engineering of the constituents [5–7]. A general overview of the current state of metasurface designs, realizations, and applications can be found in [8–10]. However, one configuration that has attracted considerable attention (in practically all frequency regimes) consists of a periodic arrangement of metal nanobricks (also known as nanopatches) on top of a subwavelength thin dielectric spacer and optically thick metal film [see, e.g., Fig. 1(a)]. At optical wavelengths, this type of metasurface can be fabricated in just one step of electron beam lithography, while proper choice of geometrical parameters allows for full control of phase and amplitude of reflected light [11–13]. For this reason, a wide range of flat optical components have been realized, such as wave plates [14–19], lenses [20], and blazed gratings [12, 21], but also more advanced applications have appeared, like polarization-controlled excitation of surface plasmon polariton [22], holography [23, 24], mathematical operations on light [25], and for determination of light’s state of polarization [26]. Regarding the possibility to control the phase and amplitude of the reflected light in these metal-backed nanobrick arrays, we would like to emphasize that this property owes to the excitation of gap surface plasmons (GSPs) that propagate in the gap between the nanobricks and metal film, thus experiencing Fabry-Perot like resonances due to multiple reflections at nanobrick boundaries [27]. The reflected light then undergoes a ∼ 2π phase shift as the nanobrick dimension (for a fixed wavelength) is varied across the resonant brick configuration, while the confinement of the GSP mode determines the level of absorption at the resonance [12].

In the above-mentioned applications, it is worth noting that the metal of choice is typically gold due to its inertness towards the surrounding air and relatively good plasmonic properties. Nevertheless, in many metasurface applications gold is far from an ideal choice. For example, when considering pure phase manipulation of light, as in wave plates and lenses, it is from a point of view of efficiency desirable to avoid Ohmic losses, which can be accomplished by replacing metallic particles with high-permittivity counterparts [28, 29]. In other applications, like for light processing [25] and true (i.e., phase and amplitude modulated) holograms [30,31], Ohmic losses seem needed in order to engineer the amplitude of the light. For these applica-
tions, however, additional material challenges exist in transferring all these exciting developments into a realm of practical applications. Gold is an expensive material and, most importantly, not integrable with industry-standard semiconductor manufacturing technologies. As a way to circumvent this problem, research in recent years has begun searching for alternative plasmonic materials [32]. Most noteworthy, transparent conducting oxides have been suggested as viable alternatives to gold and silver at near-infrared wavelengths [33,34], while the family of transition-metal nitrides, including the particularly promising titanium nitride (TiN), may find usage in the optical regime [35–38]. It should be noted that TiN does not feature smaller Ohmic losses than gold, but it is a CMOS (complementary metal-oxide semiconductor) compatible material with the additional benefit of optical properties being dependent on the processing conditions.

In this work, we numerically investigate the complex reflection coefficient of GSP-based metasurfaces, operating at a wavelength $\lambda = 800$ nm, as a function of geometrical parameters when the metal is assumed to be gold, titanium nitride, and a perfect electric conductor (PEC). The PEC calculations are included to shed light on the plasmonic effect of gold and titanium nitride on the metasurface performance, while it also makes a direct reference to the low-frequency response of the analog reflectarrays, although the typical dissipative losses in the spacer layer of these structures [39] are neglected in our study. We demonstrate that, unlike the PEC metasurfaces, the plasmonic properties of gold and TiN metasurfaces allow for a practically full control of the reflection phase for unit cells of size $\sim \lambda / 3$, while also offering the possibility to engineer the reflection amplitude. In general, realistic values of the permittivity of TiN entail stronger Ohmic losses in GSP-based metasurfaces than in the gold counterparts, but the utilization of a larger spacer thickness can partly resolve this issue. At the same time, larger absorption results in slower changes of the reflection phase in the vicinity of resonance, promising thereby less stringent requirements when designing amplitude and phase-gradient GSP-based metasurfaces. Overall, TiN seems to be an attractive alternative plasmonic material in GSP-based metasurfaces operated at the long wavelengths of the visible regime or at near-infrared wavelengths.

2. Gap plasmon-based metasurfaces and resonators

In designing inhomogeneous metasurfaces, typically featuring a phase-gradient along the metasurface, the starting point is always the homogeneous counterpart that is characterized by an array of identical nanostructures and, hence, can be efficiently modeled using a single unit cell with periodic boundary conditions applied on the side walls. By assuming that the coupling between neighboring particles are negligible, the inhomogeneous metasurfaces, incorporating a certain functionality, can be constructed from the optical properties of the simpler homogeneous metasurface. For this reason, the main part of this work focuses on the phase and amplitude of reflected light from GSP-based homogeneous metasurfaces. The unit cell, as shown in Fig. 1(a), consists of a glass spacer of thickness $t_s$ sandwiched between an optically-thick metal film and an array of nanobricks separated by the subwavelength distance $A$ and characterized by the widths $(L_x, L_y)$ and the thickness $t$. The refractive index of the glass spacer is assumed to be 1.45, while the permittivity of gold is described by interpolated experimental values [40]. In all calculations, the incident light is assumed to be a monochromatic plane wave normally incident on the metasurface and polarized along the $x$-direction. Since all our previous successful applications of GSP-based metasurfaces have been realized at a wavelength of 800 nm [12, 13, 16, 17, 20, 22, 25, 26], this is also the wavelength of choice in the following numerical study that has been conducted using the commercially available finite element software Comsol Multiphysics, ver. 5.0.

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Fig. 1. Reflection from gap plasmon-based metasurfaces. (a) Sketch of basic unit cell consisting of a metal nanobrick on top of a glass spacer and metal substrate. Panels (b)-(d) show the calculated reflection coefficient as a function of nanobrick widths for $\Lambda = 250$ nm, $t = t_s = 50$ nm, and wavelength $\lambda = 800$ nm when the metal is assumed to be gold ($\varepsilon_{\text{Au}} = -24.1 + i1.5$), titanium nitride ($\varepsilon_{\text{TIN}} = -9.3 + i8.5$), and a perfect electric conductor, respectively. Color maps show the amplitude of the reflection coefficient for $x$-polarized normal incident light, while lines are contours of the reflection phase.

2.1. The influence of metal properties

Let us start by discussing a type of gold configuration typically used for phase manipulation, which consists of an array of nanobricks with significant subwavelength periodicity $\Lambda = 250$ nm, a spacer thickness of $t_s = 50$ nm that ensures a rather weak near-field coupling between the metal layers, and a nanobrick thickness of $t = 50$ nm. Figure 1(b) displays the reflection amplitude (color map) and phase (contour lines) as a function of the nanobrick widths varying from 15 nm to 240 nm (in steps of 5 nm). It should be noted that the reflection phase is evaluated at the top most layer of the nanobrick, while the phase is presented in an unwrapped fashion, meaning that it is not limited to $[-180^\circ; 180^\circ]$ but continuously grows (for a fixed $L_y$) as $L_x$ increases. It is seen that for most nanobrick dimensions the reflection amplitude remains high ($|r| > 0.85$), which is even the case at the GSP resonance that is characterized by an accumulation of phase contour lines and (for $L_y > 70$ nm) a weak dip in the reflection amplitude. Moreover, one notices that the span of nanobrick dimensions makes it is possible to cover $\sim 90\%$ of the $2\pi$ phase space. Accordingly, the gold configuration allows for design of phase-gradient metasurfaces that manipulate light with high efficiency and the possibility to discretize the phase space in up to ten steps, though previous studies have shown a discretization in six steps to be more than adequate [12, 13].

We now discuss the same configuration as above, but this time gold is replaced with tita-
nium nitride. Without dwelling on the details, it ought to be mentioned that the exact optical properties on TiN depend on many processing parameters, like temperature, pressure, ambient composition, and substrate [37, 38], that can significantly change the strength of the metallic response. Here, we choose an intermediate value of $\varepsilon_{\text{tin}} = -9.3 + i8.5$ that was experimentally determined in [41]. The associated reflection map of the GSP-based TiN metasurface is shown in Fig. 1(c), clearly demonstrating a lower level of reflection as compared to the gold case, with the possibility of practically all light being absorbed for certain nanobrick dimensions. More interestingly, the phase contour lines seem to originate from a single point defined by the minimum in the reflection amplitude, while the upper and lower parts of the reflection map (indicated by a dashed line) show noticeably different optical properties. In the upper (lower) part of the reflection map, the reflection phase as a function of $L_x$, keeping $L_y$ constant, changes by more (less) than 180° as one moves across the GSP resonance. Regarding the design of phase-gradient metasurface, it is evident that the TiN metasurface is a viable alternative, since nanobrick dimensions can be chosen to cover the whole phase space, though the efficiency would be reduced compared to the gold configuration.

In connection with the reflection from GSP-based TiN metasurface [Fig. 1(c)], it is appropriate to mention a recent work that treats GSP-based metasurfaces as a one-port single-resonance model within the coupled mode theory [42]. Within this framework, the specific behavior of the reflection coefficient depends not only on the resonance wavelength but also on the probability that the excited GSP mode will decay either radiatively or non-radiatively (i.e., by Ohmic heating). If the two decay channels are equally probable, one reaches the condition of perfect absorption, which in our TiN metasurface corresponds to the point of which all phase contour lines emanate from. Moreover, the upper (lower) part of the reflection map represents nanobrick configurations where the GSP mode is more (less) likely to decay radiatively than non-radiatively, hence featuring a phase variation larger (smaller) than 180° as one moves across the resonance. In regard to the gold metasurface in Fig. 1(b), it is worth noting that all nanobrick dimensions feature a GSP mode that is more likely to decay radiatively than non-radiatively.

In order to better gauge the plasmonic effect of gold and titanium nitride metasurfaces, while also establishing a link to the analog reflectarrays in the low-frequency regime (e.g., microwave regime), we have calculated the reflection coefficient when the metal is assumed to be a perfect electric conductor [Fig. 1(d)]. We note that the configuration is dispersionless and, hence, can be scaled to any frequency regime of interest. As expected, the reflection amplitude is one for all nanobrick configurations, since light cannot be transmitted nor absorbed. The reflection phase, on the other hand, shows a significant dependence on $L_x$ with almost parallel contour lines, which is an advantageous property when designing birefringent phase-gradient metasurfaces [13]. The different nanobrick configurations, however, only covers $\sim 75$% of the phase space, meaning that in a corresponding phase-gradient metasurface the reflection phase must be discretized in minimum steps of 90°. As a final comment, it should be mentioned that low-frequency reflectarrays, unlike the configuration in Fig. 1(d), typically employ brick thicknesses that are significantly smaller than the dielectric spacer thickness (i.e., $t \ll t_s$). We emphasize, however, that the inability of light to penetrate perfect metals results in reflectarrays that only weakly depend on the brick thickness. For example, reducing the nanobrick thickness from 50 nm to 5 nm for the configuration in Fig. 1(d) leads to a metasurface that expands the phase-space coverage from $\sim 75$% to $\sim 80$%, with the concurrent unfortunate outcome of increased crowding of phase-contour lines. This issue will be discussed in more detail in the next section in connection with the influence of the spacer thickness.
2.2. Comparison between gold and PEC metasurfaces and resonators

In the following, we make a closer comparison of the reflective properties of gold and PEC metasurfaces as the spacer thickness is decreased to 10 nm and 20 nm, keeping all other parameters identical to the previous subsection. For the case of gold configurations [Figs. 2(a) and 2(b)], the decrease in spacer thickness implies a stronger confinement of the GSP mode in the spacer layer (i.e., higher mode index), hereby resulting in resonant behavior for smaller $L_x$ and an increase of absorption at this GSP resonance. This is particularly evident for $t_s = 10$ nm [Fig. 2(a)], where a higher-order GSP resonance appears at $L_x \approx 235$ nm, while the usual GSP resonance at $L_x \approx 85$ nm entails of significant dip in reflection amplitude, followed by a concurrent rapid change in reflection phase as a function of $L_x$. We note that the strong absorption at the GSP resonance can be used to amplitude modulate reflected light in inhomogeneous metasurfaces, but a simultaneous control of the reflection phase implies stringent fabrication tolerances of the associated nanobrick dimensions [25]. In general, we see that decreasing the spacer layer increases the possible span of reflection phase to more than 95% of the full phase space, while the reflection maps, similar to the previous TiN metasurface [Fig. 1(c)], include a point of minimum reflection from which phase contour lines originate. As such, the stronger confinement of the GSP mode implies that a larger set of nanobrick configurations [i.e., those configurations below the dashed lines in Figs. 2(a) and 2(b)] incite the mode to decay through dissipation rather than radiatively.

Shifting our focus to the equivalent PEC metasurfaces [Figs. 2(c) and 2(d)], it is clear that compared to a spacer thickness of 50 nm [Fig. 1(d)] the available phase space increases to $\sim 85\%$, however at the expense of a progressively larger part of achievable reflection phases being contained within a small region of $L_x$ values. This behavior entails an increasing demand...
on the accuracy of the fabricated nanobrick dimensions with decreasing spacer thickness. As an alternative way of extending the phase space using GSP-based PEC metasurfaces, Fig. 3 displays the reflection map for a spacer thickness of 50 nm and unit cell periods of 300 nm and 400 nm. For these two unit cell periods the available reflection phase amounts to $\simeq 80\%$ and $\simeq 85\%$ of the full phase space, respectively. It is important to notice that the reflection phase for a specific nanobrick dimension is practically independent of the unit cell period [compare Figs. 1(d) and 3], with the resonance, as indicated by a fast variation in reflection phase as a function of $L_x$, occurring around $L_x \sim 200$ nm. It is this almost twice as large resonant nanobrick dimension compared to the gold and titanium nitride metasurfaces [Figs. 1(b) and 1(c)] that hinders PEC metasurfaces in attaining the same span of reflection phase for a unit cell period as small as $\sim \lambda/3$ ($\Lambda = 250$ nm). In this regard, it is appropriate to mention that unit cells of reflectarrays in the low-frequency regime can be reduced to the size $\sim \lambda/3$ by utilizing complex-shaped elements, like fractal-shaped patches [43, 44].

As a way to better understand the underlying physics behind the markedly different behavior of gold and PEC metasurfaces, we have calculated the extinction cross section as a function of wavelength from an isolated nanobrick ($L_x = L_y = 140$ nm and $t = 50$ nm) on top of a continuous spacer layer and metal film [Fig. 4(a)]. Despite the fact that both gold and PEC configurations show distinct resonances of roughly the same strength, the dependence with respect to the spacer thickness is the opposite. For the gold configuration, the GSP resonance red-shifts with decreasing spacer thickness, which naturally follows from the fact that the GSP mode index increases with decreasing spacer thickness [27], thus requiring (for a fixed configuration) a larger free-space wavelength in order to sustain the standing-wave pattern of the resonance. For the PEC configuration, however, no plasmonic effects exist and the waveguide mode associated with the standing-wave resonance is closely related to the transverse electromagnetic (TEM) wave in parallel-plate waveguides. This means that the effective mode index for any subwavelength spacer thickness is noticeably smaller than the GSP counterpart and close to the refractive index of the spacer material, only slightly modified due to the finite $L_y$ of the nanobrick. For this reason, the observed blue-shift of the resonance wavelength for decreasing spacer thickness is a consequence of a concomitant decrease in the effective resonator length ($\neq L_x$). Physically, the effective resonator size is related to the extend of the mode field beyond the nanobrick boundaries, which will reduce as the spacer thickness is decreased [46]. With regard to plasmonic versus PEC metasurfaces, we note that the smaller mode index of the PEC waveguide mode compared to the GSP mode results for a fixed wavelength in larger resonant...
nanobrick configurations, which is the reason why PEC metasurfaces with subwavelength unit cell size of $\sim \lambda / 3$ do not reach the same span of reflection phase as the plasmonic counterparts. Furthermore, the decrease in spacer thickness of PEC metasurfaces implies (for a fixed wavelength) an increase in the size of the resonant nanobrick, hereby explaining why the reflection maps in Figs. 2(c) and 2(d) show increasing regions of practically constant reflection phase. At the same time, the reduced line widths of the resonances for decreasing spacer thickness [see Fig. 4(a)] explain why the phase contour lines of the associated metasurfaces contract. As a final remark, we emphasize a fundamentally different aspect of gold and PEC resonators. For the considered PEC resonator, the excited mode can only decay by radiation, whereas the GSP mode in a gold configuration may decay either radiatively, non-radiatively, or by excitation of an SPP. The influence of the three decay channels is shown in Fig. 4(b) as a function of spacer thickness. As expected, the non-radiative decay channel becomes progressively more important for decreasing spacer thickness due to the stronger confinement of the GSP mode. It is also this effect that underlies the occurrence of gold metasurface configurations where the GSP resonance implies a reflection phase shift less than 180° [see Figs. 2(a) and 2(b)].

2.3. Titanium nitride metasurfaces

Having presented, discussed and clarified the different optical properties of plasmonic and perfect metal GSP-based metasurfaces, we focus in the remaining part of this work on important aspects of titanium nitride metasurfaces. As already stated in the Introduction, the optical properties of titanium nitride depend in a nontrivial way on the processing conditions, meaning that TiN can show strong or weak metallic response in the visible and beginning of the near-infrared regime. The strength of the metallic response can be quantified by the magnitude of the negative real part of the permittivity, with experimental values at 800 nm ranging from $\text{Re}\{\varepsilon_{\text{tin}}\} = -4.9$ in [37] to $-12.7$ in [47]. These two values represent TiN with a less and more metallic behavior than used in the calculations of Fig. 1(c), respectively, and the associated metasurface reflection maps are displayed in Fig. 5. It is clear that the concurrent expel of the electromagnetic field within TiN as the metallic response increases leads to a reduced absorption and, hence, a larger span of nanobrick dimensions for which the GSP resonance entails a phase shift greater than 180°. For example, a too weak metallic response, as exemplified in Fig. 5(b), makes light ma-
Fig. 5. Reflection from titanium nitride GSP-based metasurfaces. (a) Highly and (b) weakly metallic metasurface configuration, with the TiN permittivity $\varepsilon_{\text{tin}} = -12.7 + i5.4$ [47] and $\varepsilon_{\text{tin}} = -4.9 + i6.1$ [37], respectively.

Fig. 6. The influence of spacer thickness on reflection from titanium nitride metasurfaces. Reflection coefficient as a function of nanobrick widths for (a) $t_s = 20\text{ nm}$ and (b) $t_s = 80\text{ nm}$ when the TiN permittivity is $\varepsilon_{\text{tin}} = -12.7 + i5.4$. The remaining parameters are kept constant at $\Lambda = 250\text{ nm}$, $t = t_b = 50\text{ nm}$, and $\lambda = 800\text{ nm}$.

The achievable range of reflection amplitude and phase of TiN metasurfaces can, similar to the gold metasurfaces in Fig. 2, also be altered by varying the spacer thickness, as shown in Fig. 6. It is clear that a spacer thickness of 20 nm [Fig. 6(a)] brings one to the regime of the GSP mode being more likely to decay non-radiatively than radiative, hereby precluding the possibility for phase control of reflected light. Increasing the spacer thickness to 80 nm [Fig. 6(b)], on the other hand, renders a reflection map that covers the whole $2\pi$ phase space (due to the presence of the zero reflection point from which all contours lines originate) while nanobrick dimensions with arbitrary reflection phases can be chosen to feature reflection amplitudes of $\sim 0.6 - 0.8$. Overall, the efficiency of these TiN metasurfaces is still not as high as for the gold counterparts, but it is evident that the higher Ohmic losses associated with TiN can be partly compensated in GSP-based metasurfaces by utilizing a larger spacer thickness. Moreover, TiN
metasurfaces seem attractive for both amplitude and phase modulation of reflected light, since fabrication tolerances on nanobrick dimensions can be relaxed compared to gold configurations due to the larger spreading of phase contour lines [compare Figs. 2(b) and 6(b)].

3. Conclusion

In summary, we have numerically studied at a wavelength of $\lambda = 800\text{ nm}$ the reflection phase and amplitude from GSP-based metasurfaces as a function of geometrical and material parameters, with the metal representing gold, titanium nitride, and a perfect electric conductor. The PEC calculations are included to shed light on the plasmonic effects of gold and TiN, but also to make a direct reference to the low-frequency analog reflectarrays. Through a careful examination of calculation results of metasurfaces and isolated resonators, we have shown that the physically different nature of resonances in plasmonic and PEC-based configurations leads to reflection coefficients that depend in fundamentally different ways on the spacer thickness and nanobrick dimensions. Particularly, gold and TiN metasurfaces with proper geometrical parameters allow for almost full phase control of reflected light with unit cells of size $\sim \lambda/3$, while these materials also offer a way to engineer the reflection amplitude by dissipation of energy in the metal. We note that the Ohmic losses associated with plasmonic materials are often considered a disadvantageous, but we would like to emphasize that certain metasurface applications, like analog light processing [25] and holograms [30, 31], may exploit this property. In fact, this point of view has recently been elaborated on for plasmonic applications in general [48]. Finally, realizing that the exact optical properties of TiN depend on the processing condition, we also studied how different (but realistic) material parameters affect the reflection from GSP-based metasurfaces. It is clear that TiN metasurfaces generally exhibit higher absorption losses than the gold counterparts, but the additional Ohmic losses can partly be compensated by utilizing a thicker dielectric spacer. Moreover, TiN losses actually appear to be an attractive property for applications requiring both amplitude and phase manipulation of reflected light. Finally, we note that recent experimental study of TiN films has shown permittivity values near 800 nm that are closer to gold than employed in this work [38]. Overall, TiN is judged to be a viable material for the GSP-based metasurfaces operating at near-infrared and longer wavelengths, with the technologically crucial property of being a CMOS compatible material. We believe that the considered range of metallic properties allows one to identify other alternative plasmonic materials (transparent conducting oxides or transition-metal nitrides) that might be suitable for specific applications of GSP-based metasurfaces.

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