Plasmonic Black Metasurface by Transfer Printing

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In various applications simultaneous large optical absorption and large thermal and electrical conductivity are desired. As bulk materials cannot fulfill that need, metamaterials have been developed that often require complicated nanotechnology. The work presents the facile fabrication of black metasurfaces consisting of silver only. Silver nanoparticles (AgNPs) are transferred printed onto a silver film from a polydimethylsiloxane (PDMS) stamp. Numerical simulations confirm that gap plasmon modes between the particles and the film are responsible for increased absorption and also show that a nanoscale variation of the AgNP size increases the spectral width and magnitude of the absorption. The presented transfer printing enables such control of the AgNP size by introducing a relief at the PDMS stamp surface. In a case study a grating relief of a period of 277 nm is used. The resulting highly conductive metasurface shows large absorption of up to 97% for transversal magnetic polarization and over 70% for unpolarized light from the near-ultraviolet to near-infrared range for incident angles below 45°.

1. Introduction

Metals exhibit a large absorption coefficient, high thermal and high electrical conductivity at the same time, but reflect most of the incoming electromagnetic radiation. However, if a metal is structured on the nanometer scale, absorption can be achieved nonetheless, due to effective reflection suppression by impedance matching. In nature, this phenomenon cannot be observed due to the lack of sufficient surface modifications on small scales. So far, metallic absorption has been demonstrated in various experimental systems such as nanocavity systems, metallic gratings, nanoparticles, and metallic slits. Recently, metamaterials have also drawn enormous attention due to their highly tunable electromagnetic properties over a broad range of frequencies and novel fields of research such as flatland optics, hyperlenses, and nanoscopic phase manipulation. Especially plasmonic metasurfaces benefit from extraordinary high field intensities, enabling improved absorption of electromagnetic radiation at visible and infrared wavelengths. Providing high electrical with thermal conductivity, plasmonic metamaterial absorbers are therefore promising for various applications such as solar energy harvesting, heat management, electro-thermal systems, as well as nonlinear optics and could also serve as electrodes and heat sinks at the same time.

In order to achieve absorption in plasmonic metasurface-based systems, several production methods have been developed. Many of them rely on expensive bottom-up processes or need relatively large amounts of metal and/or strongly roughened surfaces in order to obtain broadband absorption. On the other hand, highly absorbing systems with low material consumption have been realized using dielectric films in order to build metal-insulator-metal (MIM) absorbers. Further on, recent MIM absorbers are compatible with cost-efficient statistical large scale production methods. However, due to the dielectric films, the electrical and thermal conductivity of MIM absorbers are not ideal. If the functionality of MIM absorbers could be preserved without any dielectrics, highly conductive broadband absorbers on large scales with low production costs would be achieved.

We present a novel metasurface consisting of only silver, demonstrating the resonant nature of MIM absorbers without the necessity of any insulating spacer material. The technique is performed by polydimethylsiloxane (PDMS)-assisted transfer printing of silver nanoparticles (AgNP) directly on top of a thin continuous silver (Ag) film, thus forming a metasurface. In addition to that, the technique can easily be used for cost-efficient, large-area fabrication on arbitrary substrates. The development of the metasurface is based on the experimental observation, that the absorption of AgNP films on PDMS is significantly enhanced, when they are brought into contact with a silver thin film.

2. Results and Discussion

2.1. Preliminary Simulation of Simplified Geometries

We first study this phenomenon with numerical simulations, whereby AgNPs are approximated by perfect silver...
Figure 1. Finite difference time domain (FDTD) studies. Electrical field intensities of a nanosphere with a size of 40 nm in a) vacuum and on infinitely thick b) silver at resonance. c) Corresponding absorption. Dashed blue line: Mie resonance in vacuum. Solid blue line: broadened spectrum with higher total absorption on silver. d–f) Analogous simulation of an 80 nm nanosphere with a redshift of the resonance features. g) Two nanospheres of different sizes at large distance. h) Corresponding absorption (black solid line) and superposition of individual absorption spectra for comparison (black dashed line). i) Two nanospheres of different sizes at short distance. j) Absorption with additional resonances and further increase of total absorption and spectral width.
nanospheres. Figure 1a,b shows single nanospheres with diameters of 40 nm located in vacuum (dashed blue frame) or on top of infinitely thick silver (solid blue frame), and the cross section of their corresponding electric field intensities at significant resonance. In vacuum, a dipole-like electric field intensity distribution in the X–Z plane can be observed. When placed on infinitely thick silver, the local field intensity is increased by a factor of about ten and the largest electric field intensity is localized at the free space directly around the point of contact of nanosphere and infinitely thick silver. This is mainly due to the interaction between the silver nanosphere and its mirror image induced by the silver surface forming a gap plasmon mode.[35–37] Consequently, the absorption spectra in Figure 1c differ from each other. In vacuum (dashed blue line), the absorption spectrum is in close agreement with the Mie theory[38] and consists of a single resonance peak at a wavelength of 360 nm. Placed on infinitely thick silver (solid blue line), the interaction between the nanosphere and the infinitely thick silver introduces additional resonant gap plasmon modes at wavelengths between 350 and 450 nm, which leads to broadening and increase of absorption.

In Figure 1d,e larger nanospheres with a diameter of 80 nm are investigated analogously (Figure 1f). In comparison to Figure 1a–c, a redshift of characteristic absorption features is observed. Consequently, the nanosphere size is one possible parameter to tune the spectral absorption properties of the presented model system, as larger nanosphere diameters lead to a redshift of the absorption spectrum.

Figure 1g shows two nanospheres of different sizes on top of infinitely thick silver with a distance of 150 nm. The resulting absorption spectrum (Figure 1h, black solid line) is dominated by the simple superposition of the absorption spectra according to Figure 1c,f, which is visualized in Figure 1h as dashed black line. No significant increase of the total absorption in comparison to the absorption spectra in Figure 1c,f can be observed.

Finally, the same two nanospheres at a closer distance of 56 nm are investigated (Figure 1i). Most strikingly, in the resulting absorption graph (Figure 1j), two resonances with peak values of about 80% occur at a wavelength of 632 and 738 nm, respectively. Further on, a broadening of the absorption for wavelengths between 350 and 550 nm can be observed. Both peaks at 632 and 738 nm can be understood as resonant gap plasmon modes exhibiting field enhancements at both the interfaces between the nanospheres and the infinitely thick silver and between the two nanospheres. This becomes obvious in an exemplary plot of the electric field intensity at 738 nm, shown in Figure 1i. The remaining peaks between 350 and 550 nm are influenced by the reduced nanosphere distance as well. Compared to Figure 1h, the total absorption (Figure 1j) is strongly increased and broadened by the effects of the additional coupling between the nanospheres, although only the position of the nanospheres is changed.

The simulation results can be summarized as follows. Free-standing nanospheres exhibit well-known spectrally narrow Mie resonances. Broadening of these resonances and an increase of the total absorption is observed if the nanospheres are placed on infinitely thick silver, while the nanosphere size introduces spectral shifting of the absorption features. Wide-spaced nanospheres of different sizes exhibit absorption spectra dominated by the superposition of the individual single nanosphere spectra. At reduced distance, however, additional resonances strongly increase the total absorption as well as the spectral width of the absorption. Following this trend, it is straightforward to assume that absorption is attainable with an AgNP-based metasurface, since systems of many interacting nanospheres are expected to further broaden and increase the absorption. Therefore, broad particle size distributions as well as small interparticle distances, in other words a high particle density, are desired in order to achieve a highly absorbing silver metasurface.

2.2. Experimental Results

2.2.1. Test Device

For the experimental realization of an absorbing metasurface based on the simulated geometry, we apply transfer printing of AgNPs onto a thin silver film. It is a promising method for later large area fabrication of the absorbing metasurfaces as well.

The properties of an exemplary sample based on that geometry (referred to as test device) will be given in the following section. In order to fabricate the test device silver (AgNPs) needs to be deposited on silver, which typically causes layer-by-layer growth. To achieve structures comparable to Figure 1b, dielectrics are usually deposited on top of the silver film in order to break the layer-by-layer growth.[33] In the presented approach, dielectric cross-linked PDMS is used as stamp material to create AgNPs. These AgNPs are then transfer printed from the stamp onto the silver thin film. Figure 2a shows the PDMS stamp coated by AgNPs still being in contact with the silver film. Here, the PDMS has not been removed from the substrate yet. Both the silver mass thickness on the stamp (top) as well as on the substrate (bottom) are varied in a combinatorial approach with bottom = 0–100 nm and top = 0–35 nm. At bottom = 15 nm the interaction between the top and the bottom film is visible as an abrupt change of color of the test device. No significant dependence of the optical properties on bottom is observed above this critical value of 15 nm, which is close to the percolation threshold of silver films on glass. This confirms that assuming an infinitely thick silver film as applied in the numerical calculations (see Figure 1) is correct, even for very thin closed silver films. On the other hand, the optical properties of the test device strongly depend on top as the color change along the axis of top shows.

For a quantitative analysis, the absorption data of the test device are shown in Figure 2b at a constant value of bottom of 80 nm and normal incidence. The absorption (A) is calculated from the reflectance (R) and transmittance (T) via A = 1 – R – T.

The black arrow indicates the change of top between values of 7 and 35 nm. For every measured value of top, the absorption spectra consist of broad resonance peaks. For the most pronounced resonance peak, a redshift of its maximum position from 470 to 870 nm and an increase of the maximum value from 78% to 95% appears toward increasing values of top. Additionally, a broadening of the peak width, defined as full width at 70% absorption, from 113 to 400 nm is observed.

According to the simulation results, the dominant absorption can be explained by resonant excitation of localized gap plasmon modes due to the interaction between the silver substrate and the particles. The observed redshift of this dominant absorption peak is attributed to an increasing particle...
size toward larger values of $t_{\text{top}}$, as it is confirmed by the results of atomic force microscopy (AFM) measurements of the silver-coated PDMS stamp at various values of $t_{\text{top}}$ (Figure 2c). The cross-sectional scanning electron microscope (SEM) image (Figure 2d) confirms that the AgNPs consist of spherical particles and validate the assumed geometry used in the simulation. However, a distinct difference between the numerical simulation from Figure 1 and the measurement results from Figure 2 can be observed. The measured absorption peak widths are much broader than the simulated ones. This can be explained by interparticle interaction. As confirmed by the simulation (Figure 1h,j), the interparticle distance has a strong influence on the peak position. While in the simulation only two particles have been considered, the AgNP film deposited on the stamp consists of many particles with statistically varying distances. Therefore, this statistical distance variation broadens the observed absorption resonances.

Even though the simulation used a very simple geometry, it correctly describes the qualitative influence of the particle size. In the test device shown in Figure 2a, the local particle size was varied via $t_{\text{top}}$. Here, this variation is introduced over a long distance of 5 cm. Consequently, the particle size distribution can locally be considered constant (compare Figure 1b,e) and with increasing $t_{\text{top}}$ only a redshift of the spectrum is achieved.

As suggested by the simulation results, the particle size has to be varied on a nanoscale in order to achieve an even broader absorption spectrum. Furthermore, the experimental results clearly illustrate that a controlled local particle size can be obtained by the local control of $t_{\text{top}}$. This control must happen on subwavelength lateral dimensions to achieve improved absorption properties. It is then presumed that for high particle densities this variation of the particle size will lead to further broadening of the absorption spectra in Figure 2b. This idea is experimentally realized as described in the following section.

2.2.2. Fabrication Procedure of the Metasurface Absorber

The fabrication procedure for this experimental realization is shown in Figure 3. A PDMS (SYLGARD 184, Dow Corning) stamp is produced by cast and cure replication of a nanostructured master[39] (Figure 3a) and a silver film is deposited...
onto a glass substrate by thermal physical vapor deposition (PVD) (Figure 3b). The master is a commercially available sinusoidal grating (Thorlabs) with a period of 277 nm and an amplitude of 100 nm. Subsequently, a silver film with a mass thickness of $t_{\text{top}} = 22$ nm is evaporated onto the stamp surface (Figure 3c). First of all, similarly to the approach used for fabricating the test device, the sinusoidal curvature of the stamp leads to a spatial variation of the local silver thickness deposited on the stamp. Here, however, this variation is introduced at the nanoscale. In addition to that, the growth of the AgNPs is influenced by the curved stamp surface as well in order to minimize the surface energy.\[40\] This results in AgNP films with periodically modulated particle size as suggested in the drawing of Figure 3d.

The PDMS stamp is then pressed upside down onto the silver film on glass, resulting in a direct contact between the AgNP layer and the silver film (Figure 3e).

Finally, the PDMS stamp is removed from the substrate by a controlled peel-off (Figure 3f), resulting in a black metasurface consisting of silver, only (Figure 3g). Details of that transfer printing process are reported elsewhere.\[41\] The black square corresponds to the transfer printed area. It is worth mentioning, that all metasurfaces produced with that method exhibit an outstandingly high stability over time. Even over a period of 3 years, no significant change of the optical properties of the metasurfaces has been observed.

2.2.3. Properties of the Metasurface Absorber

The morphological and optical properties of the absorbing metasurface shown in Figure 3g will be presented in the following sections.

Figure 3. Fabrication procedure for the plasmonic black metasurface. a) Casting and curing of PDMS on a master template. b) Thermal PVD evaporation of a silver film on a glass substrate. c) Deposition of AgNP on PDMS stamp by PVD evaporation. d) Particle diameters can be influenced by the surface modulation. e) Adhesion of AgNPs from the PDMS stamp to the silver-coated substrate by direct mechanical contact. f) Subsequent release of the PDMS stamp. g) Photograph of the fabricated device. Black square: transfer printed area. The arrows indicate regions of the sample surfaces coated by Ag only, Ag and AgNPs, and AgNPs only, respectively.

Figure 4a shows the result of an AFM measurement. The metasurface consists of densely packed AgNPs. An analysis of the particle sizes is shown in Figure 4b and provides a broad variety of particle diameters with values from 30 nm up to 120 nm with an average particle diameter of 60 nm. As intended, the particle size is modulated periodically whereby larger particles accumulate on lines corresponding to the surface structure of the sinusoidally modified PDMS stamp.

Figure 5 shows the absorption of the metasurface in transversal magnetic (TM) polarization (Figure 5a) and transversal electric (TE) polarization (Figure 5b). An absorption above 70% is observed for both polarizations in the wavelength range between 350 and 1050 nm and for angles of incidence below 45°. In case of TM polarization, a high broadband absorption is even present for large incidence angles with a maximum absorption of 97% at an angle of incidence of 60° at a wavelength of 700 nm. As a matter of course, for TE polarized light, the absorption spectrum is nearly identical to the TM case at small incident angles. Toward increasing incident angles, the absorption between 350 and 1050 nm decreases approximately by a factor of 2 from 10° to 70°, whereby the maximum absorption of 83% is found at 750 nm for small incidence angles.

Different absorption mechanisms can be considered in order to explain the observed absorption data. First of all, analyzing the difference between TM and TE polarization, it might be suspected that surface plasmon polaritons (SPPs) excited by the particle grating play a substantial role in the observed absorption characteristics. In fact, the corresponding resonance, which can be calculated from the dispersion relation of the involved SPPs and the grating with 277 nm period, has only a minor influence. In contrast, a smooth metallic grating produced with the same stamp shows much stronger absorption at
SPP resonance (inset Figure 5a). While resonances correlating with the grating period are very weak, the spectra are dominated by wide bandwidth statistical phenomena. We conclude that periodicity is not required for the observed phenomenon as long as the stamp defines features creating different particle sizes at suitable distance (see Figure 1g,i).

A well-established description of statistical metamaterials is the effective medium approach.[42]

We therefore extracted the effective permittivity tensor of our metasurface by fitting a biaxial effective medium with 22 nm thickness on top of infinitely thick silver (see Supporting Information) to the absorption spectra in Figure 5. The resulting in-plane ($\varepsilon_r^{||} + i\varepsilon_i^{||}$) and out-of-plane ($\varepsilon_r^{\perp} + i\varepsilon_i^{\perp}$) components of the dielectric tensor are shown in Figure 6a (real part) and Figure 6b (imaginary part). The effective medium exhibits a hyperbolic behavior between 320 and 480 nm and elliptic behavior above 480 nm. Except for the wavelength region around the resonance at 480 nm, the calculated permittivities lead to rather poor absorption properties for the isolated effective medium. Therefore, the effective medium itself is not responsible for the high absorption of the sample. Only in combination with the silver film, the outstanding absorption properties of the metasurface are reached.

As predicted by the preliminary simulations, the metasurface is dominated by the interaction between the metal layer and particle layer. This again hints to the excitation of gap plasmons, which, in the simulation and discussion of the test device, were identified to be strongly dependent on particle size. Therefore, broadening of the absorption observed for the metasurface can indeed be linked to the modified particle size distribution due to the structuring of the stamp. This verifies the assumptions concerning the necessity of high particle densities and broad variations of the particle size to achieve transfer printed broadband absorbing silver metasurfaces.

3. Conclusion

In summary, we have shown that absorbing metasurfaces consisting of pure silver can be produced by transfer printing. In agreement with numeric simulations, gap plasmon modes between the thin silver film and the AgNPs printed on top lead to increased absorption. This was also validated by an effective medium description of the measured absorption. Only in presence of the silver layer, high absorption values are obtained. This phenomenon can be further enhanced by...
modifying the particle size at a lateral distance that is smaller than the wavelength of the absorbed light.

The nanoscale variation of the particle size is created by introducing a surface relief at the PDMS stamp used for printing. In contrast to more sophisticated approaches reported in literature, strictly speaking, the metasurface was not controlled at the nanoscale, but rather an otherwise statistical AgNP film was modified on the nanoscale.

In a first case study, a simple relief stamp was applied, which was cast and cured replicated from a commercial sinusoidal grating of the period 277 nm. The atomic force micrographs clearly confirm the corresponding modification of the particle size distribution. More importantly, this modification results in a metasurface showing an absorption of more than 70% for incident angles below 45°. The reported fabrication shows a large variety of parameters for further optimizing the optical properties of the resulting metasurface such as the stamp relief, evaporation parameters and materials, mass thickness, parameters of transfer printing, post-printing annealing, and other treatments.

From a wider perspective, the results show how efficiently the nanoscale modification of an otherwise statistical AgNP film can be applied in order to achieve improved optical properties. The introduction of more complex relief structures of optimized dimensions may lead to further improved absorbers. In general, many optical systems require large bandwidth operation. Hence, controlling statistical variations on large areas is highly desirable. The facile additive approach enables low-temperature printing of the metasurface to arbitrarily shaped metallic or metal coated surfaces. Further on, the metasurface provides hotspots with strong electrical field enhancements. Thus, together with its low material consumption, simple fabrication and avoidance of hazardous chemicals, we anticipate it to have a strong impact on the development of various environmental-friendly large area or cost-sensitive optical systems, sensors, and nonlinear optics as well as heat management systems and solar energy harvesting in the near future.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. The project has received funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation program (grant agreement No. 637367). P.G. acknowledges funding by the Emmy-Noether-Programm of the DFG (Deutsche Forschungsgemeinschaft).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

absorption, metamaterials, metasurfaces, plasmonics

Received: April 16, 2018
Revised: June 12, 2018
Published online: July 25, 2018

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