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Optical properties and fabrication of dielectric metasurfaces based on amorphous silicon nanodisk arrays

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Abstract: Dielectric metasurfaces based on amorphous silicon (a-Si) nanodisks are interesting for nanophotonic applications due to the high refractive index and mature/low temperature fabrication of a-Si. The investigated metasurfaces consist of a-Si nanodisk arrays embedded in a transparent film. The diameter-dependent optical properties of the nanodisk Mie resonators have been investigated by finite-difference time-domain (FDTD) simulations and spectrally-resolved reflectivity and transmission measurements. Well-ordered substrate-free a-Si nanodisk arrays were fabricated and characterized with regard to their broadband anti-reflection properties when placed on a crystalline silicon (c-Si) surface, and reflectivity/transmission properties when embedded in a polydimethylsiloxane (PDMS) film. Our results confirm broadband anti-reflection when placed on silicon, while the optical characteristics of the nanodisks embedded in PDMS are shown to be potentially useful for color/NIR filter applications as well as for coloring on the micro/nanoscale.

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1. Introduction

Recently metasurfaces composed of dielectric nanostructures (in the order of $\lambda_{air}/(2n_{sc})$; where $\lambda_{air}$ is the wavelength of light in air and $n_{sc}$ is the refractive index (RI) of the dielectric) working as Mie resonators and optical antennas have received much attention due to their unique optical properties for absorption enhancement and engineering light scattering [1–6]. Surface nanodisk/ pillar arrays show great promise for several optical and optoelectronic applications, e.g., in photovoltaics [7–10], (photo)detectors [11] and optical (color) filters [12–18]. A commonly reported approach for combining different materials in the fabrication of nanostructured dielectric metasurfaces is by transferring the structures to another surface by either polydimethylsiloxane (PDMS) embedding [16,19] or stamping [20,21]. Besides broadband anti-reflection [2] other reports have shown that crystalline silicon (c-Si) nanodisk arrays on/in silicon dioxide ($SiO_2$) [12–14] or sapphire ($Al_2O_3$) [15] can show specific colors in reflection mode.

High-index dielectric (e.g., semiconductor) nanostructures support geometrical (Mie-like) resonances in the visible-NIR spectral range, with scattering cross-sections larger than their geometrical cross-sections [2–6]. The Mie resonances in dielectric materials are formed by displacement currents instead of actual currents, resulting in lower losses than in metallic materials. Scattering of spherical particles in a homogeneous medium has been extensively studied in Mie theory [22] including analysis of particles weakly coupled to the substrate.

The optical antenna effect in dielectric nanostructures [1] has been utilized for broadband omnidirectional anti-reflection using etched c-Si nanodisk arrays on a c-Si substrate. Without coating an average surface reflection of ~7.5% was obtained in the spectral region of 450-900 nm [2]. An additional thin SiN-layer coating resulted in an average surface reflection even as low as ~1.3%. First and second order Mie resonances were identified, which are related to the geometrical properties of the Mie resonator. The presence of a sharp reflection dip...
(periodicity dip) is also reported and is due to the Wood-Rayleigh anomaly [23], related to the grating effect of the regular nanodisk array. A redshift of the Mie resonances is observed as the nanodisk diameter increases, indicating that specific resonance wavelengths can be tailored. These resonances were found to be related to electric/magnetic dipole (ED/MD) or quadrupole (EQ/MQ) modes for the nanodisks [2–18]. Other works regarding Mie resonator design have been reported [2–6], in which the individual modes were distinguished for the resonators and it was shown that these modes can be tuned (shifted, separated, combined or suppressed) by optimizing the nanoparticle shape, diameter and height and the RI of the surrounding medium. It was indicated that the driving mechanism for the in-plane magnetic dipole is due to the coupling of the electric component of the electromagnetic field to displacement loops (vertically oriented inside the nanoparticle). A MD moment, perpendicular to the electric field polarization, is induced by this displacement loop. For this, significant retardation of the driving field is required throughout the particle. The ED resonances require only collective polarization of the inside material of the resonator caused by the incidental light electric field component. Far-field signatures of these resonances can be observed. Scattering cross-section analysis can be used to identify the occurrence/behavior of the individual Mie resonances [3–6]. In this work, reflectance/transmittance spectra were used where modulation effects of these spectra could be influenced by interference of the Mie resonances with the incident light.

Nanostructuring of Si, including nanodisk-based metasurfaces [8–19,24–28], is widely reported in literature due to its attractive features, e.g., its mature fabrication/processing technology, suitable electrical and optical properties, low cost and non-toxicity. While the etching of c-Si nanodisk (array) structures is well-known, transferring the etched nanostructures to other substrate surfaces can still be an issue. In comparison, very few works have addressed the fabrication and optical properties of amorphous silicon (a-Si) nanodisks [18]. Due to the typically large thickness of the (common) c-Si layers, it is unsuitable to create substrate-free structures. For high aspect ratio structures (e.g., nanopillars or nanowires) it is possible to “break/scrape-off” the structures from the substrate. For the fabrication of substrate-free low aspect ratio Si nanodisks, silicon on insulator (SOI) can be used for etching the patterns and where the insulator (typically SiO2) can be sacrificially etched away.

An alternative way is to deposit a layer of a-Si on a sacrificial layer/substrate surface. The typical RI of a-Si is ~3.6–4.5 in the UV-NIR part of the wavelength spectrum, and has a reported bandgap of ~1.1-1.5 eV. For large scale production, the advantage of a-Si is its low cost and possibility for low temperature layer deposition. Plasma-enhanced chemical vapor deposition (PECVD) of a-Si:H is possible at temperatures between 30 and 300 °C, which makes this low temperature processing available for substrate materials like glass, plastics and metals. This paves the way for integration of nanodisk-based metasurfaces in a broader scope of material combinations than has been reported. Furthermore, the choice of a-Si for nanodisk fabrication has advantages compared to using SOI since the a-Si can, in principle, be deposited on any type of (sacrificial) layer with better flexibility over the design of the layers.

Dielectric metasurfaces composed of a-Si nanodisk arrays can be used for omnidirectional broadband anti-reflection and optical (filter) applications. In the first case, when they are placed on a high RI substrate (e.g., c-Si), surface reflections can be significantly reduced due to strong forward scattering into the substrate, and thereby increasing the absorption in the substrate (e.g., as in solar cells). On the other hand, when they are placed on/embedded in a low RI (transparent) medium (e.g., SiO2 or polydimethylsiloxane (PDMS)), coloring in the reflectance mode can be obtained, determined by the geometrical properties of the nanodisks and their array period. Similarly, in transmission mode these a-Si nanodisk metasurfaces have interesting properties for transmission filter applications. Both the reflectance and
transmittance peaks of such metasurfaces can be tuned for the (visible-)NIR wavelength region.

In this work, we report the fabrication and optical characterization of a-Si nanodisk (Mie resonator) arrays. Substrate-free a-Si nanodisk arrays were fabricated with a hexagonal array period of ~500 nm, height of 200 nm and diameters varying between ~180-380 nm. The dimension range of the nanodisks used in this work was chosen due to their ability to support multi-modes; this in order to use the relative spectral location of the occurring Mie resonances for transmission (optical) filter applications. In previous works, most of the reported Mie resonator Si structures were fabricated by either electron beam lithography (EBL) or nanoimprint lithography (NIL). Here, we use a simple method to fabricate nano-sized Mie resonator structures by a cheap, flexible and straightforward method based on a combination of colloidal lithography (CL) and inductively coupled plasma reactive-ion etching (ICP-RIE). The obtained nanodisk arrays placed on top of a c-Si substrate and embedded in PDMS are characterized by finite-difference time-domain (FDTD) simulations and spectrally-resolved specular reflectivity and transmission measurements. The main focus of this work is on the transmittance properties of the a-Si nanodisk arrays embedded in PDMS with regard to possible optical filter applications.

2. Fabrication of a-Si nanodisk-based metasurfaces

The a-Si layers were deposited by PECVD on a c-Si substrate coated with a SiO2 layer. The substrate-free a-Si nanodisk arrays were fabricated by CL, dry etching and selective wet etching. Figure 1 shows the schematic of the process steps involved in the fabrication of the a-Si nanodisk-based metasurfaces and Fig. 2 presents representative scanning electron microscopy (SEM) images of the fabrication steps.

First a 100 nm sacrificial SiO2 layer was deposited on a c-Si (100) wafer by PECVD. Next a 200 nm layer of hydrogenated a-Si (a-Si:H; further on specified as a-Si) was deposited by PECVD (SiH4 flow of 100 sccm, temperature of 250 °C, RF power of 30 W, pressure of 200 mTorr and a planar (calibrated) deposition rate of ~8.3 nm/min). This a-Si layer thickness specifies the height of the nanodisks. Next, an additional ~55 nm SiO2 cap layer is deposited by PECVD to improve the wettability of the surface and to provide an additional etch mask layer for the a-Si nanodisk etching.

CL was performed using the self-assembly of SiO2 spheres for surface patterning. Substrate pieces of ~2 cm² were prepared and oxygen plasma treatment (O2 flow of 500 sccm, RF power of 1 kW and process time of 10 min) of the surface was done to improve the monolayer coverage of the SiO2 spheres. A SiO2 colloidal particle solution (Sigma Aldrich; 500 nm (5 wt%) in H2O) was deposited on the surface and a mild spin coating process resulted in the self-assembly of the SiO2 colloidal particles in a hexagonally close-packed array coverage (Fig. 2(a)) in several mm²-sized patches, where the size of the particles determines the hexagonal array period of the a-Si nanodisk arrays. For obtaining larger (>1 cm²) single monolayer area coverages, optimization of the CL monolayer coverage is required.

To vary the nanodisk diameter, the self-assembled SiO2 spheres were size reduced (Fig. 2(b)) by reactive-ion etching (RIE) using a CHF3/Ar-based chemistry (CHF3/Ar flow of 20/10 sccm, RF power of 200 W and pressure of 40 mTorr). The ‘vertical’ etch rate of the SiO2 spheres was approximately ~20 nm/min. An estimate for the resulting diameter, based on the vertical etch rate, is reported in [29]. Simultaneously, the exposed PECVD SiO2 thin film layer will be etched (etch rate of ~20 nm/min), resulting in a circularly shaped hard mask underneath the SiO2 colloidal particles. The extra hard mask layer avoids undesired etching at the top part of the nanodisks; caused by the relatively small contact point of the colloidal particle with the underlying surface. In Fig. 5 (Appendix A) schematics and representative SEM images are included, indicating the influence on the etch shape when the additional hard mask layer is absent.
The etching of the a-Si nanodisk arrays was done by ICP-RIE (STS Multiplex ICP DRIE) based on an optimized variant of the Pseudo Bosch process (SF₆/C₄F₈/Ar flow of 21/48/5 sccm, ICP power of 600 W, RF power of 10 W, pressure of 10 mTorr and etch rate of ~40 nm/min) in order to obtain vertical disk walls. A representative SEM image of the etched structures is shown in Fig. 2(c). Due to slight chemical etching (etch rate=5 nm/min) in the lateral direction, a slightly smaller diameter for the a-Si nanodisks was obtained than the diameter of the SiO₂ etch mask. By this method a-Si nanodisk arrays were fabricated with a hexagonal array period of ~500 nm; the a-Si nanodisk height is 200 nm as in the vertical layer structure, but the diameter was varied between ~180-380 nm by size reducing the SiO₂ spheres. For a specific targeted diameter fabrication, the obtained nanodisks had a variation in diameter, typically ±30 nm, which we believe is primarily due to the size dispersion of the SiO₂ colloidal particles. To make the a-Si nanodisks substrate-free, a HF-treatment (HF5%) was applied to subsequently etch away the underlying sacrificial SiO₂ layer and to remove the SiO₂ etch mask at the same time. To maintain the hexagonal array period of the nanodisks, movement of the nanodisks during wet chemical processing steps should be prevented. Due to the removal of the sacrificial layer, the nanodisks drop to the substrate surface and are kept in place by Van der Waals forces. By limiting the forces on the nanodisks during dilution and drying procedures, lateral movement of the nanodisks can be limited. SEM imaging showed the hexagonal ordering to a reasonable extent; the inter-disk spacing varies typically ±100 nm. However, from a manufacturing perspective this fabrication step must be made more robust for example by developing suitable selective dry etching techniques. Figures 2(d) and 2(e), respectively, show representative top and side view SEM images of the fabricated substrate-free a-Si nanodisks. These nanodisk arrays were embedded in PDMS (Dow Corning; Sylgard 184 Silicone Elastomer) and subsequently lifted-off from the substrate.
(Figs. 1(e)-1(g)). A representative SEM image of the embedded nanodisks is shown in Fig. 2(f).

![SEM images of the fabrication process steps.](image)

**Fig. 2.** Scanning electron microscopy (SEM) images of the fabrication process steps. (a) Colloidal lithography resulting in a close-packed hexagonal array of SiO$_2$ colloidal particles (diameter: ~500 nm). (b) Fabrication of the etch mask by reactive ion etching (RIE) based on a CHF$_3$/Ar chemistry. (c) Inductively coupled plasma reactive-ion etching (ICP-RIE) of the a-Si nanodisk array by a SF$_6$/C$_4$F$_8$/Ar-based chemistry. (d) Top-view of a substrate-free a-Si nanodisk array on the c-Si surface. (e) Side-view of a substrate-free a-Si nanodisk array structures. (f) Top-view of embedded a-Si nanodisk structures in PDMS.

3. Optical properties of the a-Si nanodisk-based metasurfaces

Lumerical’s finite-difference time-domain (FDTD) simulation tool was used to investigate the reflectivity and transmission properties for a-Si nanodisk array metasurfaces on c-Si and embedded in PDMS, respectively. The geometrical properties of the a-Si nanodisk array structures are similar to those fabricated. These studies were performed to investigate the tuning of the optical properties by varying the nanodisk diameter. The values for the height and hexagonal array period were taken as 200 and 500 nm, respectively. The optical constants for a-Si and c-Si were taken from Palik [30]. The RI of a 200 nm PECVD deposited a-Si layer was determined by ellipsometry and resulted in a measured value of ~3.63 (in the NIR range). This value agrees well with the optical constants taken from literature [30]. The RI data for
PDMS was taken from the Dow Corning product information and the small (to no) absorption in PDMS was neglected for the wavelength range that was used. The c-Si substrate and the PDMS slab were taken as semi-infinite, where the a-Si nanodisk array on c-Si is in direct contact with the substrate surface and the top surface of the a-Si nanodisks embedded in PDMS are on the same plane as the PDMS surface. Both those geometries mimic the fabricated structures. Using a plane wave source and periodic boundary conditions, the total reflectivity and transmission was determined for the wavelength range of 400-1000 nm. Polarization effects can be neglected for both the measurements and simulations due to the (near) normal incidence angle of the incident light source. Though, it should be noted that polarization effects should be taken into account when using angular incidence angles.

The fabricated substrate-free a-Si nanodisk arrays on c-Si surfaces and those embedded in PDMS were optically characterized by spectrally-resolved reflectivity and transmission measurements. As discussed earlier, the coverage of the a-Si nanodisks on the sample surface resulted in fields of uniform coverage of typically several mm²-sized patches due to colloidal lithography influences. Thus, to overcome inconsistencies a home-built small spot setup was used to investigate the (embedded) nanodisk arrays for the wavelength range 400-1000 nm. Due to different experimental configurations, the spot size was ~350 $\mu$m² for the specular reflectance measurements (microscope built-in lamp, Andor Shamrock spectrophotometer and Andor Newton CCD; NA = 0.13) and ~900 $\mu$m² for the specular transmittance measurements (supercontinuum source, fiber setup, lens system and fiber-coupled (diameter = 600 $\mu$m) AvaSpec spectrometer.

The surface reflectivity was measured and simulated for the a-Si nanodisk arrays on a c-Si surface. The obtained data confirms the broadband anti-reflection application of this type of metasurfaces where average surface reflections as low as ~7.5% were obtained, which are comparable to the results reported in [2]; where additionally the omnidirectional broadband anti-reflection property of this type of structuring was shown. It should be noted that the obtained surface reflection is not as low as values reported for the state-of-the-art anti-reflection by direct structuring [31]. Though, an additional dielectric layer coating can further reduce the surface reflections. Additionally, the data show a redshift of the reflectivity dips for increasing disk diameters due to the shifting of the Mie resonances to longer wavelengths [2–6]. The measurements show a good agreement with the simulation data obtained for a-Si nanodisk arrays similar to the fabricated structures. The geometries (nanodisk height of 200 nm and nanodisk diameters of 150-400 nm) and spacing (hexagonal array period of 500 nm) used in this work indicate applications for broadband anti-reflection applications for the spectral range of ~550 to >1000 nm, when placed on a c-Si substrate surface. However, a-Si nanodisks absorb in the visible wavelength spectrum and thus it would be more desirable to use transparent anti-reflection metasurface materials (e.g., TiO$_2$). Since the anti-reflection application is not the main focus of this work, the results are implemented in Fig. 6 (Appendix B).

Figures 3(a)-3(c) show the simulation results for the transmittance of the a-Si nanodisk arrays (hexagonal array period of 500 nm and height of 200 nm) embedded in PDMS, where the disk diameter is varied between 150 and 400 nm. Figure 3(a) presents a contour plot (increasing diameter with a step size of 5 nm) for the total transmittance in which the relevant transmittance peak and dip shifts are indicated. Figure 3(b) shows the data taken from the contour plot for a nanodisk diameter of 300 nm where the transmittance peaks and dips are labelled accordingly. Several transmittance peaks are observed which are labelled as T1-4, respectively, and transmittance dips as Tdip1(a&b) and Tdip2(a&b). Tdip1 represents an overlap of Tdip1a and Tdip1b. Tdip2 represents an overlap of Tdip2a and Tdip2b. Figure 3(c) shows the simulated transmittance data taken from Fig. 3(a), indicating the shift of the peaks and dips for nanodisk arrays similar to the fabricated structures. A clear transmittance peak (T1) is observed with a peak value of ~60% and shifts from ~675 to ~850 nm as the nanodisk diameter increases from 200 to 350 nm.
Figure 3(d) shows the measured specular transmittance data for the a-Si nanodisk arrays embedded in PDMS for which the relevant peaks and dips are labelled accordingly. For the optical characterization, nanodisk diameters were taken based on diameter values (~275-375 nm) determined by the simulations. For these disk diameters a single peak occurs without a high shoulder at either side of the peak. The experimental results show a clear peak (combination of T1&T2), located between two transmittance dips (Tdip1b and Tdip2), with a peak value of ~23-25% and shifts from ~800 to ~850 nm as the nanodisk diameter increases from ~300 to ~380 nm. The lower measured peak value compared to the simulation data is most likely due to a higher fill factor (smaller inter-disk spacing) of the disks compared to the 500 nm hexagonal array spacing used in the simulations. As mentioned before, the inter-disk spacing shows a variation of ±100 nm. A higher fill factor results in the reduction of the overall transmission and blue-shifts the peak. The effect of the variation of the inter-disk or inter-sphere spacing on the shift/coupling of the Mie resonances has been reported in [25,32]. Supporting data regarding the influence of the inter-disk spacing variation for the a-Si nanodisk arrays is shown in Fig. 7 (Appendix C). Furthermore, specular (measured) vs total (simulation) transmittance influences should be taken into account. Tables 1 and 2 show the...
Table 1. Measured data for the transmittance peaks/dips for a-Si nanodisks in PDMS

| Disk Diameter [nm] | Wavelength Range [nm] | Average Shift [nm/nm > diameter] |
|-------------------|-----------------------|----------------------------------|
| Tdip1a            | ~300-380              | ~600-620                         | ~0.21                              |
| Tdip2             | ~300-380              | ~910-960                         | ~0.59                              |
| T1&T2             | ~300-380              | ~790-840                         | ~0.58                              |

Table 2. Simulation data for the transmittance peaks/dips for a-Si nanodisks in PDMS

| Disk Diameter [nm] | Wavelength Range [nm] | Average Shift [nm/nm > diameter] |
|-------------------|-----------------------|----------------------------------|
| Tdip1             | 150-200               | ~600-615                         | ~0.34                              |
| Tdip1a            | 250-400               | ~615-650                         | ~0.25                              |
| Tdip1b            | 250-400               | ~680-830                         | ~0.93                              |
| Tdip2             | 350-450               | ~1040-1160                       | ~0.60                              |
| Tdip2a            | 150-300               | ~680-930                         | ~1.63                              |
| Tdip2b            | 150-300               | ~750-980                         | ~1.50                              |
| T1                | 150-350               | ~615-840                         | ~1.50                              |
| T2                | 350-400               | ~840-900                         | ~1.07                              |

determined values for the shifts of the relevant transmission peaks and dips with regard to the diameter variation and occurring wavelength range for the measured and simulated data, respectively. This data shows an agreement between the measured and simulation data for similar structures.

Figure 4(a) shows a schematic sketch of the embedded a-Si nanodisk array in PDMS and Fig. 4(b) the simulated reflectivity, transmission and absorption spectra of the structure. The hexagonal array has a period of 500 nm; the nanodisk height and diameter are 200 and 300 nm, respectively. Figure 4(b) indicates that the transmittance dips (Tdip) can be related to the absorption peaks for the nanodisk arrays embedded in PDMS and are thus an indirect indication for the Mie resonances. The relevant absorption peaks are labelled as peak A1-4, along with the relevant reflectance peaks (R1-3) and transmittance peaks (T1-4). It can be observed that Tdip1a is related to peak A4, Tdip1b to peak A3, Tdip2a to peak A2 and Tdip2b to peak A1. Where peaks A1 and A2 are due to ED and MD modes, respectively; A3 and A4 are due to MQ and EQ modes, respectively (see also Fig. 8 (Appendix D)) [2–6]. Tdip2a&amp;b is additionally overlapping with a strong reflectance peak R1, though the location and shift of the small dips alongside peak T3 are still related to the peaks and shifts of A1 and A2.

Peak T1 or T2 (both a peak value of ~60%) is located between a combination of peaks A3&amp;A4 and A1&amp;A2&amp;R1(or R2), where peak R1 (or R2) can be assigned to a strong reflection (back scattering) peak (see Fig. 4(b)). Peak T3 (peak value of ~30%) is located between peaks A1 and A2 and disappears when both peaks spectrally overlap. Peak T4 (peak value of ~12%) is located between peaks A3 and A4 and disappears when both peaks spectrally overlap. Peak T1 (or T2) is interesting for optical (color/NIR) filter applications for nanodisk diameters varying between ~275-375 nm due to the single peak with relatively high transmittance (see Figs. 3(a) and 3(c)). The average peak shift for peak T1 has been determined to be ~1.50 nm/nm diameter increase for disk diameters increasing from 150 to 350 nm. This indicates that the spectral location of this peak can be tuned.

The occurrence of a strong reflection peak (R1) was also mentioned in previous works on Si nanodisk arrays [12–17] and can be used for optical (color) filter applications in the reflection mode. Detailed simulation data regarding the a-Si nanodisk diameter influence on the reflectivity data is added in Fig. 9 (Appendix E). For nanodisk diameters ranging from 150 to 400 nm three reflectance peaks can be distinguished: R1, R2 and R3 (see Fig. 4(b)). Peak R1 (peak value of ~75-80%) shifts from ~725 to >1000 nm for nanodisk diameters increasing from 150 to 325 nm with an average shift rate of ~1.52 nm/nm diameter increase.
Peak R2 (peak value of ~80%) shifts from ~775 to >1000 nm for nanodisk diameters increasing from 275 to 400 nm with an average shift rate of ~1.42 nm/nm diameter increase. Peak R3 (peak value of ~20%) shifts from ~625 to ~725 nm for nanodisk diameters increasing from 275 to 400 nm with an average shift of ~0.48 nm/nm diameter increase. The result shows a single peak for R1 (or R2) with a relatively high reflectance and thus is interesting for optical filter applications, in reflectance mode, in the NIR wavelength range.

Figure 4(c) shows the average shifts and occurring wavelength range obtained from the simulation data for the relevant reflectance (R1 & R2) and transmittance (T1 & T2) peaks. The average shifts of the relevant absorption peaks (A1-3) are added which are due to Mie resonances. This data indicates the tunability of peak R1 (or R2) and T1 (or T2) by varying the nanodisk diameter, which is directly related to the shifts of the occurring Mie resonances.

The measurement and simulation data show that the optical properties of the a-Si nanodisk arrays (on c-Si or embedded in PDMS) can be tuned by varying the diameter of the nanodisks. When embedded in PDMS, the a-Si nanodisk array metasurfaces show very interesting optical characteristics directly relevant for optical filter applications in the (visible-)NIR region, both in reflectance and transmittance modes. Figure 4(d) shows data taken from Fig. 4(b) and highlights the relevant reflectance (R1) and transmittance (T1) peaks for optical filter applications. Both these peaks can be tuned in the (visible-)NIR wavelength region.
4. Conclusion

In this work metasurfaces based on high refractive index amorphous silicon (a-Si) nanodisk arrays were fabricated and embedded in a flexible and transparent substrate (e.g., polydimethylsiloxane (PDMS)), using a low cost fabrication process.

Well-defined substrate-free a-Si nanodisk array metasurfaces were successfully fabricated by a combination of colloidal lithography (CL) and inductively coupled plasma reactive-ion etching (ICP-RIE), with a hexagonal array period of ~500 nm, a height of 200 nm and with diameters varying between ~180-380 nm. These nanodisk structures were subsequently embedded in PDMS to investigate their optical properties depending on their disk diameter.

Finite-difference time-domain (FDTD) simulations were used to design and investigate the optical properties of the fabricated a-Si nanodisk array structures in the visible-NIR range. Mie resonances were determined by tracking the transmittance dips in the obtained transmission spectra. Experimental transmission spectra showed transmission dips with a good agreement on the spectral location of the occurring Mie resonances and their dependence on the nanodisk diameter. The results show that a high transmission window can be obtained between these dips and additionally the occurrence of a high reflectivity window. For a nanodisk diameter range of 150-350 nm, a strong transmittance peak is observed between two transmittance dips with an average peak value of ~60% and shifts from ~600 to ~950 nm with a shift rate of ~1.50 nm/nm diameter increase. For the nanodisk diameter range of 150-400 nm, a reflectance peak can additionally be observed having an average peak value of ~75% and shifts from ~725 to >1000 nm with a shift rate of ~1.52 nm/nm diameter increase.

The obtained results in this work show that metasurfaces based on a-Si nanodisk arrays are interesting for (omnidirectional) broadband anti-reflection applications as well as for optical filter applications for the (visible-)NIR wavelength region. Furthermore, these nanodisk-based metasurfaces could be very interesting for both large-scale as well as micro/nanoscale coloring applications.

Appendix A: The influence of an additional hard mask for colloidal lithography

Fig. 5. Influence of the additional hard mask layer, below the hexagonal array of the size reduced SiO₂ spheres, on the shape of the etched structures by inductively coupled plasma reactive-ion etching (ICP-RIE). (a) Schematic showing the influence of the etched structures without the additional hard mask layer. (b) Schematic showing the etched a-Si nanodisk structures due to the additional hard mask layer. (c) Scanning electron microscopy (SEM) images showing the influence on the etched structures when the additional hard mask layer is absent (from left to right: 1 to 4 minutes ICP-RIE time with a time-step of 1 minute); the scale bar is shown in red.
Appendix B: Measurement and simulations for a-Si nanodisks on a c-Si substrate

Fig. 6. Reflectivity data for a-Si nanodisk arrays on a c-Si surface with a hexagonal array period of 500 nm, height of 200 nm and varying nanodisk diameter. (a) Schematic of the nanodisk-based metasurface for (omnidirectional) broadband anti-reflection applications. (b) The measured specular reflectivity data for the fabricated structures with nanodisk diameters increasing from ~180 to ~270 nm. The relevant dips are labelled accordingly for the data shown for the nanodisk diameter of 220 nm. A redshift of the reflectance dips (Si-Rdip2 and Si-Rdip3) is observed for an increasing nanodisk diameter. (c) Contour plot (diameter step of 5 nm) showing the total reflectance obtained by FDTD simulations for nanodisk diameters varying between 150 and 400 nm. The shifts of the reflectance dips (Si-Rdip1-3) and location of the periodicity dip are indicated with a white dashed line and labelled accordingly. Optical constants for a-Si and c-Si have been taken from Palik [30]. (d) Data taken from (c) showing the redshift of the relevant reflectance dips (Si-Rdip2 and Si-Rdip3) for nanodisk diameters increasing from 200 to 300 nm. The relevant dips are labelled accordingly for the data shown related to a nanodisk diameter of 300 nm.
Appendix C: Inter-disk spacing FDTD simulations for a-Si nanodisk arrays on c-Si and embedded in PDMS

Fig. 7. FDTD simulations regarding the influence of the inter-disk spacing on the reflectivity and transmission spectra for a-Si nanodisk arrays with a nanodisk height of 200 nm, diameter of 300 nm and varying hexagonal array period (inter-disk spacing). Optical constants for a-Si and c-Si have been taken from Palik [30]. Optical constants for PDMS were taken from the Dow Corning product information. (a) Contour plot of the reflectivity data with regard to the hexagonal array period (inter-disk spacing = period-diameter; step size of 50 nm) for a-Si nanodisk arrays on a c-Si surface. The shifts of the reflectivity dips (Si-Rdip1-3) and the periodicity dip are indicated with white dashed lines and labelled accordingly. (b) Data taken from (a) showing the redshift of the reflectivity dips for an increasing inter-disk spacing where the reflectivity dips are labelled accordingly. (c) Contour plot of the transmission data with regard to the hexagonal array period (inter-disk spacing = period-diameter; step size of 50 nm) for a-Si nanodisk arrays embedded in PDMS. The shifts of the transmission peak (T1), dips (Tdip1b and Tdip2) and the periodicity dip are indicated with white dashed lines and labelled accordingly. (d) Data taken from (c) showing the redshift of the transmittance peak (T1) for increasing inter-disk spacing where the transmission peak is labelled accordingly.
Appendix D: The E- and H-field plots for a-Si nanodisk arrays embedded in PDMS obtained by FDTD simulations

| Embedded in PDMS | X-Field Monitor | Y-Field Monitor |
|------------------|-----------------|-----------------|
|                  | E-field         | H-field         | E-field         | H-field         |
| A1               | ![Image](image1) | ![Image](image2) | ![Image](image3) | ![Image](image4) |
| A2               | ![Image](image5) | ![Image](image6) | ![Image](image7) | ![Image](image8) |
| A3               | ![Image](image9) | ![Image](image10) | ![Image](image11) | ![Image](image12) |
| A4               | ![Image](image13) | ![Image](image14) | ![Image](image15) | ![Image](image16) |

Fig. 8. Cross-section plots of the E- and H-field profiles for the wavelengths associated to the absorption peaks A1-4 for a-Si nanodisk arrays embedded in PDMS; with a hexagonal array period of 500 nm, height of 200 nm and diameter of 300 nm. The cross-sections are taken along the Z-axis of the unit cell and through the middle of the nanodisk. The field profiles are shown for both the X- and Y-field monitors, respectively, where the X- and Y-axis span the horizontal plane of the simulated unit cell. The plots are obtained for a 0° (E_x) polarized source at a normal (vertical) incidence angle. It should be noted that the plots show the sum of the incident and scattered field components.
Appendix E: Reflectivity FDTD simulations for a-Si nanodisk arrays embedded in PDMS

Fig. 9. Reflectivity data for a-Si nanodisk arrays embedded in PDMS with a hexagonal array period of 500 nm, height of 200 nm and varying nanodisk diameter. Optical constants for a-Si have been taken from Palik [30]. Optical constants for PDMS were taken from the Dow Corning product information. (a) Contour plot showing the total reflectivity data obtained by FDTD simulations for nanodisk diameters varying between 150 and 450 nm. The shifts of the reflectivity peaks (R1-4) are indicated with a white dashed line and labelled accordingly. (b) Data taken from (a) showing the redshift of the relevant reflectance peak (R1 or R1&R2) for nanodisk diameters increasing from 200 to 400 nm.

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