Summary

Photovoltaics (PV) has the potential to become one of the major energy sources in a future with carbon-neutral energy supply. Two key aspects to achieve large-scale implementation of PV in our society are the seamless integration into buildings and infrastructure and a further increase of photovoltaic conversion efficiency. Integration of PV in our environment requires the development of solar cells with a colorful appearance. An increase of the efficiency in PV can provide a reduction in cost per generated power, as well as the demanded area required for the installation of PV systems. Achieving high efficiency requires the development of multijunction solar cells composed of multiple materials.

In this thesis we develop dielectric nanostructures to create tailored colorful appearance of solar cells and show how similar structures can increase the efficiency in tandem cells. For this, we design layers composed of dielectric nanoparticles that show strong light scattering in the visible spectral range. In particular, we develop dielectric metasurfaces and combined (gradient) metagratings that enable the control of spectrum and directivity of scattered light. In the first part of the thesis, we present novel ways to realize coloration of PV (Chapter 2-5), and design metasurfaces with tailored spectral and angular control of light (Chapter 4, 6). In the second part of the thesis, we theoretically and experimentally develop metasurface spectral splitters to improve the performance of two-terminal and four-terminal perovskite/silicon tandem solar cells (Chapter 7, 8).

In Chapter 2 we integrate crystalline Si nanocylinder arrays (radius = 100–120 nm, height = 240 nm, period = 325 nm) in the front module layer of mini silicon PV modules and demonstrate efficient coloration of the modules. The characteristic Mie resonances in the visible of the nanocylinder cause strong (35-40%) specular light scattering around a wavelength of λ = 540 nm and create a green coloration of the module that is quite independent from incoming angles over the 8–75° angular range. The colored module only shows a reduction of 10–11% in short-circuit current and efficiency. The green solar modules presented here can find applications in building- and landscape-integrated photovoltaics of many different kinds. For the fabrication we use the soft imprint conformal lithography (SCIL) nano-imprint technique that is applicable to full-area (15 x 15 cm^2) solar cell patterning.

Chapter 3 expands the technology of Chapter 2 to an increased color space. By changing the size and the distribution of the scattering nanocylinders (radius = 50–150 nm, period = 160-300 nm) we control the interference of electric and magnetic dipoles to create resonant scattering over a distinct spectral band. Pure colors from blue to red can be created with a scattering efficiency of up to 75%. The technique is extended by creating mixed colors (e.g. white) by combined pixels of nanocylinder arrays. We fabricate a white solar mini-module that shows a current loss of only 19%, due to the fact that only light in a well-defined visible spectral band is reflected. The route of combining differently structured metasurface pixels shows a great flexibility in creating colors by choice

and demonstrates the large potential for this technique to be widely applied.

In Chapter 4 we demonstrate metasurfaces composed of combined metagratings with tailored angular scattering profiles of Si Mie resonators. We control the spectral response by tailoring the interference of the modes of the nanoparticles and arrange them into patches of different periodicities to control the directivity of the scattering into diffraction orders. Based on a theoretical dipole scattering model, we designed these ultrathin surfaces (height = 185 nm) to realize broad-angle (35-75°) and Lambertian-like large-angle (0-75°) scattering profiles on resonance (λ = 650 nm). Specular reflection is canceled out by destructive interference of the scattering of the particles and reflection of the substrate, and high-order diffraction modes are strongly suppressed by shaping the scattering of a unit cell with several nanoparticles. Experimentally, the metasurfaces are composed of Si nanobars on a Ag mirror protected by a thin dielectric layer, and reach scattering efficiencies of 73-84%. The ultrathin resonant metagratings effectively create the functionality of “nanostructured paint”, which can find applications in photovoltaics with colored appearance and angular behavior by choice as demonstrated in Chapter 5. Similarly, the presented metasurfaces can serve as spectrum splitting architectures in tandem solar cells as described in Chapter 7 and 8.

In Chapter 5 we follow up on Chapter 4 by experimentally demonstrating spectral and angular control of scattered light using combined resonant metagratings that create light scattering only for a determined set of angles. By tailoring the interference between the light scattering modes of the nanoparticles we control the scattering spectrum, and by arranging them into patches of different periodicities, we control the directivity. Silicon nanowires were fabricated on a transparent substrate and resonantly scatter light around λ = 650 nm. The nanowires were placed in a supercell structure composed of multiple gratings with different pitches (675-1300 nm), creating a wide-angle scattering distribution within the 30-75° range. The metagratings are designed to suppress specular refection and show efficient transmission outside the resonant scattering band. The metagratings are integrated with a silicon heterojunction solar cell creating a bright red scattering appearance under a limited range of angles. The external quantum efficiency is only reduced in the scattering spectral band, with a short-circuit current drop of only 13%. This structure can find application in rooftop photovoltaics and other building integrated PV that benefits from light reflection towards a well-defined angular range.

Chapter 6 shows the theory, design and experimental realization of a broadband back reflector metasurface based on the Huygens-Fresnel principle. Using a phase-gradient approach we design a theoretical metasurface with unity efficiency for back reflection. By creating a periodic phase change of light reflected from the surface, light is efficiently channeled into the first negative diffraction order over a broad angular range and over bandwidth in the visible spectral range. In the experimental realization, the calculated phase gradient is discretized and arrays are formed by unit cells of TiO_2 nanowires of different height, placed on a Ag mirror. This metasurface allows back reflection for a broad range of wavelengths from λ = 490 - 940 nm with an efficiency above 85%. The advantages of the design lie in its low profile, simplicity of design, broad bandwidth and wide acceptance angular range. The ultrathin metasurface can replace more bulky back reflectors and find application in many optical technologies.

In Chapter 7 we theoretically study the benefits of a spectral splitter integrated into a perovskite/silicon two-terminal (2T) tandem solar cell. The spectral splitter reflects light with energy above the bandgap energy of the top cell and therefore enhances the absorption in the perovskite to benefit from the higher photovoltage in the top cell. We distinguish between a planar and a Lambertian scattering spectral splitter and find that by choosing the right splitting condition for the strength and spectral onset of the reflectivity, both spectral splitter configurations are beneficial for 2T tandem cells. For top cell materials with bandgaps above 1.7 eV, a spectral splitter is always beneficial as it enhances absorption in the top cell and creates better current matching with the bottom cell. Detailed-balance calculations show that including a Lambertian spectral splitter interlayer for 500 nm thick perovskite top cells with bandgap higher than 1.7 eV leads to 5%-6% absolute efficiency gain. Using experimental parameters of realistic cells, we predict a potential efficiency gain of 2.8% (absolute) for a top cell with a bandgap energy of 1.77 eV.

In Chapter 8 we experimentally demonstrate a spectral splitting light trapping metagrating as an interlayer in 4T perovskite/silicon tandem cells. This metagrating has the property to efficiently diffract light with energies above the bandgap of the perovskite into the top cell under high angles and thus create light trapping of the reflected light in the perovskite top cell. At the same time, the spectral splitter metasurface reduces reflection in the infrared and therefore enhances photocurrent generation in the silicon bottom cell. We fabricate the metasurface spectrum splitter using SCIL and integrate it in a perovskite/silicon 4T tandem cell, and demonstrate a current increase in the top cell of 0.5 mA/cm^2. We predict an efficiency gain for the fabricated geometry in a wafer-sized 4T perovskite/silicon tandem solar cell of 0.26% (absolute). We present a further optimized metasurface design for which we predict a 1.4 mA/cm^2 enhancement in short-circuit current in the perovskite top cell and an efficiency enhancement of the 4T tandem solar cell of 0.40% (absolute). The metagrating geometry is very flexible in design and can be adjusted to other top and bottom cell bandgap combinations. The spectrum splitting metagratings present an optimized balance between spectral splitting, light trapping and optimized transmission in the infrared.

Overall, this thesis provides novel solutions based on nanophotonic light scattering to create colored PV and tandem solar cells with enhanced efficiency. It employs control over the scattering properties of resonant dielectric nanoscatterers and metasurfaces to create desired scattering distributions that are tailored in angular and spectral range. The metagrating concepts for photovoltaics developed in this thesis can be applied on a wide range of solar cells and can be scaled up to practical large-area fabrication technologies.

List of Publications

This thesis is based on the following publications

- *Efficient colored silicon solar modules using integrated resonant dielectric nanoscatterers*, V. Neder, S. L. Luxembourg, and A. Polman, Appl. Phys. Lett. **111**, 073902 (2017). (**Chapter 2**)

- *Colored solar modules using integrated pixelated resonant dielectric nanoscatterer arrays*, V. Neder, S. L. Luxembourg, and A. Polman, Proc. 33^{rd} European Photovoltaic Solar Energy Conference and Exhibition, 34-37 (2017). (**Chapter 3**)

- *Combined metagratings for efficient broad-angle scattering metasurface*, V. Neder, Y. Ra'di, A. Alù and A. Polman, ACS Photonics **6**, 1010–1017 (2019). (**Chapter 4**)

- *Resonant metagratings for spectral and angular control of light for colored rooftop photovoltaics*, F. Uleman,V. Neder, A. Cordaro, A. Alù, and A. Polman, ACS Applied Energy Materials **3**, 3150-3156 (2020). (**Chapter 5**)

- *Visible light, wide-angle graded metasurface for back reflection*, V. Neder, E. M. Estakhri, M. W. Knight, A. Polman, and A. Alù, ACS Photonics **4**, 228-235 (2017). (**Chapter 6**)

- *Detailed-balance efficiency limits of two-terminal perovskite/silicon tandem solar cells with planar and Lambertian spectral splitters*, V. Neder, S. W. Tabernig, and A. Polman, preprint at https://arxiv.org/abs/2012.12636 (2020). (**Chapter 7**)

- *Four-terminal perovskite/silicon tandem solar cell with integrated Mie-resonant spectral splitter metagrating*, V. Neder, S. D. Zhang, S. Veenstra, and A. Polman, preprint at https://arxiv.org/abs/2012.12649 (2020). (**Chapter 8**)

Other publications by the author

- *Broadband angular colour stability of dielectric-coated pyramidal textured Si for photovoltaics*, N. Roosloot, V. Neder, H. Haug, C. C. You, A. Polman, and E. S. Marstein, in preparation

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About the author

Verena Neder was born in Schweinfurt, Germany, on April 16, 1991. She received her high school diploma in 2010 from the Jack-Steinberger Gymnasium in Bad Kissingen. She studied Physics at the Ruprecht-Karls University in Heidelberg and at the University of Amsterdam. She graduated in 2016 with cum laude, with a thesis on a gradient metasurface back reflector at AMOLF in Amsterdam under the supervision of prof. dr. Albert Polman. After her graduation she stayed at AMOLF and continued as a PhD student in the same group, working on dielectric metasurfaces for light management in photovoltaics. The results of her research are shown in this thesis. During her PhD in 2017 Verena spent one month at the University of Texas in Austin under the supervision of prof. dr. Andrea Alù, working on a Lambertian metagrating metasurface. In 2019 she went on a 4 month maternity leave after the birth of her daughter. In her free time Verena enjoys running, biking, gardening, cooking for friends and in a volunteer-run restaurant and inventing new activities with her family.