Theoretical and experimental analysis of the structural pattern responsible for the iridescence of *Morpho* butterflies

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Abstract: *Morpho* butterflies are well-known for their iridescence originating from nanostructures in the scales of their wings. These optical active structures integrate three design principles leading to the wide angle reflection: alternating lamellae layers, "Christmas tree" like shape, and offsets between neighboring ridges. We study their individual effects rigorously by 2D FEM simulations of the nanostructures of the *Morpho sulkowskyi* butterfly and show how the reflection spectrum can be controlled by the design of the nanostructures. The width of the spectrum is broad (≈ 90 nm) for alternating lamellae layers (or "brunches") of the structure while the "Christmas tree" pattern together with a height offset between neighboring ridges reduces the directionality of the reflectance. Furthermore, we fabricated the simulated structures by e-beam lithography. The resulting samples mimicked all important optical features of the original *Morpho* butterfly scales and feature the intense blue iridescence with a wide angular range of reflection.

OCIS codes: (350.3950) Micro-optics; (220.4241) Nanostructure fabrication; (310.6628) Subwavelength structures, nanostructures; (050.1755) Computational electromagnetic methods; (330.0330) Vision, color, and visual optics; (000.1430) Biology and medicine; (350.4238) Nanophotonics and photonic crystals.

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1. Introduction

Many insects feature resourceful structural colors [1–6]. Especially the famous iridescence of *Morpho* butterflies has attracted the attention of researchers for a long time. Several research

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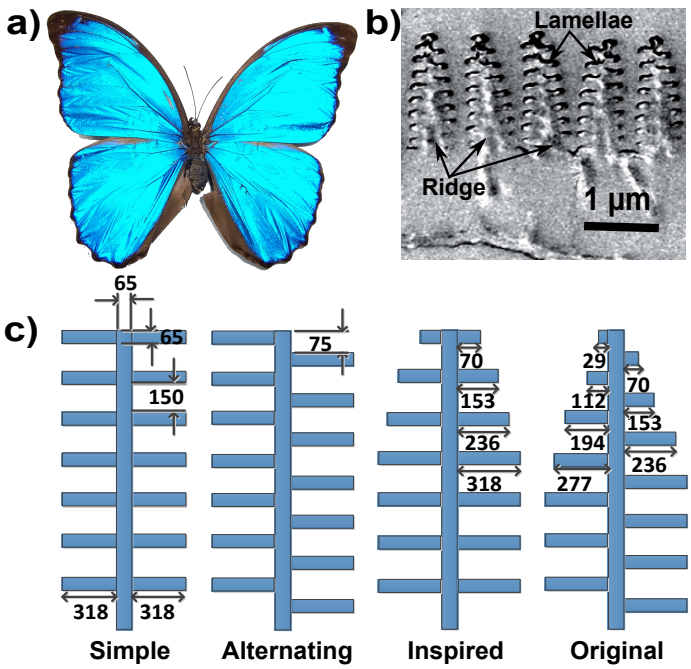

Fig. 1. (a) The wings of blue *Morpho* butterflies like *Morpho menelaus* show strong blue iridescence. (b) A TEM image of the cross-section of the *Morpho sulkowskyi* ground scale reveal the "Christmas tree" like structure responsible for the famous blue iridescence. Reproduced from Pris *et al.* [10]. (c) Schematics of the structures and dimensions (all units in nm) used in this study for simulation and fabrication. All values are extracted from the TEM and SEM images of *Morpho sulkowskyi* [9]. The 'simple' grating like structure resembles a simple stack of thin films. The structure shown next consists of 'alternating' lamellae layers without "Christmas tree" type shape. The 'inspired' structure is shaped like a "Christmas tree" without alternating layers. Finally, the 'original' *Morpho*-type structure features both properties: alternating layers together with "Christmas tree" like shape.

groups analyzed the architecture of the wings of *Morpho* butterflies in order to understand their blue or white-purple iridescence (Fig. 1(a)) and developed fabrication methods to replicate their design for various applications [7–11]. Potyrailo *et al.* [9], for example, revealed that the optical response of the nano-scale structures of the *Morpho* butterfly changes for different vapors. Therefore, they suggested that mimicking of this feature might outperform existing photonic vapor sensors. Recently, the same authors were inspired by the *Morpho* butterfly to design an artificial thermal sensor with high sensitivity and spatial resolution [10]. Furthermore, the *Morpho* butterfly architecture inspirited researchers to enhance the efficiency of solar cells [11–13].

It is commonly believed that the multilayer interference from the stack of lamellaes of regular periodic ridges on the scales (Fig. 1(b)) is the origin of the blue iridescence of the *Morpho* butterflies [14–16]. Kinoshita *et al.* explained the blue coloring of *Morpho* butterflies by the combination of interference and diffraction of light from ridges with irregular height differences in the sub-wavelength range in combination with pigmentation [17, 18]. Employing the same hypothesis, Saito *et al.* [7, 19] and Chung *et al.* [20] used multilayer deposition of TiO_2/SiO_2 on an irregular support to successfully mimic the wide angular reflectance of the blue color artificially. Recently, however, Chung *et al.* [21] found that chitin/air multilayers provide larger color

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gamut and better color stability against random structural variations than multilayers based on typical inorganic materials. Watanabe *et al.* [22] successfully fabricated replica of Morpho butterfly scales using focused-ion-beam chemical-vapor-deposition (FIB-CVD) and observed brilliant blue color refection. Aryal *et al.* [23] recently introduced a method for the large area nanofabrication and subsequent nanoimprinting of three dimensional butterfly structures.

Here, we analyze the three design principles causing the wide angle reflection observed in blue *Morpho* butterflies: alternating lamellae layers, "Christmas tree" like shape, and offsets between neighboring ridges. In order to study their individual effects rigorously, we simulate the optical properties of the nanostructures found in the *Morpho sulkowskyi* butterfly (see Fig. 1(b)) with the finite element method in two dimensions (2D FEM) and calculate the corresponding reflection of light. We observe that the reflection depends strongly on the design of the optical active structures. The width of the reflection peak is very broad (≈ 90 nm) for alternating lamellae layers (or "brunches") in the "Christmas tree" like structure. In addition, the reflection intensity for large angles is higher for this design.

In a second step, we accurately replicate the already simulated structures by e-beam lithography, especially the "Christmas tree" like structure of the *Morpho* butterfly. The optical spectra of the fabricated structures are in general agreement with our optical FEM simulations. The experimental results reveal that the blue iredescence is caused by the structural pattern of alternating lamellae layers ("brunches") within the "Christmas tree" like structure and the height differences between neighboring ridges. The experimental reflection spectrum for this design is broad (≈ 60 nm) and greatly effects the shift of the peak also for large viewing angle. Therefore, we conclude that the combination of these three design principles is responsible for the famous wide angle reflection of blue light with high intensity observed in blue *Morpho* butterflies.

2. Optical simulation

Several electromagnetic/optical approaches have been introduced already to analyze the scattering from butterfly scales to predict the observed blue color [9, 16, 17, 24–28]. The analytical approaches usually assume a simplified model for the original structure. Some of them model the whole scale as a stack of infinite long parallel plates with the effective refractive index alternating between high and low values [16]. The plane wave reflection coefficient for the stack is determined using a standard analysis for layered media. The simple transfer matrix method has been used in which the lamellae are treated as infinitely thin plates of finite width [17]. Other approaches used rigorous lamellar grating theory [24] and 3D ray tracing [9]. The most recent and accurate approaches utilize numerical methods like finite difference time domain (FDTD) for solving Maxwell's equations which can model the fine details of the structures [25–28]. Here, we apply a similar approach to model the complex butterfly structures using the finite element method (FEM) to solve Maxwell's equation with a commercial software package [29]. This approach is much less time consuming because of its flexible triangle-shaped mesh but as accurate as the other models mentioned before.

The computational domain for solving optical scattering measurements is illustrated in Fig. 2(a). The computational method starts with the definition of the parameters for solving the electromagnetic wave equation in the frequency domain

$$\nabla \times \mu_r^{-1}(\nabla \times \mathbf{E}) - k_0^2 \left(\varepsilon_r - \frac{j\sigma}{\omega \varepsilon_0} \right) \mathbf{E} = 0, \tag{1}$$

where k_0 is the wave number defined by $(2\pi/\lambda)$ and $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m is the electrical permittivity in free space. The relative magnetic permeability μ_r is equal to 1 as the magnetic permeability μ is taken to be everywhere equal to its free space value of $4\pi \times 10^{-7}$ H/m. The

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conductivity σ and the relative electrical permittivity ε_r are defined as [25]

$$\sigma = 2n_r n_i \omega, \tag{2}$$

$$\varepsilon_r = n_r^2 - n_i^2, \tag{3}$$

where n_r and n_i are the real and imaginary parts of complex refractive index n. Initially, we considered the complex refractive index n of the butterfly scales as $1.55 + i0.06$ [16]. Comparing the results with the reflectance spectra of the original butterfly *Morpho sulkowskyi* [9] we observe good agreement. However, in order to make the simulations comparable to the experimental results of our fabricated structures described in the next sections we use $n = 1.50$ for PMMA (Polymethyl methacrylate) at 450 nm [30] in the following. The maximum mesh element size was $1/10^{th}$ of the smallest wavelength of inspection (300 nm) at the interfaces and $1/5^{th}$ elsewhere.

The scattered field formulation is used to measure the scattering of light from the nanostructures mimicking the optical active parts of the butterfly scales. It is based on the pure scattered field formulation in which the total field is the sum of the known incident field and the unknown scattered fields [31]. Here, the incident field is defined as a plane transverse magnetic (TM) wave with unit amplitude and intensity, eliminating the need for normalizing the output further. In order to have a finite computational space, the perfectly matched layer (PML) technique [32] is applied by constructing a 300 nm wide PML absorber on top and bottom of the computational domain. Periodic boundary conditions (PBC) are utilized to simulate the large system by modeling a small unit cell containing two ridges of the butterfly structure. As it is a periodic cell, the length of the right and left side in the computational domain are taken as half of the distance between the two ridges. To reduce computational time calculations are done in the two dimensional $x - y$ surface of the 3D nanostructures. The near-zone scattered field on the top surface of the domain mainly determines the reflection intensity [26]. We calculate the scattered output power by integrating the square of the near-zone scattered field over the top boundary before the PML.

The simulated geometries start with a 'simple' stack of thin films of 65 nm with a gap of 150 nm and transform gradually to an 'alternating', to an 'inspired', and finally to the 'original' structure as shown in Fig. 1(c). The middle pillar (or "trunk") stabilizes the nanostructures to keep them free standing. Already the scattered electric field norm displayed in Fig. 2(b) reveals that the scattering behavior depends on the structure. The 'simple' and 'original' structure have a different scattering pattern at 490 nm.

The simulated reflection spectra are compared in Fig. 2(c) for all four structures. The reflection intensity is largest at 492 nm for the 'simple' structure and we defined this value as unity. The wavelength of the reflection peak follows directly from the thin film interference phenomena (see Eq. (1) in [33]). The first order reflection at normal incidence is obtained at $2(n_1 d_1 + n_2 d_2)$ which is 495 nm in our case ($n_1 = 1.50, d_1 = 65$ nm, $n_2 = 1, d_2 = 150$ nm). This result proofs the accuracy of our FEM simulations by a comparison with well established physics. Shortening the upper lamellae layers (or "brunches") of the 'simple' structure results in an 'inspired' "Christmas tree" like structure but reduces the intensity to 60% compared to the simple structure. Obviously, this 'inspired' structure does not mimic the features found in *Morpho* butterflies. However, introducing an offset of 75 nm into the right portion of the ridges of the 'simple' structure creates an 'alternating' lamellae pattern with a much broader reflection spectrum with two peaks at 425 nm and 492 nm (see Fig. 2(b), green dotted line). The reflection intensity, however, is reduced as the reflection energy is distributed over a wide range of frequencies. On the other hand, shaping this 'alternating' pattern into "Christmas tree" type shape gives the 'original' pattern and increases the intensity to 60% again (see Fig. 2(b), blue dashed line). This result is comparable with previous simulations [9] assuming structures of the same

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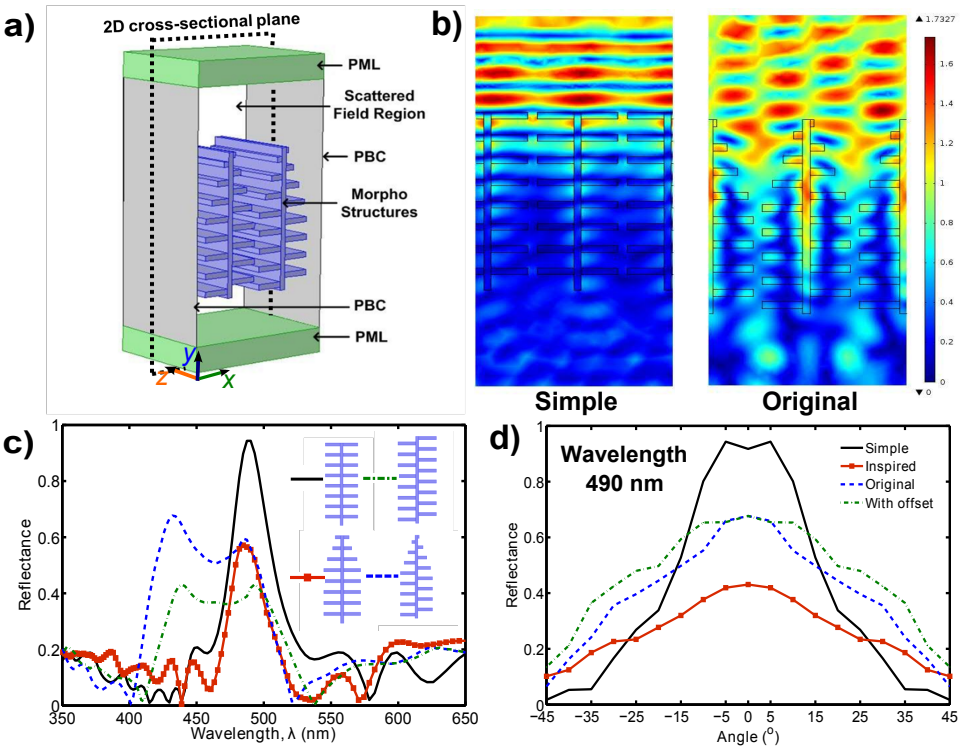

Fig. 2. Optical FEM simulations of *Morpho* butterfly nanostructures. (a) 3D Schematic of the periodic cell assumed in the simulations. The dotted cross-sectional plane in the schematic corresponds to the 2D simulation plane. Periodic boundary conditions are used on the left and right side of the cell to repeat the structures in horizontal direction. To confine the cell in top and bottom direction, perfectly matched layers are applied. Two ridges are considered to implement the offset between the structures. (b) A comparison of the scattered electric field norm at 490 nm in the $x-y$ plane for the 'simple' (left) and the 'original' (right) type structures with four ridges already reveal differences in the scattering behavior. (c) The reflectance is numerically calculated for different types of structures by measuring the scattered field power normalized by the incident power. The simulated structural patterns are displayed as insets. A broadening of the reflection spectrum is noticed for alternating layers of lamellae. The intensity peak, however, is highest for the simple grating like structure. (d) Simulations of the reflectance for variable incidence angles at 490 nm for the 'simple', 'inspired' and 'original' structures with and without an inclusion of 50 nm offset between successive ridges.

size. Therefore, considering the offset on the right portion of the ridges and the "Christmas tree" like shape creates the broad spectrum observed in blue *Morpho* butterflies.

All calculations presented so far did not consider the height differences between neighboring ridges frequently described in literature [7, 9, 17]. Adding an offset of 50 nm between neighboring ridges [9] of the 'original' design does not change the reflection at normal incidence but maintains higher reflection intensity in the blue region for higher incidence angles. The angle resolved reflection spectrum in Fig. 2(d) represents this dependency of the structural pattern on wide angle single blue color. The result clearly indicates the wide angular reflection property of original structure with an offset between neighboring ridges over the other structural pattern. The wavelength considered here is 490 nm as it shows highest reflection at normal incidence

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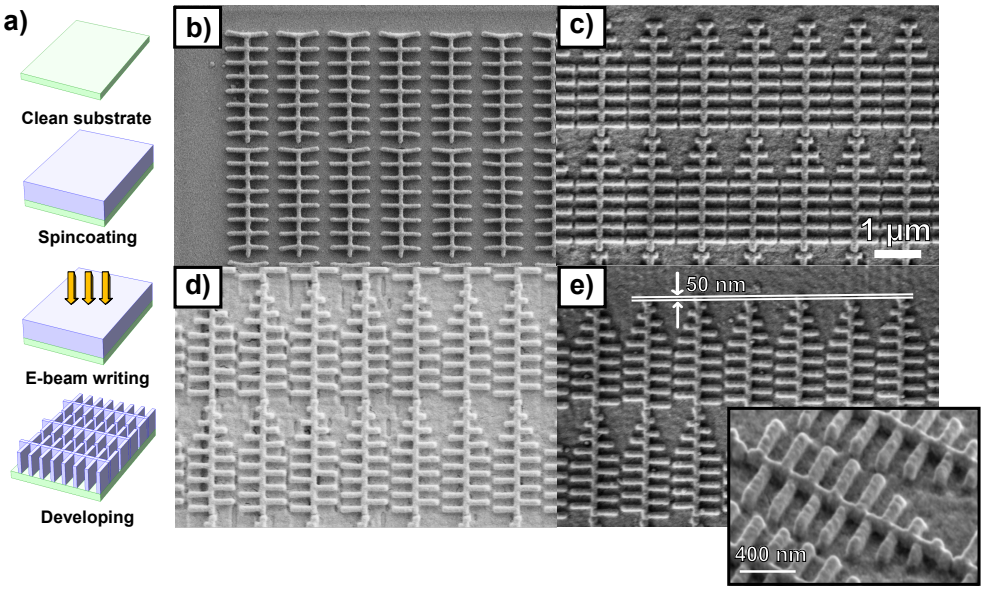

Fig. 3. Fabrication by e-beam lithography and corresponding SEM images of the Morpho inspired nanostructures . (a) The process flow diagram of the fabrication procedure. SEM images of (b) the 'simple' grating like structure with dimensions corresponding to the original butterfly; (c) the 'inspired' "Christmas tree" like structure without the alternating lamellae pattern; (d) the 'original' *Morpho* like structure with the dimension of the *Morpho sulkowskyi* butterfly, and, (e) the 'original' *Morpho* structure considering also the 50 nm offset between two neighboring ridges. The inset at the lower right shows a single "Christmas tree" structure from an inclined view angle.

for these structures.

3. Fabrication of Morpho-inspired nanostructures

Motivated by the results of our simulations, we fabricated real samples to prove our hypothesis that the "Christmas tree" like structure with alternating brunches and offsets between neighboring ridges is responsible for the famous blue iridescence. As the structures are extremely small, we applied e-beam lithography to replicate the structures with exactly the same dimensions already considered in the simulations. The fabrication steps are summarized in Fig. 3(a).

The fabrication procedure starts with spincoating of 200 nm PMMA (Polymethyl methacrylate, AR-P 672.045, 1550 rpm for 60 s with an acceleration of 150 rpm/s) on a clean Silicon wafer followed by soft baking (180° C for 5 minutes). As already mentioned in the previous section we used PMMA as material because it has a refractive index ($n = 1.50019$) comparable to butterfly scales ($n = 1.55$). A JEOL Electron beam writer is used for the structuring of the samples with a dosage of 300 μC/cm^2 considering the proximity effect correction. The lateral area of each structure is $500 \times 500\,\mu$m^2 large and written with a current of 100 pA. A frame area of 3×3 mm^2 is exposed with a current of 1000 pA in order to reduce the writing time. Overall, it takes 12 hours to write one complete structure. After writing the structure is developed for 45 s with MIBK:IPA (1:3) using an OPTIwet SB 30 system. Finally, the remaining PMMA outside the frame is mechanically removed to ease subsequent the optical characterization.

The SEM images in Fig. 3(b)-(e) display four different structures analyzed in the following: a 'simple' grating like structure (Fig. 3(b)), a "Christmas tree" like 'inspired' structure with-

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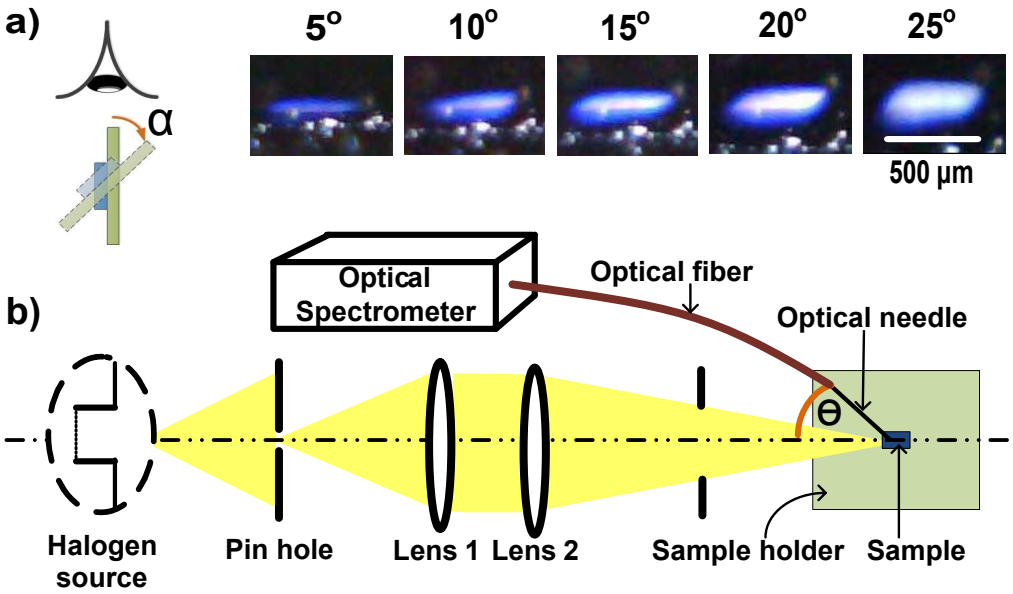

Fig. 4. (a) Photos of the blue reflection from the artificial *Morpho* structure with the inclusion of an offset between neighboring ridges. The illumination area of this sample is $500\,\mu m \times 200\,\text{nm}$ at an incidence angle of 0°. A bright reflection is still visible in the blue color regime for a tilting angle of 25°. Since the "Christmas trees" lay flat on the sample the size of the visible area increases with the tilting angle. (b) Schematic of the optical setup to measure the angle resolved reflectance quantitatively. A halogen lamp with an aperture to reduce the beam spot is used as the light source. It is followed by back to back lenses to focus the light on the sample. An optical needle with a diameter of 50 μm is used to collect the reflected light which is fed into an optical spectrometer for the subsequent analysis.

out alternating layers (Fig. 3(c)), one with exact 'original' dimension and alternating layers (Fig. 3(d)), and the final one with the dimension of the 'original' but 'with an offset' of 50 nm between the neighboring ridges (Fig. 3(e)).

4. Experimental results and analysis

The structures fabricated by e-beam lithography are not free standing like the ridges in the butterfly scales. Instead they lay flat on the Silicon wafer. Nonetheless, they still maintain the intense blue iridescence over a wide angular range. Figure 4(a) displays the artificial reflected blue color of the 'original structure with offset' (Fig. 3(e)). As seen on the photos the 'original structure with offset' maintains the high intense blue reflection for an angle up to 25° (realized by the tilting of the sample).

The optical setup [19] schematically drawn in Fig. 4(b)) is used to detect the reflective properties of all structures in more detail. A Halogen lamp with a pinhole to reduce the beam diameter serves as a source. A lens system focuses the light on the sample. The reflectance is measured through an optical needle (diameter of 50 μm) coupled into a fiber-coupled USB spectrometer (Ocean Optics HR2000+ with a span region of 394.12 nm - 840.11 nm and a resolution of 0.5 nm). The subsequent data analysis is done with Matlab together with the commercial software package SpectraSuite for data collection from the spectrometer. The optical needle is rotatable in the horizontal plane to measure angle resolved reflectance spectra. All measurements were taken in the dark to avoid possible stray lights from the surrounding. A reference

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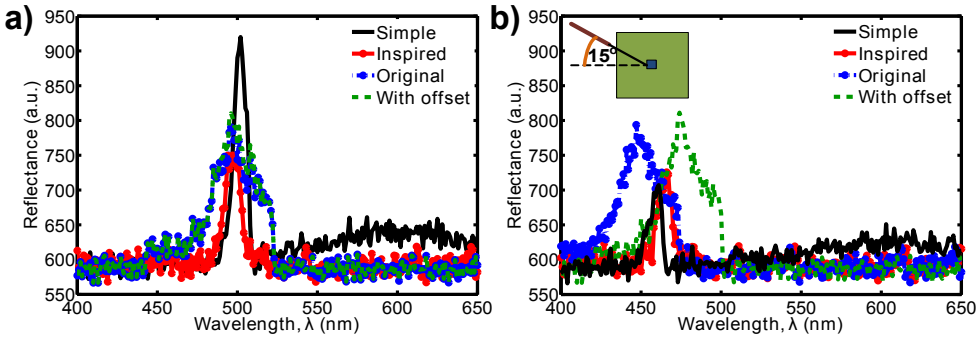

Fig. 5. (a) The reflectance spectra of the four fabricated samples for a normal light incidence. The 'simple' grating like structure (Fig. 3(b)) shows the highest and sharp intense blue reflection (solid line). The 'inspired' structure (Fig. 3(c)) reveals a narrow band spectrum as expected from the simulation (circles). The 'original' structure (Fig. 3(d)) has a broader spectrum with a bandwidth of 50 nm (quadrangles). The inclusion of an offset of 50 nm between neighboring ridges (see Fig. 3(e)) does not change the spectrum for normal illumination (crosses) and the corresponding curve partly covers the data of the 'original' structure without offset. (b) The reflection spectra change for a reflection angle of 15°. The reflection of the 'simple' grating reduces by 25% and moves out of the blue regime. The 'original' structure has a reduction of only 5% in intensity. The intensity reduction is even lower for the structure 'with offset'. Consequently, the alternating lamellae pattern and the alternating offsets between the ridges are responsible for the wide angular reflection of the *Morpho* butterfly in the blue regime .

measurement is done first without sample and subtracted afterwards from the measurements with sample to avoid unwanted disturbance from the background.

Quantitative reflection measurements are displayed in Fig. 5 and 6. The 'simple' grating like structure shows acute reflection at normal incidence as expected from the simulation. The "Christmas tree" like structure with alternating layers (both 'original' and 'original with offset') reflects a band of 470-520 nm of the incoming white light. This result deviates a little from the spectra of the original Morpho nanostructures that show highest reflection in the 400-500 nm region. This might be due to the slightly curved shape of the "trunks" of the "Christmas trees" observed in the original Morpho scales [16, 17]; a feature which is not considered in our structures. The fabricated 'original' structure, however, preserves the same blue color even for large angles of incidence (Fig. 6). The structure with 'original' pattern has only a 8% reduction in intensity (without offset between the ridges) as observed for an angle of 25°. Interestingly, the intensity of the 'original with offset' structure merely decreases and stays nearly the same compared to the reflection at normal incidence. For the 'simple' structure the reflection drops abruptly (around 30%) at 15° and loses its reflection at an angle of 20° (not shown). This is a well-known effect of thin film interference and our 'simple' structure behaves nearly like a stack of alternating layers of different refractive indices (air and PMMA). The spectrum of the 'inspired' structure with "Christmas tree" like shape moves less towards the UV region and undergoes less reduction in the blue reflection intensity at 15° compared to the 'simple' structure. Nonetheless, the spectrum is less broad as the 'original' ones with and without offset between the ridges. The improvement caused by this offset becomes most evident by the spectra shown in Fig. 6. 'with offset' the intensity reduces only slightly for angles of 15° and 25° and shift only 45 nm to the UV compared to 75 nm for the 'original' structure without offsets between the ridges.

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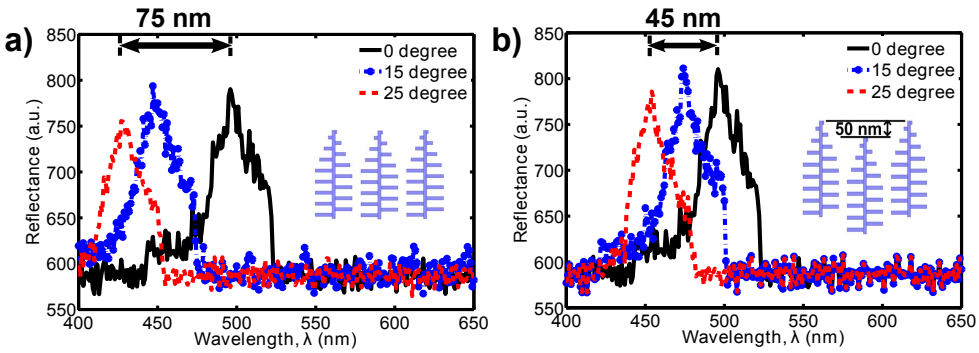

Fig. 6. Reflection spectra of fabricated original *Morpho*-type structures with and without offset between the ridges are compared in (a) and (b), respectively. With an offset of 50 nm between neighboring ridges, no reduction of intensity is observed for an angle of 15° and only 5% reduction of intensity with a shift of 45 nm towards UV region for an angle of 25°. The reduction of intensity is much higher for original structure without offset for an angle of 15° degree. This effect becomes even more evident at an angle of 25°.

5. Conclusion and outlook

To conclude, our work explains the wide angle reflection of the famous iredescence from blue *Morpho* butterfly scales. To analyze this famous optical property, we performed optical simulations on various nanostructures mimicking *Morpho* butterfly scales. FEM simulations were applied to calculate the scattering of incoming white light. To validate the numerical results, Morpho nanostrucutures are fabricated accurately in terms of structure and dimensions using high precision e- beam lithography. The wide angular blue reflection proves that it is possible to fabricate such tiny structures with state-of-the-art technology. Although, the structures are not free standing and oriented in the 2D plane they might be used in integrated optics for research purposes. The aspect ratio of fabricated structures can be improved by increasing the PMMA layer and improving the parameters of e-beam lithography. Furthermore, they might be replicated by hot embossing in combination with the LIGA technique [34]. Since *Morpho* butterfly structures show a lot of promises for technological applications and our horizontal design might help to accelerate scientific research on them.

Probing the numerical and experimental results, it is evident that the alternating layers are responsible for broad band and wide angle reflectance at the same time. Furthermore, the inclusion of an offset between the ridges increases the intensity of reflection for a wide range of angles. Only 5% reduction of intensity in reflection is noticed for the 'original structure with offset' at an angle of 25°. The "Christmas tree" structure removes the directionality of the blue iredescence. With simple interference structures, high intense reflection can be achieved but the original structural color of *Morpho* butterflies can only be achieved by creating alternating layers in "Christmas tree" shape with height offsets between neighboring ridges. Consequently, these design principles have to be used to produce surfaces with high iridescence in the future.

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