

Supplementary information: Structural colors with angle-insensitive optical properties generated by *Morpho*-inspired 2PP structures

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I. DISORDER CHARACTERISTICS IN BIOMIMETIC 2PP STRUCTURES

In the article, we presented the application of two-photon polymerization (2PP) for mimicking the lamellar ridges of the blue *Morpho* butterflies. We applied the interference-assisted polymerization method,^{1,2} since otherwise, the resolution of the conventional 2PP structuring strategy would be too low.³ In the interference-assisted polymerization method, a thin film of the photosensitive material is used for manufacturing arbitrary structures with modified cross-section (see manuscript). Therefore, polymer line structures with multilayer systems consisting of polymer and air, similar to those of the *Morpho* ridges, can be produced. If the fabrication process is performed close to the polymerization threshold, other effects can also be observed, which leads to specific disorder characteristics in the structures.

First, small but significant differences in the dimensions of single polymer layers and air cavities in the biomimetic multilayer systems were identified (see Fig.S1). The resulting inhomogeneity was mainly caused by the fluctuation of the average power of an ultrashort pulsed laser.³⁻⁶ In addition, individual line structures were partly tilted against each other by up to 10° due to the low degree of polymerization⁷ (see Fig.S1). Furthermore, the polymer lines showed slight differences in height, which can be explained by a low shrinkage of the polymer during the development process (see Fig.S1).

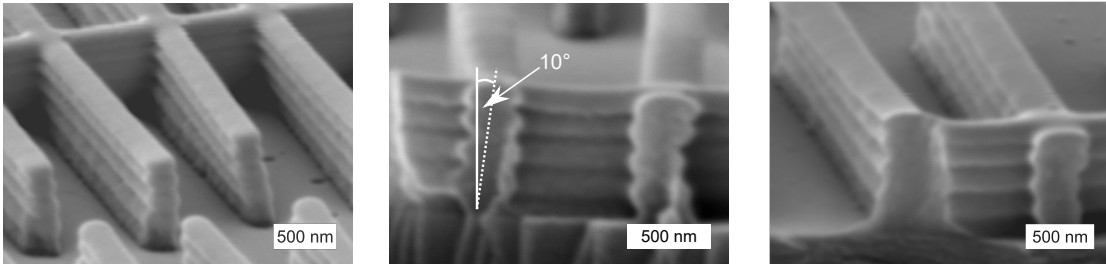


FIG. S1. **Disorder characteristics in individual polymer lines.** The three SEM images illustrate single polymer lines that form a multilayer system consisting of polymer and air. Such structures reflect the light in blue with almost angle-independent properties. The exceptional optical properties can be attributed to the disorder characteristics: (1) inhomogeneity in the structure of the multilayer system, (2) tilting of individual polymer lines, and (3) height differences between the polymer line structures.

Although these three effects influence individual structures randomly, this is advantageous for biomimetics, since disorder characteristics comparable to those found in nature can be mimicked. Therefore, the interference-assisted polymerization method led to the generation of 2PP structures that strongly resemble the *Morpho* butterflies' surface structures on their wing scales and thus, exhibit angle-insensitive optical properties.⁸⁻¹²

II. OPTICAL PROPERTIES OF BIOMIMETIC 2PP-STRUCTURES WITH MODIFIED GEOMETRIES

The following results are a summary from *Zyla et al.*¹³ and are supplemented by another structure geometry, which we call circle structure. In this case, we intended to demonstrate that the biomimetic structural color does not result from the overall structure, but from the multilayer system in the cross-section of single structural features (see Fig.S2). It should also be noted that even a varying distance between individual structural features only affects the color intensity but not the color hue.¹³ However, it is possible to adjust the direction from which the angle-insensitive optical properties appear. Therefore, we used specific structural geometries with different symmetry properties. If, for example, a colored surface, produced with grid structures, is tilted around the horizontal or vertical sample axis, the color is angle-insensitive for both directions. In contrast, the color surface of uniformly oriented arc structures exhibits the angle-insensitive optical properties only when the sample is tilted around its vertical axis, since it is only in this direction that a sufficiently high density of multilayer systems participate in the color formation process. In addition, these optical properties can also be adjusted omnidirectionally using the circle structure, since here, the same number of multilayer systems exist in each direction.

To substantiate the claims, the optical properties of the respective color surfaces were determined from different directions (I1, I2, I3) using angle-resolved spectroscopy. In this case, the structured areas were illuminated at an angle of 45° to their surface normal, as shown (see Fig.S3A-C). The detection of the reflected light was performed for the same angle value. The structured surface fabricated by circle structures, was set for three random orientations. The optical measurements in Fig.S3A-C underline our aforementioned statements.

Furthermore, angle-insensitive optical properties can be achieved independently on the

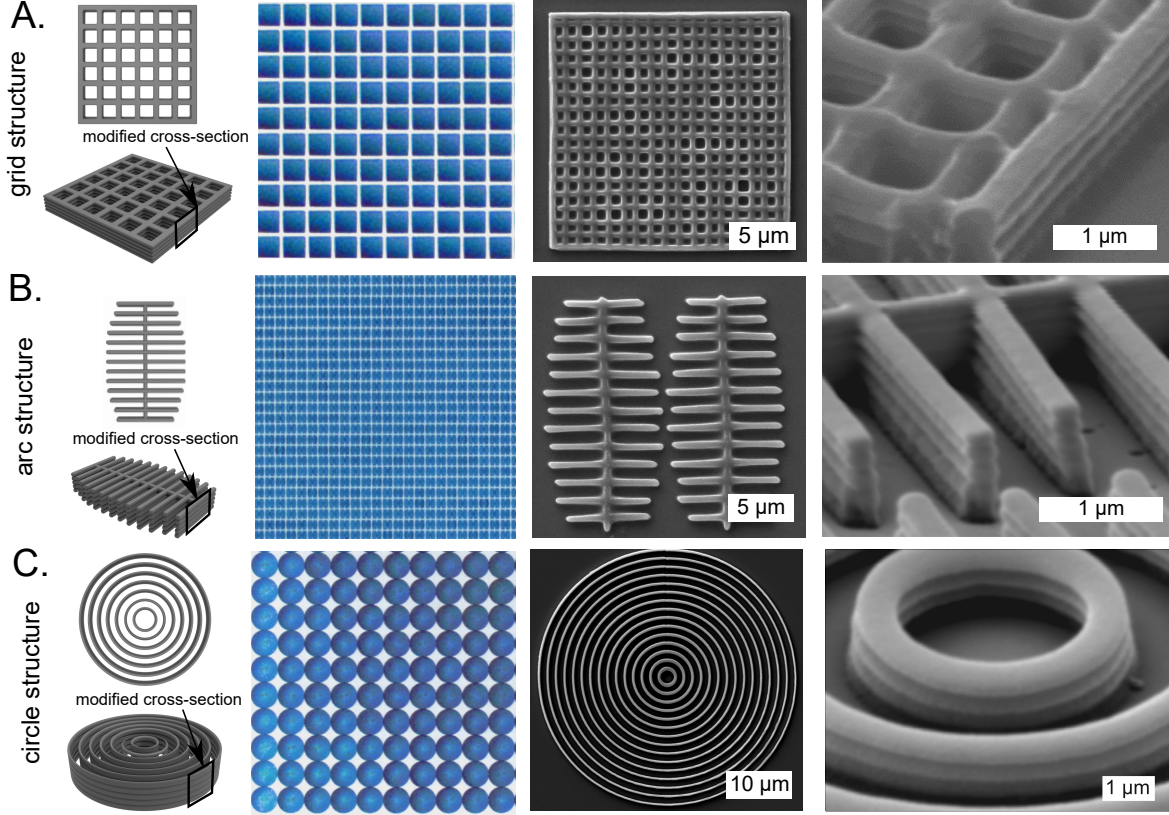


FIG. S2. **Generation of a blue structural color with different structure geometries.** Arbitrary structure geometries, e.g., (A) grid, (B) arc, or (C) circle structures, can be used to manufacture structural colors. All structures were produced with the same average laser power and the same velocity of the galvo scanner. Therefore, all multilayer systems are the same. The colored area in (A) measures $62.500 \mu\text{m}^2$ while the color areas in (B) and (C) are 0.25 mm^2 . Arc structures in (B) are uniformly orientated in the structured area.

structure geometry in the direction (I1) where the light is highly intense reflected. This opens up a wide range of possible applications, e.g., in the field of coating or anticounterfeiting.

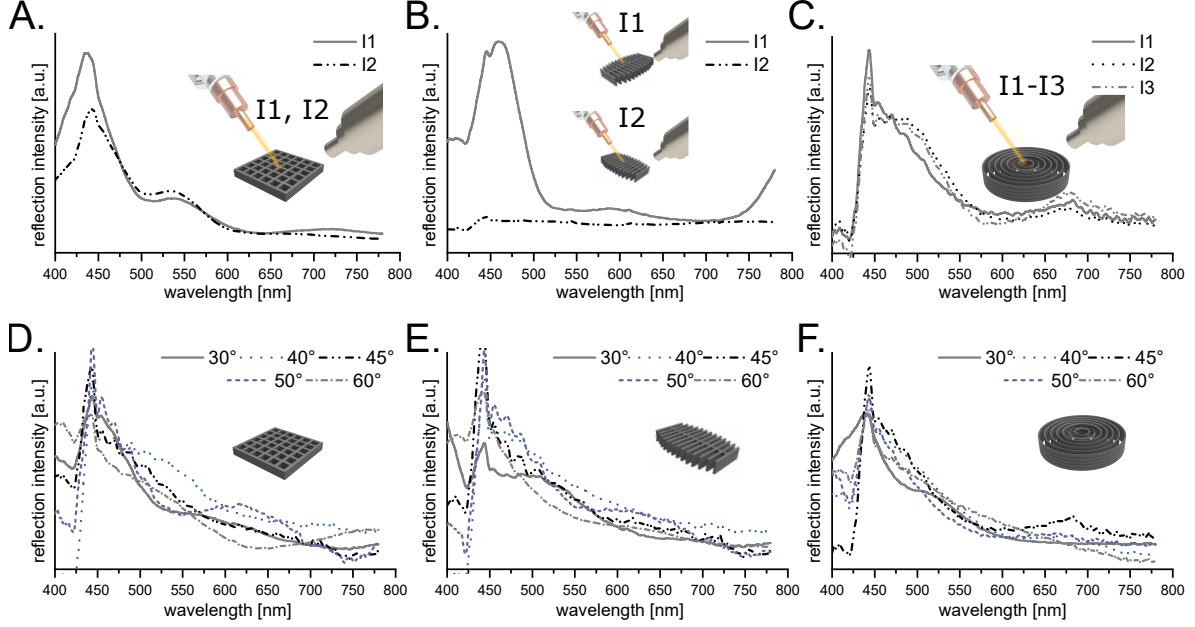


FIG. S3. Optical properties of color surfaces produced by different structured geometries. (A)-(C) Optical properties of colored areas using the grid, arc, and circle structure. Here, the angle of incidence for illumination and the angle of reflection for the detector position was 45° . Depending on the structure geometry and on the structure's orientation, the blue color occurs in a specific direction. In that direction (I1), in which the blue color occurs for all structure geometries, the angle-insensitive optical properties still exist (D)-(E), and they are almost identical for each structure geometry.

The optical measurements in Fig. S3A-C underline our aforementioned statements. Furthermore, almost angle-independent optical properties can be achieved independently on the structure geometry in the direction (I1) where the light is highly intensely reflected. This opens up a wide range of possible applications, e.g., in the field of coating or anticounterfeiting.



FIG. S4. **Reproducibility of different structural colors.** Manufacturing of different colors by successively increasing the average laser power ($\Delta P_a=2.5$ mW) for each structured area from left to right. The lowest average power ($P_a=10$ mW) was used to produce a blue color. The individual color areas measured 0.375 mm².

III. REPRODUCIBLE GENERATION OF DIFFERENT STRUCTURAL COLORS

In the article, we mentioned the reproducible manufacturing of different colors. Furthermore, we also state that even minimal variations in the biomimetic multilayer system in the range of approximately 20 nm can be adjusted with the interference assisted polymerization method^{1,2} by applying different average laser powers. To further support these statements, we produced different colors on one cover glass (see Fig.S4). The average laser power was successively increased by $\Delta P_a=2.5$ mW from left to right for each structured area, starting at $P_a=10$ mW. The scan of the galvo scanner was kept constantly at 1.4 mm/s. In addition, the result presented demonstrates that a wide range of colors in the visible spectrum can be easily produced by this method.

IV. ADDITIONAL MOLDING RESULT OF BIOMIMETIC 2PP STRUCTURES

The replica produced by molding, which is discussed in the article, indicates a shift in the color hue. We assume that this color shift may be caused by manually applied pressure with a cover glass during the preparation of the negative. Therefore, the multilayer system in the cross-section of the individual lines in the negative was modified in the nanometer range. Thus, the interference condition was varied. However, due to the slightly yellowish color of polymerized Spurr, the impression could arise that the green color may be caused by the optical properties of the material. For this reason, we repeated the molding on the structured area, which is illustrated in Fig.S2C, but the pressure we manually applied was

lower than before. The result is demonstrated in Fig. S5A where it is shown that the same color can be produced. Therefore, our assumption seems to be correct. However, the color intensity of the blue replica was lower than those from the original, which we can explain by the partially damaged structure features illustrated in the scanning electron microscope (SEM) image in Fig. S5B.

Nevertheless, the SEM image in Fig. S5C shows the multilayer system for undamaged replica features. Furthermore, the replica's optical properties, as determined by angle-resolved spectroscopy, also exhibited angle-insensitive optical properties (see Fig. S5D). Therefore, we can also state that the molding process is reproducible to structural colors generated similar to those of the blue *Morpho* butterflies.

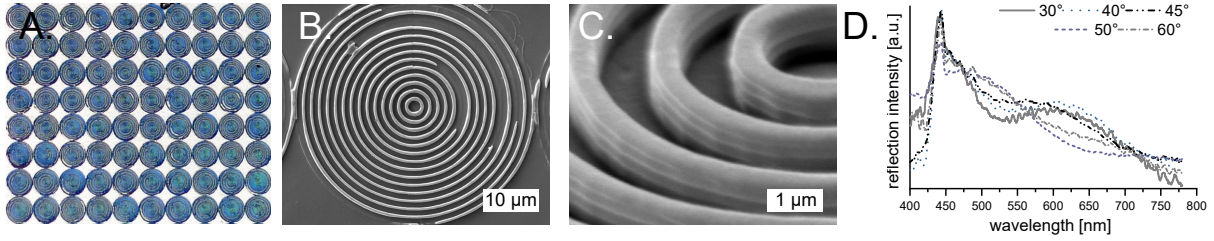


FIG. S5. **Results of biomimetic replica using molding.** The structured area from Fig. S2C was used for molding. The replica indicates the same color but with a lower color intensity, which is due to partially damaged structure features. Undamaged structure features exhibit a similar multilayer system as the original circle structures, and, therefore, the replica also has angle-insensitive optical properties.

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