Morpho - blue principle and: its lithographic reproduction

Akira Saito, Shinya Yoshioka, Shuichi Kinoshita

Since more than 100 years ago, the Morpho butterflies of South America have attracted interests because of their brilliant metallic blue wings. The blue luster is produced by a transparent protein in the cuticle of the butterfly's scales. The origin of the coloration is then not pigment but rather the interference effect of light scattered from microscopic structures. The structural color is not affected by longtime chemical change, and the color does not fade even after 100 years.

The interference also explains the high reflectivity of the blue coloration. The principle of this coloration was then simply considered due to a grating or multilayer structure for longtime. In fact, the analysis of origin of the blue coloration attracted the attention of researchers as well, and seems to be a history shot with big scientists (A.A.Michelson (1911) Phil.Mag.21, p.554; Lord Rayleigh (1918) Phil.Mag. 37, p.98). After the electron microscope caught clearly the microscopic structure of the scale, the traditional dogma on the origin of the metallic blue has been accounted to be proved.

However, the optical characteristics of the scattered light cannot be explained by a grating or multilayer, and the traditional dogma is faced with a serious mystery: the color is independent of the observation angle (i.e. it appears blue for wide angle more than ±40 degrees from the top). This nature of lack of rainbow-coloration contradicts the essential properties of interference, which was remained unnoticed for many years.

This mysterious feature has recently been explained by a peculiar optical structure that includes "discrete" multilayers [1]. This model can totally explain the specific characteristics of the Morpho blue: hot brilliant blue in broad angular range, high reflectance, speckle aspect, slight change of the color tone into violet phase at a grazing observation angle, and one-dimensional anisotropy of the brilliance.

As shown in figures 1 and 2, the essence of the blue can be summarized in five principles. In these figures, scanning electron microscopy (SEM) images of the wing scale from the typical Morpho butterfly (Morpho Didius) are shown on the left, while schematic model depicting their function are shown on the right. The red numbers in these two figures correspond to the following five principles:
1. A discrete multilayer composed of alternative layers of high and low refractive index is origin of the blue. The blue coloration is produced by interference in a single discrete multilayer.
2. Blue light is diffracted into a wide angular range because of a limited size of the scatterer. The width of each discrete piece must be in the order of a wavelength.
3. Random heights of the discrete pieces prevent a rainbow-coloration by precluding interference between the lights coming from neighboring discrete pieces. The randomness of the height must be on the order of a wavelength. The randomness also produces the speckle-like luster.

4. A narrow gap between the discrete pieces generates high reflectivity. Otherwise, the incident light penetrates the bottom of the multilayer and absorbed or transmitted through the substrate. The gap must be on the order of a wavelength.

5. The scale is made up of a quasi one-dimensional pattern as revealed by SEM (Fig.2, left). In other words, the pattern extends along Y-direction, but it is broken into a staggered array of discrete linear sections. This anisotropy plays an important role to make high reflectivity in a finite direction. Otherwise, two-dimensional (isotropic) randomness (Fig.2 (c)) would scatter the light in all solid angles, leading to a critical decrease of reflectance at any observing angle. In addition, a continuous one-dimensional pattern (Fig.2 (a)) would produce a strong diffraction in too narrow solid angle along the Y-direction. Therefore, the random brakes in the Y-direction scatter the blue light in adequate angular range and prevent a rainbow-interference, as well. The brakes along the Y-axis have a random interval distributed around $2 \mu m$. This quasi one-dimensional pattern can be approximated in Fig 2 (b) as rectangular pieces distributed randomly with an interval around $2 \mu m$ in Y-direction.

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However, a theoretical hypothesis needs an experimental proof and we attempted to fabricate an optical film by changing and controlling the five conditions on an order of 100 nm. Nevertheless, the fabrication process needs an artifice. The necessary structure is ideally as shown in Fig.3: multilayer, limited width and gap, random height, and
quasi one-dimensional structure. One of the ways to produce this composition is shown in Fig.4 [2]. As a first step, a randomly polished surface is prepared on a glass or plastic substrate. A multilayer composed of alternating layers of high and low refractive index is deposited on the surface to produce the blue coloration. This is followed by a gravure process to obtain the adequate gap and width of the discrete pieces. Materials of the multilayer is desired to be oxides for high controllability of thickness and selectivity of the refractive index.

Fig.3 Schematic view of the structure necessary to reproduce the Morpho blue.

Unfortunately, this process is hardly accomplished because of difficulty to realize a high aspect ratio on the oxide film of 1-2 μm thick in a practical area (~mm size). We therefore tried another process by extracting the minimum essence of the principles. The most important step is to first engrave the substrate surface, considering the above mentioned 4 principles except multilayer. In other word, we graved the surface by controlling the gap and width, which incorporate simultaneously randomness and quasi one-dimensional anisotropy as shown in Fig.5 [3]. The surface pattern is composed of randomly distributed rectangular units of 300 nm x (2000 ± a) nm (Fig 2(b)), where a is a finite standard deviation. To achieve this delicate first step, we used conventional lithography employed by the semiconductor industry for quartz substrates.

Fig.5 Fabrication process for obtaining the minimum features necessary to reproduce the Morpho blue. SEM image of the substrate surface prior to deposition [3].

The process was finished by deposition of TiO₂ (high refractive index layer, ~80nm thick) and SiO₂ (low refractive index layer, ~150nm thick) without a final step of engraving. This minimized the cost and process under the limited condition of fabrication. This optical film thus generated successfully reproduced the Morpho blue (Fig.6) with its fundamental characteristics: hot blue in a wide angular range, high
reflectivity, slight change of the color tone at a grazing angle, speckle feature, and one-dimensional anisotropy of the brilliancy.

Fig.6 Photograph of *Morpho Didius* (left) and the reproduced film (right).

Fig.7 shows the measured angular dependence of reflectance from the normal incident light for different samples: (a) a real *Morpho*-butterfly, (b) a continuous multilayer deposited on a flat substrate, (c) a patterned bare (deposition-free) substrate, and (d) an artificial *Morpho*-like film. The smooth angular dependence in the blue range (Fig.7 (a)), characteristic of the *Morpho*-butterfly's wing, was reproduced (Fig.7 (d)), whereas this property was lost for the others (Fig.7 (b), (c)). The results represent the role of the multilayer and the substrate. A still sharp profile of Fig.7 (d) is attributed to the lack of randomness due to the use of a simplified design with a single graving depth, which produces a specular reflection especially at longer wavelength.

![Graphs showing the measured angular dependence of reflectance from the normal incident light for different samples: (a) a real *Morpho*-butterfly, (b) a continuous flat multilayer, (c) a patterned bare (deposition-free) substrate, and (d) an artificial *Morpho*-like film.](image)

By comparing twelve substrates fabricated with different parameters, as we predicted, we found that the optical properties were influenced by the structure on a 100 nm scale (data not shown): a film on a substrate fabricated with larger gap and width (900nm) generated the blue coloration in too narrow angle, similar to the result shown in Fig.7.
(b). A continuous one-dimensional pattern (Fig.2 (a)) produced the blue in too narrow angular range and also glittering multi-coloration because of strong interference due to a lack of randomness. On the other hand, the two-dimensional pattern (Fig.2 (c)) scattered the light in all solid angles, causing a critical decrease in reflectance. Importantly, this model is based on both ordered (multilayer) and disordered (random) structures, but it is not a simple medium between them but a delicate combination of both. This "controlled irregularity" demonstrates the surprising skill and art of nature.

In the past years, there were many trials for reproducing the attracting *Morpho* blue. However, certain critical conditions were fatally overlooked, especially the randomness, narrow width and gap, and quasi one-dimensional structure. Therefore, the past products have produced a mixture of different colors, or a decrease in reflectivity. A practical area size of the film is also an important point as a product.

Such artificial structural color can be used in a wide variety of applications, including decoration, textile, paints, and security (like a hologram). It produces color without pigment, it makes tone that is qualitatively impossible by pigment, and also resistant to discoloration due to chemical change over time. Furthermore, the artificial film is resistant to fire, heat, sunshine, and it should be possible control the specific coloration; for example, it should be possible to produce a red *Morpho* film).