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PAPER

Unparalleled sensitivity of photonic structures in butterfly wings

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Butterflies are famous for their brilliant iridescent colours, which arise from the unparalleled photonic nanostructures of scales on their wings. In this paper, the sensitivity characteristics of photonic structures in butterfly wings to surrounding medium were found. Firstly, it was showed that the iridescent scales of *Morpho Menelaus* butterfly give a different optical response to surrounding vapours, such as water, ether and ethanol. Then, the ultra-depth three-dimensional microscope and FESEM were used to observe the morphology and nanostructures of butterfly wing scales. The highly spectral response characteristics were identified by using Ocean Optics spectrometer USB4000. It was found that the reflectance spectra of *Morpho Menelaus* butterfly scales could provide information about the nature of the surrounding vapours. Afterwards, the theory of multilayer-thin-film interference was used to analyse the mechanism of this sensitivity. It was the multilayer-thin-film interference structure constituted by alternating films with high and low refractive indexes leading to the sensitivity of butterfly wings. The refractive indexes of surrounding mediums play an important role in gas sensitivity. These characteristics dramatically outperform that of existing nano-engineered photonic sensors and maybe potential to design efficient and high sensitivity optical gas sensors.

Introduction

Over the past few years, there has been increasing interests and efforts working in the photonic crystals in the visible wavelength range research of *Morpho* butterflies,[1-4] spectroscopes, or display screens.[5-6] Although photonic structures of biological origin have been well studied for their optical and morphological properties,[7] the methods for the potential applications in selective gas sensors,[8] functional coatings,[9-10] structural color devices[11-12] that help make large, cheap and operable photonic crystals working in the visible spectrums are highly required.[13]

Butterfly is one of the insects with brilliant iridescences. The multi-functional characteristics of *Morpho* butterfly wing scales, including lightweight, visual effects, hydrophobic, mechanically strong and thermal regulations, are revealed and have a relationship with the photonic nanostructures in the ridges of the scales.[14] The optical properties due to the multi-functional characteristics of the butterfly wing scales are associated with photonic crystal, and such optical properties depend strongly on wavelength and incidence angle of the incident light and the viewing angle.[15] The colors of its wings could change with the viewing angle, which is referred to as structural color.[16-17] It was found that some butterflies are famous for their structural color[18] and multifunctions[19-21]. *Morpho aega* and *Eryphanis reevesi* exhibit a strong iridescent, which are attributed to the discrete "Christmas-tree" structures in butterfly wings.[22] Photonic crystal structures of *Morpho* butterfly wings can lead to diversity of colors and patterns.[23] The photonic structures of butterfly wings are well called photonic band gap materials (PBG).[24] The revealed "Christmas-tree" nanostructures in the ground and cover scales of *Morpho Menelaus* butterfly are responsible for the observed iridescent blue color and the diffraction pattern of the wings.[25]

Although the materials replicated from photonic structures in butterfly wings may have numerous applications ranging from optical computing[26] through tunable photonic circuits,[27] more efficient lasers[28] and fibers for optical communications,[29] less harmful pigments[30] to photonic paper,[31] its gas sensitivity was mentioned only a few. These nanostructures of butterfly wings have been found to be sensitive to surrounding medium.[7] In fact, the nanostructures that caused the brilliant iridescent colors is formed by discrete multi-layers of cuticle and air, which is also the basic reason of butterfly's sensitivity to environmental surrounding mediums.[32-34] The brightness and color can be changed with the alteration of surrounding mediums.[35] This special property could be implemented into the engineering designs for highly sensitive detection of optical gas sensors.[36]

In this work, the optical sensitivity of *Morpho Menelaus* butterfly wings to surrounding vapours, such as water, ether and ethanol, were investigated, which is mainly based on several elementary optical processes including multilayer interference,

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diffraction grating,[37] light scattering and so on.[38] It was found that the sensitivity of *Morpho Menelaus* butterfly wings to gaseous ether and ethanol is more complicated than gaseous water, although the optical sensitivity of *Morpho Menelaus* butterfly wings to surrounding vapours of water, ether and ethanol were very significant. What's more, not only the different sensitivity were identified, but also a color change accompanied with the different sensitivity were also identified. The theory of multilayer-thin-film interference was used to analyse the sensitive mechanism of *Morpho Menelaus* butterfly wings to ether and ethanol. This unparalleled sensitivity of photonic structures in Morpho Menelaus butterfly wings could be applied to highly sensitive and selective bio-inspired sensors by designing and fabricating artificial nanostructures.

Experimental

In this work, the scales of *Morpho Menelaus* butterfly were taken as biological experimental sample. Due to the back wing's surface of *Morpho Menelaus* butterfly is covered by mysteriously brilliant blue scales, only one part of *Morpho Menelaus* butterfly wing was chosen as experimental areas to be studied carefully.

Analytic grade reagents NaCl, ether and absolute alcohol were provided for experimental pre-treatment. The macroscopic morphology of the butterfly wings was characterized using a camera. The spatial arrangements of the scales were characterized using the ultra-depth three-dimensional microscope (VHX-2000). With the help of scanning electron microscope (FESEM: JSM 6700-F), dimensions and characteristics of the cross-sectional ultra-structure were obtained. Spectrometer USB4000 was provided by the Ocean Optics for determining reflectance spectra of the scales during the process of interaction with vapours of water, ether and ethanol.

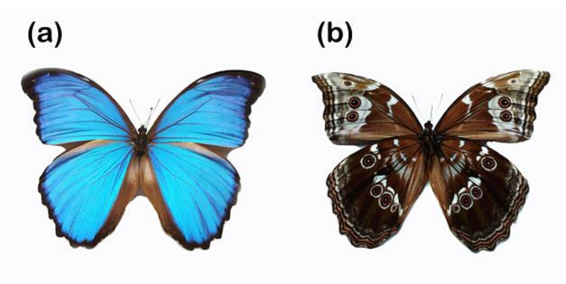

Fig. 1 Macroscopic appearance of the original *Morpho Menelaus* butterfly wings: (a) The dorsal wing exhibits blue coloration and (b) The ventral surface is brown in appearance.

In this experiment, specimens of butterfly wings were rinsed three times by 0.65% NaCl solution, and then soaked into ether for 10 min to get rid of the grume, fattiness, and proteins on the wing surface.[39] Afterwards, a series of dehydration pretreatments in graded ethanol solution was conducted. At last, in order to demonstrate that the *Morpho Menelaus* butterfly wings have highly sensitivity response to vapours of water, ether and ethanol, the specimens were placed into a sealed box and in a darkness room. Aim of this operation is to exclude the interference of other light. The water was heated by alcohol lamp. The ether and ethanol was heated by hot water bath to make water, ether and ethanol evaporate fully. Then, the vapours of water, ether and ethanol were filled into the equipment gradually, and the reflectance spectra was recorded by Spectrometer USB4000 during the whole experiment process.

Results and discussion

The beautiful colors of butterfly wings are usually contributed by two sources: chitin periodical structures and pigments, which are also referred as "physical" colors and "chemical" colors, respectively.[24] Notably, the spatial structure arrangement of scales plays the key role for structural (physical) color.[17, 40] For the purpose to verify the high selectivity of butterfly wing's surface structure to surounding mediums, the "physical" color of its wings is the prerequisite condition for choosing the right type of butterfly to be studied. *Morpho Menelaus* butterfly has bright blue wing scale, and it is well known by its iridescent blue color as shown in Fig. 1. The wingspan is about 12.0 cm. Through a simple alcohol discoloration experiment, it is proved that its iridescent color is structre-based and has angle-dependent discoloration effect. It can be distinguished by variation observation angles using the naked eye. The *Morpho Menelaus* butterfly lives in tropical forest, in particular the central and south America, including Brazil, koda, and venezuela. It is the kingdom of butterfly in Brazil. The fore and hind wings all exhibit brilliant blue with edged black color. However, only the dorsal surface exhibits the blue coloration shown as Fig. 1a. The ventral surface is brown in appearance shown as Fig. 1b.

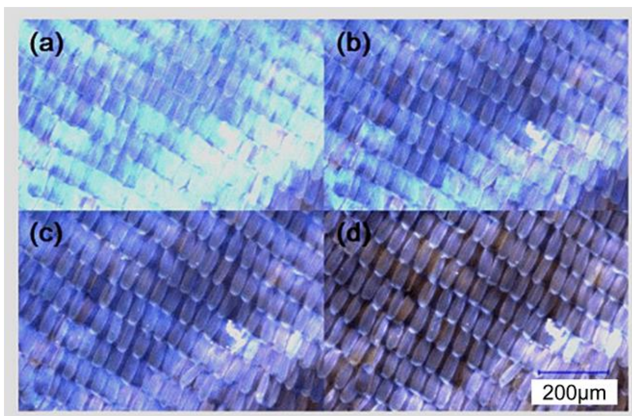

Fig. 2 The color change of *Morpho Menelaus* butterfly scales under different light intensities. This pocess were characterized using a ultra-depth three-dimensional microscope. It was magnified 200 times. With the order of light intensity: (a) > (b) > (c) > (d), the scales changed from blue to colorless. It is also indicated that the blue color of the ground scales is structure-based.

The revealed "Christmas-tree" nanostructures in the scales of *Morpho Menelaus* butterfly wing are responsible for the observed iridescent blue color.[41] In fact, this kind of "Christmas-tree" nanostructure is an alternative multilayer structure of chitin and air. The cuticle of chitin is transparent with little pigment. The reflection and absorption of the chitin/air multilayer system can provide not only the most pure structural colors but also the stability of color against variations in multilayer structure.[42] The iridescent blue color of *Morpho Menelaus* butterfly dorsal wing was shown in Fig. 2. It has two layers of scales: cover scales without color and ground scales with structural color. Along with the light intensity weakening, the blue color of ground scales was changed from blue to colorless. However, there is no color change of cover scales. It is also indicated that the color of the ground scales is structural color. The reason is that the color would not be changed if it is pigment (chemical) color. So, the functions of its wings must have relationship with it surface microstructures. Then, the surface ultrastructures of butterfly scales will be meticulous researched. The cover scales lie above the ground scales and exhibit high levels of transparency as shown in Fig. 3a. Generally, the iridescent blue color of *Morpho Menelaus* butterfly wings is mainly caused by strong constructive interference within the nanostructures of each delicately sculpted scale. So, the high-magnification cross-section images of the "Christmas-tree" nanostructure of *Morpho Menelaus* butterfly's ground scales are the fundamental reason of the iridescent blue as show in Fig. 3. The tower structures are composed by a thin-film multilayer nanostructure as shown in Fig. 3b. It can be observed that the *Morpho Menelaus* butterfly scales have tower structures, which are composed of alternating layers of chitin and air. This multilayer system has some multilayered exhibition arms of chintin is interspersed with air gaps layers of approximately 80 and 110 nm in thickness, respectively. The multilayer structure has a size of 1.8 μm height and 0.5 μm width. The distance between adjacent ridges is approximately 0.8 μm. It is obvious that each ridge has 8-9 pairs of exhibition arms (chitin layers) and the lamellae distributes on both sides of the ridge axis symmetrically in width. Their branches of the tree-like structure run parallel or near-parallel to the base of the scale. These dimensions are very important because such a structure will cause the bright blue color and interact with vapors.

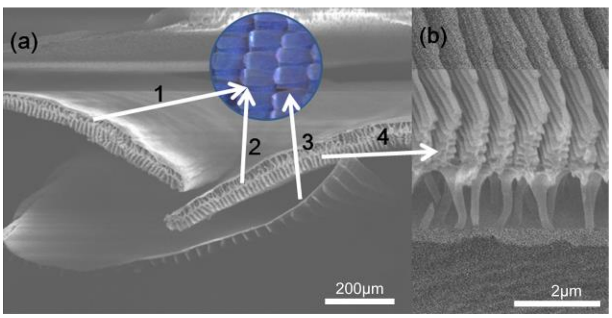

Fig. 3 (a) High-magnification FESEM images of the cross-section microstructures of *Morpho Menelaus* butterfly wing scales. Its configured multilayer structures are revealed clearly. Arrow 1 and arrow 2 are ground scales with blue color and arrow 3 is cover scale without color. (b) The multilayer structure of the ground scales is arrow 4. A shelf-like multilayer structure and its dimensions are obtained from observing the FESEM images.

The reflective spectrum was measured to detect the sensitivity of *Morpho Menelaus* to vapours of water, ether and ethanol. The incident light was emitted from an LS-1 tungsten halogen light source and transported by an optical fiber. Then, the light was irradiated on the scale samples. The reflected light was captured and inputted into an Ocean Optics USB4000 spectrometer. The experimental device and its working principle is provided in ESI†.

The reflective spectrums of the *Morpho Menelaus* butterfly scales to vapours of water, ether and ethanol were acquired respectively shown in Fig. 4. The real concentrations of the vapours have been provided in ESI†. Although the reflection intensity of *Morpho Menelaus* butterfly scales changed during vapours of water, ether and ethanol concentration increasing, the reflection intensity is not same. It is indicated that the structure of butterfly scales has highly spectrum selective response to different vapor.[7,43] Vapours of water molecular enhanced the reflection. When the concentration of pneumatolytic water is added up to a certain level, the fog droplet will formed. Then, the structure will be

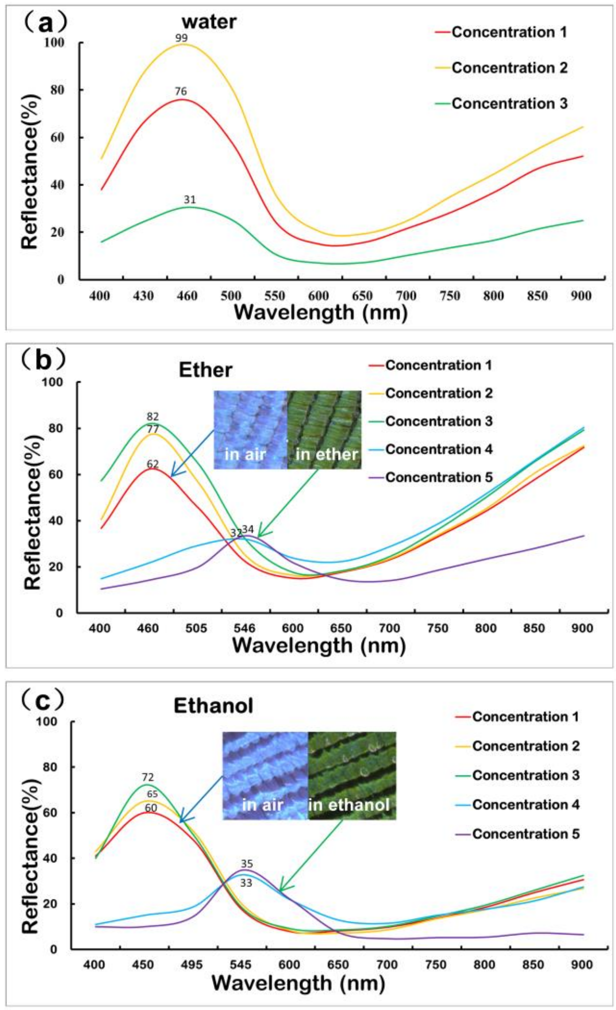

Fig. 4 The reflection spectrum of *Morpho Menelaus* butterfly wings under different vapours of water, ether and ethanol. Along with the increase of vapours concentration of pneumatolytic water, ether and ethanol, the butterfly scales show different spectral response. In reflection spectrum image of pneumatolytic (a) Water, it can be found that along with the increase of concentration vapours of water the reflection spectrum increased firstly and then decreased. However, the peak have no shift. In particular, along with the increase of vapor concentration of (b) Ether and (c) Ethanol, the reflection spectrum increased firstly. Then, the peak appears red shift. At last, when the pneumatolytic ether and ethanol increase and form liquids, the peak wavelength of reflection efficiency was at 548.45 nm and 547.19 nm and the color of scales is green as the arrow point.

Table. 1

Butterfly	Surrounding medium	Incident angle	r_1	r_2	h_1	h_2	n_1	n_2	λ_T	λ_E
Morpho Menelaus	air	0	0	0	80	110	1.555	1	468.80	463.77
	ether	0	0	0	80	110	1.555	1.349	546.90	548.45
	ethanol	0	0	0	80	110	1.555	1.362	548.44	547.19

Data used in Eq. (1). The experimental reflective spectrums of *Morpho Menelaus* in air, ether and ethanol are obtained, respectively, as shown in Fig. 4. The wavelength of diffraction efficiency peak was named λ_E. $\lambda_T=2(n_1h_1\ cosr_1+n_2h_2cosr_2)$, λ_E is diffraction efficiency peak of experimental data.

displayed as a hydrophobic behavior[44] and fog droplet could reduce the absorption of the probe light. So, the reflectivity is gradually reduced shown as the green curve in Fig. 4a. Vapor of ether and ethanol can not only make reflection intensity change higher but also color variation of *Morpho Menelaus* scales shown as Fig. 4b and c. From reflective spectrums of the pneumatolytic ether and ethanol, it can be found that the pneumatolytic ether and ethanol can make reflection intensity change higher. When the vapor concentration increases up to liquid, the reflective peak will be happened to red shift (blue and purple curves in Fig. 4b and c) and the color of *Morpho Menelaus* scales change from blue to green (insert illustration in Fig.4b and c). It is indicated that the structure demonstrates highly selective to pneumatolytic ether and ethanol before the two gases condense into liquid.

Models

The 2D optical models simplified from *Morpho Menelaus* scales were designed to analyse the sensitivity mechanism of the nanostructure shown in Fig. 5a. Here, the models with three periods was used. This period are set to infinite periods along the x-axis in this example.[45,46] The lamellae of the model are symmetrically distributed on both sides of the trunk and parallel to the base. The theory of thin-film multilayer interference of optical models was used to analyse the sensitivity mechanism of *Morpho Menelaus* butterfly scales to ether and ethanol. Considering the light is incidented on the scale where interference occured, the reflected light beams on the interfaces of the chitin and air may interfere with each other. If the constructive interference occurs, the light intensity will increase under conditions that the wavelength of the interference light is multiple of the half wavelength. If contrary, it will reduce. It was viewed that the thin-film multilayer interference occurred in the periodic multilayer structure. The chitin and air layers were designated as x and y with thicknesses h_1 and h_2 and the refractive indices were set to n_1 and n_2, respectively, as shown in Fig. 5b. it is assumed $n_1 > n_2$, for the present.[36] Here, the x layer is the material of chitin and the y layer is the surrounding medium. Thus, if the refractive index of the y layers was changed by surrounding mediums, the wavelength of reflective light will also change. It is the mechanism of the unparalleled sensitivity of photonic structures in *Morpho Menelaus* butterfly wings to surrounding mediums. Considering the multilayer horizontal model layers, the phases of the reflected light are both at the upper and lower interfaces between chitin and air. The thin-film multilayer interference can be applied as:

$$\lambda=2(n_1h_1cosr_1+n_2h_2cosr_2) \qquad (1)^{36}$$

There, the angles of refraction in layers of chitin and air are r_1 and r_2, respectively. The wavelength of reflective light changes with the thickness "h", refractive indexes "n", and the reflection angle "r". Next, considering the visible spectrum effect, the lamellae height h_1 and height between lamellae of the nanostructure h_2 of *Morpho Menelaus* butterfly are measured. The contrast of calculated values and experimental results are shown in Table 1. It is obvious that the peak wavelength of the reflectance and transmittance spectra undergos major changes when only the parameter of the refractive indexes of surrounding mediums chages and all other parameters remain unchanged. So, this result proved the conclusion again that it is the change of refractive indexes of surrounding mediums making butterfly wings have excellent sensitivity properties to surround mediums.

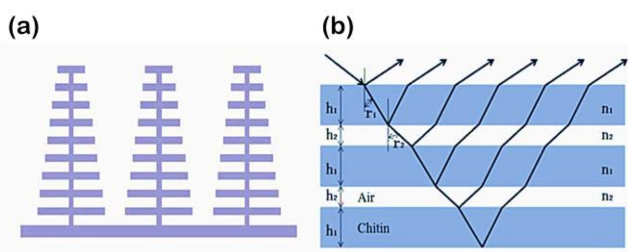

Fig. 5 (a) The horizontal model simplified from *Morpho Menelaus* butterfly and (b) Configuration of multilayer-thin-film interference.

Conclusions

The sensitive photonic crystal type nanostructures in *Morpho Menelaus* butterfly wings was investigated by electron microscopy and reflectance spectroscopy in this paper. This functional structures were composed of alternating layers of chitin and air. It was found that the blue color of ground scales is structure-based. And, it was also found that its iridescent color has angle-dependent discoloration effect. The unparalleled sensitivity of photonic structures in *Morpho Menelaus* butterfly wings to different surrounding vapours was found. From the reflectance spectra it can be found that the wavelength and efficiency of the reflectance peak could change with the alternation of concentration and type of surrounding mediums. In fact, it was the surrounding medium causing the change of refractive index of the air layer in the thin-film multilayer nanostructure. So, the structural color of the scales would also change accordingly. In addition, The refractive indexes of surrounding mediums in the thin-film multilayer nanostructure can affect the interference phenomenon. So, The refractive indexes of surrounding mediums play an important role in gas sensitivity. More importantly, the calculation result of the theory of the thin-film multilayer nanostructure was consistent with the experimental result, which provide a strong evidence for the analization of the sensitivity mechanism of butterfly scales to surrounding mediums. The sensitivity of the nanostructures of *Morpho Menelaus* butterfly scales is meaningful. It bodes well for the discovery of new gas sensitive structural materials. And its structural parameters maybe provide the basis for various artificial manufacturing practical applications. The works in this paper could steer the design of new optical gas sensors.

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