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Golden periodic nanostructures in
fauna and flora - Relations between
structure and function

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Abstract

Some animals and plants display iridescent color, generated through periodic nanostructures. The color derives from interference of light, which grant a bright, shiny appearance. In this study several samples were investigated with regard to their golden coloration. An overview of the different types of structural colors is given to form the boundaries, that will help to asses whether structural effects generate the samples´ colors. Several methods of analysis are presented, which were used for examination. Another concern of this study was to examine and simulate fluid run off properties of moths, because many butterflies and moths´ wings show hydrophobic properties and the ability to direct water flow. Simulation was performed with carved glass structures that tried to imitate the surface structure of butterfly scales.

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1 Introduction

The objective of this study is to investigate whether the golden color of the chosen samples is based on structures or pigments and to explore structure related properties from a biomimetic perspective. Biomimetics is the imitation of biological systems with the purpose to adapt these biological solutions for solving technological problems in physics or engineering. Due to evolutionary pressure, nature has pressed living organisms to develop highly optimized systems, granting them advantages over their competition. Valuable and desirable properties found in nature are, for instance, superhydrophobicity, self-healing abilities, low-friction coatings and iridescent colors. Iridescent objects gradually change their color as the angle of observation changes. Their colors result from microscopic structures, not from pigments. Therefore, one could create colors on surfaces without using paints or varnishes, which often contain toxic elements and have a negative impact on the environment.

Bionics and biomimetics have caught my interest many years ago and to this day remained fascinating and inspiring. The combination of technological problems and nature's solutions is a scientific approach that absolutely corresponds to my interests.

The purpose of this study is to further the understanding of the iridescent effects that generate structural colors, with the main objective being golden structural colors of two moths and golden colored bark. A lot of the systems in nature show a combination of properties, often achieved through one single structure, such as iridescent colors, thermal regulation and hydrophobicity. Therefore, it was decided to examine the hydrophobic aspects of the sample moths as well.

This study was conducted under the supervision of Ilse C. Gebeshuber at the Vienna University of Technology. Furthermore, the investigation was supported by Karin Whitmore, Thomas Schachinger and Nick Sinner.

2 Materials and methods

2.1 Studied samples

2.1.1 General description of *Lepidoptera* wings

In this study, the main objects of interest are *Lepidoptera*, which include butterflies and moths. This order of insects includes around 160.000 species. They can be found on every continent except Antarctica. Here, a short description of the general structure elements of their wings will be given. The terminology is taken from Serge Berthier's book on iridescent colors (Serge Berthier, *Iridescence-The Physical Colors of Insects*, 2007).

Lepidoptera have four wings in total, two on each side, the frontal and the hind wing. The wings develop when the *Lepidopteran* is still in its pupal stage, which is also called chrysalis. The wings are covered with several veins, normally visible to the naked eye. In the pupal stage, two separate membranes develop and merge together to form the wing membrane, which is the basic structure or foundation of each wing.

On these membranes, the scales are positioned orderly in areas, commonly designated through the veins. The shape of scales differs a lot, among species and even among a single specimen. They vary in length and width, have smooth or jagged ends or can be flat, bent or curled. They are oriented towards the edge of the wings. Very similar to bird feathers, they are attached to the membrane with small stems. A lot of iridescent *Lepidoptera* have two different kinds of scales, ground and cover scales, where one is pigmented and the other one has nanostructures that generate interferential light effects. Almost all scales show a very periodic structure of longitudinal striae, also called ridges. These striae are composed of many lamellae that all partially overlap in the same direction. The striae are supported by counter-striae, which are periodically arranged as well.

2.1.2 Mongolian moth

This moth was found in the steppes of southern Mongolia, in Ömnö-Gobi-Aimag. It shows various shades of brown, yellow, silver and gold all over its wings, abdomen, legs and even antennae. No difference in color was discerned between dorsal and ventral side of the wings. Depending on the angle of incidence, especially the silvery parts stand out, when observed under an optical microscope. Also a lot of the scales seem to be transparent at most angles and only show golden coloration under specific angles of incidence. However, most of the scales seem to contain some kind of brown pigmentation, especially when turning to the exterior parts of the wings.

When observed with the optical microscope, with a high magnification, iridescent effects can be seen on single scales, showing light colors ranging all over blue, green, yellow and red. The scales themselves are nearly flat, showing only slight undulations. Two different shapes of scales were distinguished. One was long and narrow, with three to five clearly visible prongs on the end, while the other was short and broad, with only three small prongs, that were more wavy. Both of these scale types showed the same



Figure 1: Mongolian moth; overview(left) and close up of a wing area, displaying iridescent effects (right)

coloration under optical observation, brightly flashing when illuminated under a right angle to the striae structure.

2.1.3 Common clothes moth - *Tineola bisselliella*

The clothes moths that were gathered and investigated all have a golden coloration, which ranges from completely golden all over to a brown golden mixture. The color is not dependent on the observation angle or light incidence, only the intensity increases at certain angles. Effectively all parts are golden, except for the eyes. Here as well, the same two types of scales were discerned. It has to be mentioned, that this is no classification of sorts, but only a visual distinction. Just like with the Mongolian moth, no difference in optical properties can be discerned under observation with the optical microscope. Also, the same iridescent colors can be observed on single scales, regardless their shape and size. In terms of shape, the same two kinds of scales are found, long and narrow, and short and broad, but both being of smaller size compared to their counterparts of the Mongolian moth.



Figure 2: Common clothes moth; overview(left) and close up of the tail end (right)

2.1.4 Mongolian bark

The third sample for investigation is bark from a bush in the South of the Changai mountains in Central Mongolia. It has a very smooth, shiny surface. On the inner side of the sample, fibrous liber¹ can be seen, that has a very similar color, compared to the outer bark. The bark itself is surprisingly resistant to physical stress. When the liber is scraped off the durability remains. It was chosen as a sample because of its radiant appearance, which is rarely seen in bark.

Viewed with the naked eye or through an optical microscope, no iridescent color effects are observed.



Figure 3: Golden bark from Mongolia, outer side on the left, inner side on the right picture

2.2 Methods of investigation

The main methods used for investigation were scanning electron microscopy (SEM) and transmission electron microscopy (TEM). These two microscope techniques enable high resolution imaging with a magnification of over 100 000. This allows to study the nanostructures of interest on butterfly wing scales, which would be impossible with simple optical microscopy, due to the dimensions of the structures.

The preparation of the samples was performed in a similar way for either the SEM or TEM studies. A micromanipulator was made out of an eyelash, that was glued to a stick and cut off at the end to increase its mechanical stability. This allowed careful treatment of the moths, in order not to destroy any scale structures and the moth wings themselves. Due to the electrostatic attraction between the scales and the eyelash, transferring them onto the sample holder was possible. Only the positioning of the scales posed a problem, because it was near to impossible to accurately place them intentionally on their top or bottom side.

¹plant tissue between wood and bark

2.2.1 Scanning electron microscope (SEM)

The scanning electron microscope uses a focused beam of accelerated electrons to produce images of solid samples by scanning the surface in a raster scan pattern. Signals are generated through the interaction between electrons and atoms of the sample. These signals can give information about the surface texture, crystalline structure and chemical composition. Achievable magnification lies in a range of about 6 orders of magnitudes, depending on the diameter of the electron beam. To avoid interaction of the beam with polluting atoms and molecules in the probe chamber, the process is performed in high vacuum. The resolution usually is below 1 nm. For better resolution and to prevent electrical surface charging, the samples were sputter coated with an alloy of gold and palladium, increasing the conductivity.

The electron beam is produced with a hot cathode or a field emission cathode, which emit high energy electrons. These are accelerated through an electric field, gaining energies, usually ranging from 0.2 keV to 40 keV. With the assistance of magnetic condenser lenses, the electrons get focused to a beam, which has a diameter of about 0.4 nm to 5 nm. The beam direction is controlled with deflection coils, screening the surface of the sample in a raster scan pattern.

Within the so called interaction volume, the primary electrons interact with sample atoms, losing energy through absorption and scattering effects. The most common method for imaging is through the analysis of secondary electrons. Secondary electrons are the ones, that get ejected from the probes atoms through inelastic scattering processes, when the primary electrons hit the sample. These secondary electrons have low energies. They are conducted to detector and a photomultiplier, after getting accelerated with an electric grid. The photomultiplier enhances and transforms this into an electrical signal. The signal output is then displayed as a two-dimensional intensity distribution, that images the surface of the sample. Edges and steep areas appear brighter than flat surfaces, due to an increased emission of secondary electrons.

2.2.2 Energy-dispersive X-ray spectroscopy (EDX)

For analysis of the chemical composition of the samples, energy-dispersive X-ray spectroscopy was used. Here, the electrons in the sample material are being excited with electron beams of certain energies. Once excited, they fall back to lower energy levels, radiating photons with characteristic wavelengths, specific for each kind of atom. Therefore, one can conclude the chemical composition from the measured photon emission spectrum. This method grants information on the atoms, that make up the sample material, and the amount of these atoms. It does not give any information on the composition of molecules. Therefore, one can only estimate the actual molecular composition of the material from the received spectrum.

2.2.3 Transmission electron microscope (TEM)

Transmission electron microscopy functions similar to scanning electron microscopy. A TEM consists of an electron emission source, electromagnetic lenses and electron detec-

tors. An electron beam is focused on a sample, where absorption, elastic and inelastic scattering happens. The main difference to the SEM is that the image is gained through measurement of the electrons that pass the sample. Therefore, the sample has to be thin enough, around 100 nm at most, so that a sufficient amount of electrons can pass through. For investigating our samples, two methods of preparation were tried. The first one was to embed several moth scales in resin and then creating thin slices with a microtome, enabling a view of the cross section of the scales. The embedding is usually used to prevent degassing of the sample, reducing the vacuum quality. This proved to work quite well. The only occurring problem was, that a few scales were ripped out of the sample slices, likely during the cutting process.

Another thing tried, was simply placing scales onto the sample holder, without any embedding. Surprisingly, image quality was high, even though degassing of the sample should have lowered the vacuum. Placing the scale onto the sample holder enabled TEM imaging of the scale surface and measuring the scale with a cathodoluminescence device, which proved valuable for color analysis.

2.2.4 Cathodoluminescence spectroscopy (CL)

Luminescence is a phenomenon that occurs when a surface is radiated with photons or electrons, causing it to emit electromagnetic radiation itself, beyond its black body radiation, which is caused by its temperature. Cathodoluminescence is the emission of photons of characteristic wavelengths, that a material emits, when radiated with high energy electrons. The electron beam elevates the sample to an excited energy state.

When returning to their non-excited state, photons with characteristic wavelengths are emitted, mostly in the visible spectrum. From this emission spectrum, one can calculate the color of the material, but without taking into account any structural or interferential effects. A convenient way to conduct measurements is to create a CL map by scanning an area of a material, which shows at which point which colors are emitted.

2.2.5 Diamond cutter setup

This test was constructed to try and simulate the water run off properties, many *Lepidoptera* wing structures show. To achieve this, a setup (figure 4) was created to produce a structure, similar to the ridges found on scales. Glass slides used in optical microscopy served as base material (Glaswarenfabrik Karl Hecht GmbH & Co KG, Sondheim, Germany).

For carving the ridges, a diamond writer (Hex-scribe diamond writer, Plano, Wetzlar, Germany) was mounted on a micromanipulator (Newport, California, USA), which granted the ability to move the glass cutter along the three space axes. The micromanipulator allows to control the distance between the carved ridges accurately in dimensions of micrometers.

Several trials were necessary to create homogeneous, periodic grooves that did not show microfractures. The best results were carved ridges with a spacing of 25 and 30

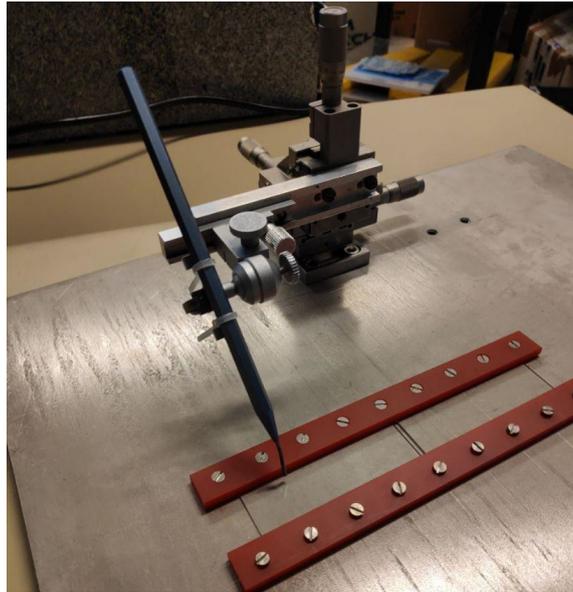


Figure 4: Diamond writer setup for carving grooves

micrometers. Ridges with smaller spacing often displayed a high density of fractures, which was deemed too inaccurate for the experiment.

One problem that occurred, when carving with the diamond writer was that due to the uneven twisting hand motion when turning the steering screws of the micromanipulator, small irregularities appeared in the ridge lines. Also the carving process consumes much time when performed this way. Due to that, a wire was wrapped around the screws and through pulling, a steady, even motion across the glass surface was achieved, which also drastically improved the working time.

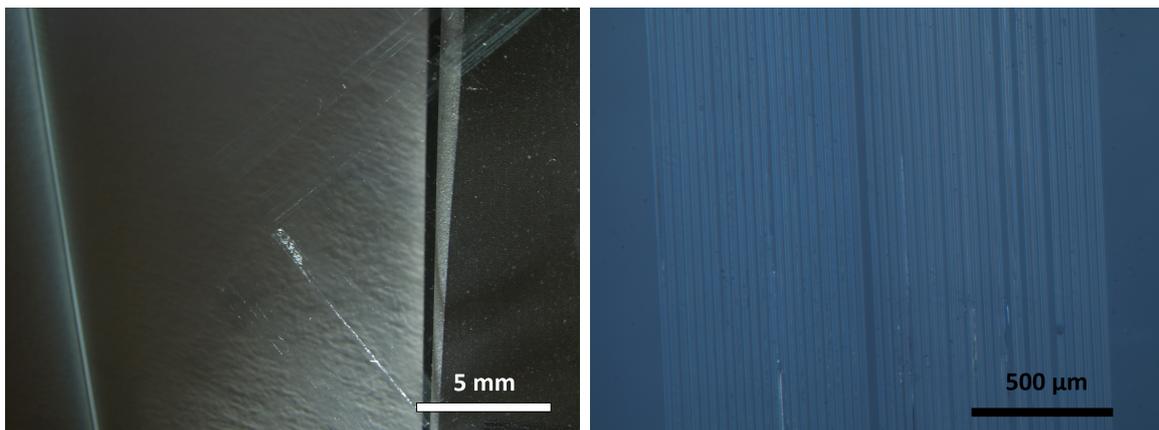


Figure 5: Carved V-shape structure (left) and close up of one of the four lines, each consisting of about 50 grooves (right)

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The final structure, on which tests were conducted, has a V-shaped form, as seen in figure 5. With this shape, water run off tests can be conducted from different directions and angles and with changing water flow directions. The double lining was chosen, to try to lead the water between the lines, and protect the inner part of the V-shape. Each line has 50 carved ridges and an approximate length of 2 centimeters. Due to mistakes made during the carving process sometimes a line was skipped unintentionally, but this should not alter the results in any substantial way. The glass slides themselves were not chemically treated or coated with any kind of substance. Due to the similar structure to diffraction gratings, as shown on the right in figure 5, the glass structures generated iridescent colors.

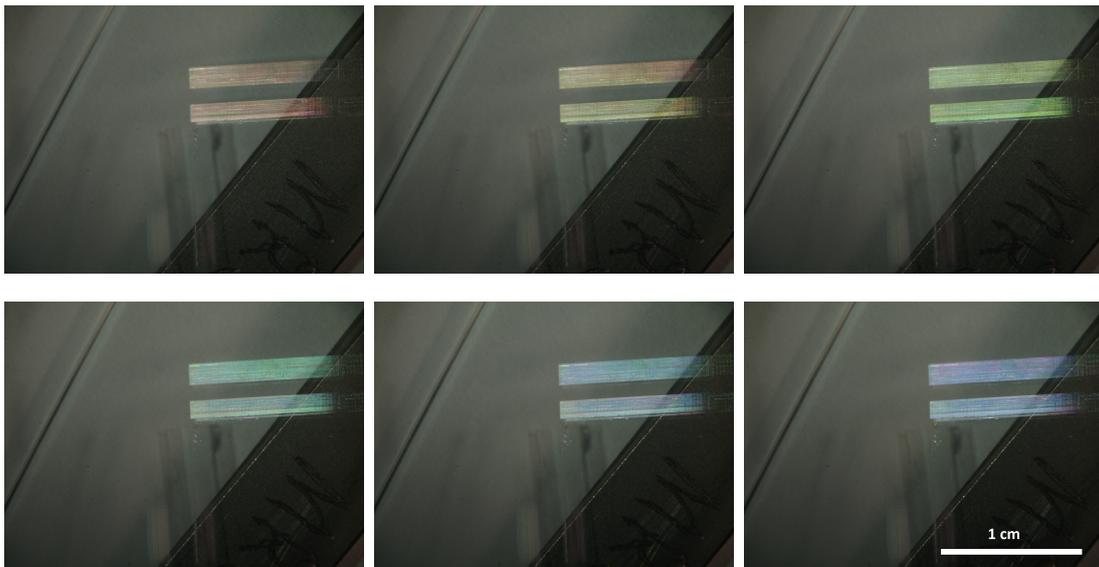


Figure 6: Iridescent colors, generated by the grating structure

3 State of the art

3.1 Theoretical basics of electromagnetic waves

Electromagnetic waves are propagating coupled electric and magnetic fields. Electric fields describe the fundamental interaction between charged particles. Magnetic fields originate from moving charged particles or from the spin of fundamental particles and interact with moving particles or ones that have a spin (e.g. interaction between two electric currents). These fields propagate through space with the speed of light. One has to consider that this speed is dependent on the medium the wave is passing through.

3.1.1 Maxwell equations

Maxwell's equations describe electromagnetism, meaning they state the correlations between electric and magnetic fields, charged particles and electric currents. They are a set of four differential equations and, together with the Lorentz force, describe all fundamental electromagnetic interactions. In vacuum these equations are:

$$\nabla \cdot \vec{E} = 0 \quad (1)$$

$$\nabla \cdot \vec{B} = 0 \quad (2)$$

$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (3)$$

$$\nabla \times \vec{B} = \frac{1}{c^2} \frac{\partial \vec{E}}{\partial t} \quad (4)$$

\vec{E} ... electric field

\vec{B} ... magnetic flux density

c ... speed of light in vacuum

The Lorentz force is defined by

$$\vec{F} = q(\vec{E} + \vec{v} \times \vec{B}) \quad (5)$$

q ... electric charge

\vec{v} ... velocity of the charged particle

When propagating through a medium, the Maxwell equations change to

$$\nabla \cdot \vec{D} = \rho \quad (6)$$

$$\nabla \cdot \vec{B} = 0 \quad (7)$$

$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (8)$$

$$\nabla \times \vec{H} = \frac{1}{c^2} \frac{\partial \vec{D}}{\partial t} + \vec{j} \quad (9)$$

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\vec{D} ... electric displacement field

\vec{H} ... magnetic field

When combining the four Maxwell equations, one obtains the two following sets of wave equations:

$$\nabla^2 \vec{E} = \epsilon_0 \mu_0 \frac{\partial^2 \vec{E}}{\partial t^2} \quad \nabla^2 \vec{B} = \epsilon_0 \mu_0 \frac{\partial^2 \vec{B}}{\partial t^2} \quad (10)$$

One possible particular solution is

$$\vec{E} = \vec{E}_0 \cos(\vec{k} \cdot \vec{r} - \omega t + \phi) \quad (11)$$

This symbolizes a plane wave, where \vec{k} describes the propagation direction of the wave, \vec{E}_0 the amplitude, ω the angular frequency and ϕ the phase.

3.1.2 Electromagnetic waves

Parameters of a wave are its frequency f , wavelength λ , phase ϕ and amplitude E_0 and B_0 . The correlation between frequency and wavelength is defined by $\lambda \cdot f = c$.

When inserting the particular solution from equation 11 into the wave equation, we obtain

$$k = \frac{n \cdot \omega}{c} \quad (12)$$

Here, n stands for the refractive index. It describes how light propagates through a medium and is also the ratio of the speed of light in vacuum to its speed in a medium. The directional energy flux of an electromagnetic wave is represented by the Poynting vector \vec{S} . The intensity I is defined by absolute value of \vec{S}

$$hI = |\vec{S}| = c\epsilon_0 \vec{E}^2 \quad (13)$$

The range of frequencies electromagnetic radiation contains is called its spectrum. A light source usually emits waves with many different wavelengths. Measuring a spectrum can be useful to obtain information on the composition or temperature of a material. There are several different measurement techniques to study the interaction between electromagnetic waves and matter, usually information is displayed through various spectra, such as absorption or emission spectra, where the intensity varies as a function of frequency.

3.1.3 Interference

In physics, interference is the superposition of two waves with equal or nearly equal frequencies, resulting in a new wave with a changed amplitude. If the two waves overlap in a way that they cancel each other out, it is called destructive interference. If the waves are maximally amplified, it is called constructive interference. Whether the waves interfere constructively or destructively with each other is determined by their phase

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difference. If two waves have a time-invariant phase difference, they are called coherent and their wave field is stationary.

3.1.4 Refraction

The phase velocity of an electromagnetic wave changes when it passes from one medium to another, causing a change in its propagation direction. This is called refraction. The phase velocity change is due to the wave inducing a driven oscillation in the atoms of the medium. How the oscillation overlaps with the initial wave, depends on the refractive index of the medium, which is proportional to its permittivity ϵ and permeability μ . The medium's electrons oscillate with the same frequency as the incident wave, therefore its wavelength has to change.

The change of direction depends on the angle of incidence and the refractive indices and is described by Snell's law

$$n_1 \cdot \sin(\theta_1) = n_2 \cdot \sin(\theta_2) \quad (14)$$

- θ_1 ... angle of incidence
- θ_2 ... angle of refraction
- n_1 ... refractive index of the medium on the incident side
- n_2 ... refractive index of the medium on the refraction side

3.1.5 Reflection

When a wave reaches an interface between two media, parts get reflected, parts absorbed and parts transmitted. If the interface is specular, the wave gets reflected at the same angle as the angle of incidence is, in respect to the normal. If the reflecting surface is rough, the reflection is diffuse, meaning there are several different angles of reflection that do not all coincide with the angle of incidence. Therefore no reflected image can be observed, only diffuse light. An ideal diffuse reflection shows luminance, distributed equally all over of the incidence side.

3.1.6 Absorption

Electromagnetic waves propagating within a medium continuously lose their energy. This is called absorption and is described by Beer's law

$$I = I_0 \cdot e^{-\alpha \Delta z} \quad (15)$$

- I_0 ... intensity of the incident wave
- α ... attenuation coefficient

The absorbance of a material is characterized by its attenuation coefficient, which is frequency-dependent. Therefore materials can strongly absorb some wavelengths,

whereas others are let through without losing intensity. The absorbed radiation can then for example be converted into thermal energy.

3.2 Pigmentary colors

Most materials in our world have a body color based on pigments. Pigments consist of either organic or inorganic molecules. For example, organic molecule pigments would be chlorophyll in green plants or melanin in human skin, whereas inorganic pigments could be cobalt blue or lead white.

When light falls onto an object, some parts of it get transmitted, some get absorbed and some get reflected. What we now perceive as color is the part of the light that gets reflected from the object into our eye. This color is called subtractive color, meaning parts of the incident light do not get reflected, thus only a part of the spectrum reaches the eye.

When white light (meaning light containing many different (visible) wavelengths), falls onto a green leaf, containing chlorophyll, the molecule absorbs the blue and red wavelengths. The green part of the spectrum gets reflected, we perceive the leaf as green.

The selective absorption and reflection of electromagnetic waves in atoms and molecules comes from resonance absorption. Electrons bound to atoms and molecules have intrinsic energy levels. To change between these energy levels, electrons can either emit energy, by emitting electromagnetic waves with certain wavelengths, thus losing some of their energy and moving to a lower energy state. Or they absorb a photon, gaining energy and moving to a higher energy state.

All atoms and molecules have discrete electron energy levels, meaning their electrons cannot freely emit or absorb electromagnetic waves with any random energy, but can only interact with waves that give exactly enough energy to move them to a higher state, respectively emit photons so that they lose exactly enough energy to transit to a lower state. Therefore the electromagnetic waves a pigment can absorb or emit are limited to certain wavelengths, which are distinct for each atom or molecule.

These wavelengths of photons, that can be absorbed or emitted by the molecule, are called resonance wavelengths. One important thing to point out is that when an electron is in an excited state, it will, within a certain amount of time, fall back into a lower energy state, once again, emitting energy by photon irradiation. But it does not have to give off all of its energy through photon emission. It is possible for an electron to transfer part of or all of its energy to other electrons, exciting them, or make a crystalline grating or another molecule vibrate. This transferred energy leads to, for instance, an increase in temperature in the object or a chemical reaction, changing or breaking down the molecule structure, and therefore is not emitted as electromagnetic wave that would give color to the object.

Because the absorption peaks of pigments are commonly wide, resulting colors are not very pure. Also scattering surface structures often contribute to a rather matte than specular appearance. Nonetheless bright colors resulting from pigments do exist, as seen in the case of butterflies like *Lycaena phlaeas* or *Ornithoptera priamus poseidon*, which show metallic aspects, but have no periodic nanostructures, two- or three-dimensional,

capable of producing such optical effects, thus only gaining their bright color from pigments. (Serge Berthier, *Iridescence-The Physical Colors of Insects*, 2007, page 49).

Pigmentary colors carry a great role in the world of *Lepidoptera*. Generally, colors in nature have many different purposes. They can function as deterrence against predators or camouflage. Also colors can have intra species signaling purposes, for example between female and male specimen as sexual recognition tools. There are two kinds of scales that can give color to *Lepidoptera*. Firstly are the structural scales. These give color through periodically nano-structured surfaces and inner scale composition. The structural scales will be discussed in chapter 3.3. The other kind of scale is the pigmentary scale. These pigmented scales are found on every *Lepidoptera* and are usually important for various reasons. Also there are some scales to be found that have both, color producing nanostructures and pigments.

For one, pigmentary scales are generally located below the structural scales and absorb the remaining light, that gets transmitted through the structural scales positioned above them, therefore hindering the transmitted light to alter the perceived color, when it gets partially reflected on lower layers of the wing structure, leaving only the fraction of the reflected spectrum that originates from the nanostructure. But they do not necessarily have to hinder the reflection of light completely, they can also give different coloring to *Lepidoptera*, meaning they do not absorb all of the transmitted light, but just a part, reflecting parts of the spectrum back, so that the perceived color of the *Lepidopteran* changes from the color it would have without the pigmented scales to a color that is composed of the combined spectra.

Pigments usually produce colors in the range of red, orange or yellow. Blue and violet are rather scarce, but can be found for instance in some mollusks and crustaceans. (Serge Berthier, *Iridescence-The Physical Color of Insects*, page 4)

The other feature of pigmentary scales comes from the already mentioned selective absorption of incident light, that gets converted into thermal energy, hence heating the *Lepidoptera*. This is key because butterflies need a certain muscle temperature, around 36° to 38° to take off. Also they have a limited temperature range within which their muscles work accordingly without getting damaged from overheating. To heat themselves they either position their wings directly at the sun, if the wings are dark-colored. If the wings are of lighter color, they angle them in a way that the light gets reflected from the wings onto their dorsal side. (Serge Berthier, *Iridescence-The Physical Color of Insects*, page 135,136) There are several pigment families in *Lepidoptera*. First of all melanins, which are present in all species. Their color ranges from yellow to brown and black. There are two kinds of melanins predominant in the insect world, phaeomelanins, which are yellow to red, and eumelanins, which are brown to black. Other important pigments in *Lepidoptera* are Ommochromes, Papiliochromes, biliary pigments (their color tends to be green or blue) and Pterines. Pterin can form small oval granules, called pterinosomes, which can be situated between striae, taking an important role in absorbing and scattering light (Serge Berthier, *Iridescence-The Physical Color of Insects*, page 130).

Some caterpillars, that feed on plants that contain toxic substances, stock these and transfer them to the later developed scales and wing membranes, where there is no

fluid circulation. This has the advantage that the poison is isolated and furthermore, seen from an evolutionary point of view, grants protection against predators. Coloring through these toxins is therefore an additional effect, not the main cause.

3.3 Structural color

3.3.1 Scattering

A plane wave incident on a small particle locally perturbs the local electron distribution and induces an oscillating dipole on the particle. This dipole radiates electromagnetic waves into space that deviate at a certain angle α from the angle of incidence. This deviation effect is called scattering. There are two important kinds of scattering, Rayleigh and Mie scattering. Rayleigh scattering describes scattering effects of electromagnetic waves on atoms, molecules or particles, that have a small diameter compared to the wavelength of the incident wave. The scattering cross-section σ is proportional to the forth power of frequency. Therefore blue light gets scattered stronger than red light, due to its higher frequency. This, for instance, is one of the reasons, why we observe a blue sky during daylight. During sunset, light has to travel a much longer distance through the atmosphere, so the more scattered blue light does not reach the eye, only the less scattered red light does, therefore a sunset appears red.

Mie scattering is the scattering of radiation on particles, whose wavelength is approximately equal to or greater than the diameter of the scattering particles. Examples for this are smoke particles, small ice crystals or water droplets in the air. Here, the scattering intensity depends strongly on the diameter and surface structure. An important aspect of Mie scattering is that the scattered radiation is stronger focused in forward direction, with growing particle diameters. Therefore one can deduce the structure or surface of particles from an angle dependent scattered intensity measurement.

3.3.2 Diffraction gratings

The optical properties of diffractive gratings are based on interference. They have periodic structures of slits or ridges and are therefore either transmissive or reflective. The slits or ridges have a periodic spacing d , which is called grating constant. This constant is substantial for the properties of the grating, and has to be bigger than the incident wavelength to generate diffraction. Two electromagnetic waves that incide onto two adjacent ridges at an arbitrary angle propagate from them in all directions and interfere with each other. If the path difference is equal to a $\frac{\lambda}{2}$ they destructively interfere and cancel each other out. If the path difference is λ they add together and maxima occur. The path difference depends on the angle of incidence. Therefore the requirement for constructive interference is contingent on the the angle of incidence, the grating constant and the wavelength. Thus the observed color of a grating that is radiated with polychromatic light is dependent on the observation angle, the incident light gets split up, similar to a prism. Usually there are several angles at which a color can be observed. The orders of interference maxima are enumerated with the integer m , the observed

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intensity declines with increasing values of m .

The formula which describes this optical effect for normally incident wave maxima under the viewing angle θ is called grating equation:

$$d \sin(\theta) = m\lambda$$

3.3.3 Thin-film interference

A major source of structural colors derives from thin-film interference. Here, an incident wave gets separated into two parts at the surface of the film, a reflected and a refracted beam. When reaching the lower surface, the same thing happens to the refracted wave again. Of interest are now the initial reflected beam and the one that gets reflected at the lower side. If these beams are in phase, constructive interference happens, if they have opposite phases, they cancel each other out. The phase difference depends on the length of the path the second beam has to travel through the film. Therefore, thin-film interference is dependent on the initial angle of incidence and the refractive index of the film and can be calculated with the following formula:

$$2n_2 \cdot d \cos(\theta_2) = m\lambda \quad (16)$$

One thing to consider when calculating thin-film interference phenomena is that phase shifts of 180° occur when light gets reflected from materials that have a higher refractive index than the material on the incident side. When the light is incident from a material with a higher refractive index, no phase change happens. Also, depending on the reflectivity of the film surface, multiple reflections and refractions of one beam can happen, but usually only the first beam is important, the others have more or less insignificant intensity. The resulting pattern of light from interference can either be light and dark, if the light source is monochromatic, or differently colored areas, for polychromatic light sources.

3.3.4 Multilayer interference

The reflection factor of a thin layer is rather weak. A much better reflectivity is given by multilayer structures, where two different materials are stacked periodically. The working principle is the same as with the above mentioned thin-film interference, where one material is the one on the incident side and the other is the thin film. As there are more layers, the reflectivity increases. Therefore a high number in layers results in a high reflectivity, and a very precise reflection peak, which therefore results in very pure colors. In the world of *Lepidoptera*, multilayer structures consist mostly of periodically changing patterns, that are composed of organic layers and layers of air. In other realms of nature, one can also find materials like nacre, which gains its iridescence from a multilayer structure of aragonite and elastic biopolymers or the Australian beetle *Notasacanta dorsalis*, which can switch its color by altering the humidity of one of its layers, therefore changing the refractive index (Serge Berthier, *Iridescence-The Physical Colors of Insects*, 2007, page 55).

As one can see, multilayers can consist not only of solid materials, but also of liquids or gases, for interferential effects, only the refractive index matters. It has to be mentioned, that until now, absorption was not considered. For animals who want to emphasize their structural colors it is important to reduce the back scattered light, which propagated through the multilayer structure, because it would alter the returned color spectrum. Absorptive materials are added beneath to reduce this background reflection. Therefore only the iridescent colors are returned but the transmitted, disruptive ones are absorbed at the lower levels of the structure (Kinoshita Shuichi, Structural Colors in the Realm of Nature, 2008, page 25). As a great example of structural color serves the tropical butterfly *Morpho peleides*. It gains its bright blue color from a ridge structure that shows a multilayer structure in cross section view, consisting of several overlapping lamellae and absorbing pigmentary ground scales.

3.4 Hydrophobicity

3.4.1 Adhesion

An interesting property of many *Lepidoptera* scales is their strong hydrophobicity, partly due to their surface structure. Hydrophobic surfaces repel water extremely well, droplets most often leaving no water residue behind at all. This can have two reasons. One is, that a material can be composed of or coated with hydrophobic molecules. The other reason is due to microscopic structures of the surface.

Adhesion is the tendency of different particles to increase their contact area. This is due to molecular forces of attraction. The stronger these forces are, the more the contact area increases. Counteracting that, cohesion pulls equal particles together. Therefore, the wettability of a surface depends on the overall sum of cohesive and adhesive forces. If adhesion outweighs cohesion, a fluid increases its contact area with the surface. To the contrary, if cohesion outweighs, the fluid minimizes the contact area with the surface.

Adhesion between non-gaseous and gaseous phases is usually very small and will not be considered here. To measure the adhesive force a surface exerts on a fluid, the contact angle is measured. When the contact angle is known, the interfacial tension can be calculated and in case of water, determination of whether the surface is hydrophilic, hydrophobic or superhydrophobic is possible. A contact angle under 90° means, that the surface is hydrophilic, greater angles mean its hydrophobic and angles around 150° and more mean its superhydrophobic.

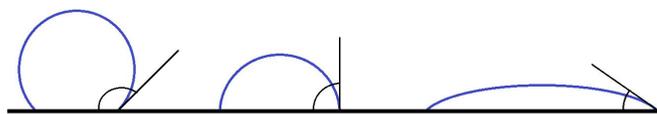


Figure 7: Decreasing contact angle from left to right, with the left figure displaying wetting of a hydrophobic surface (large contact angle) and high wettability on the right.

3.4.2 Wettability of butterfly wings

When viewing butterfly scales, one can see, that they show specific ridge structures. These ridges have usually spacings of submicrometers to about 2 micrometer, depending on the species. This effectively creates air pockets, decreasing the contact surface between water and scales drastically. Therefore, the adhesion is reduced and a mechanical hydrophobic surface is formed (G. Chen, Q. Cong, Y. Feng and L. Ren, Study on the wettability and self-cleaning of butterfly wing surfaces).

Probably the most famous natural hydrophobic surface is that of the Lotus plant. The working principles are nearly the same as with butterfly scales. The surface shows 10 to 20 micrometer high conical structures with a distance of about 10 to 15 micrometers. Because of these structures, water droplets have minimal contact are with the leaf surface. They are also coated with a wax, which further improves the hydrophobicity.

Another effect of the ridge structure concerns the run off direction of water. A droplet rolls off alongside the radial outward direction, meaning parallel to the ridges, much more easily than orthogonally to the ridges. This has the biological advantage that dust or other small polluting particles are washed off by water droplets, away from the torso of the butterfly (Yongmei Zheng, Xuefeng Gao and Lei Jiang, Directional adhesion of superhydrophobic butterfly wings).

4 Experimental approach

4.1 Analysis of coloration effects

4.1.1 Mongolian moth

A lot of the structural colors of *Lepidoptera* originate from the ridges on their scales. For example, the ridges on the scales of *Morpho peleides* show multiple overlapping lamellae, creating Christmas tree-like shaped ridges from a cross sectional view. This overlapping structure acts as a multilayer reflector, composed of layers of chitin and air. It displays bright, metallic, iridescent blue shimmering, because of the high density of ridges and their stacked layers.

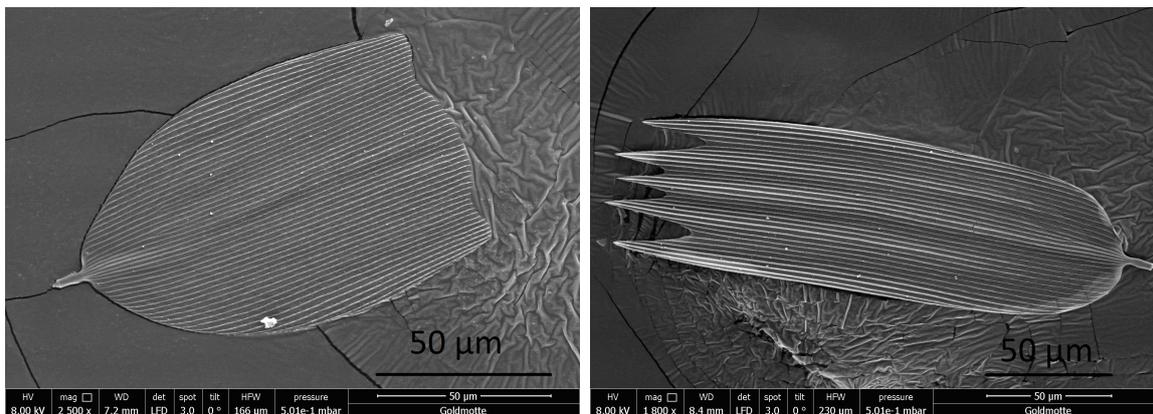


Figure 8: Two scales, taken from the Mongolian moth, showing very different shapes. Under observation with the light microscope, no color differences could be determined.

Another option for generating structural colors with butterfly scales is the formation of multilayers within the basis of the scale on which the ridges reside. Here, several layers of chitin make up the multilayer reflector. Depending on the dimensions and the observation angle, colors are generated through interference.

When observing the upper side of the scales of the Mongolian moth, a periodic structure of ridges can be found. The distance between ridges has been measured to be $1.6\mu\text{m}$ on average. The lamellae show only small overlapping areas. Because of this, iridescence based on multilayer ridge structure was excluded from the possible reasons for the moths color. Under the optical microscope, iridescent colors can be seen. A reason for this may be the entirety of the periodic structure of the ridges on each scale. Because of their periodicity, they function as a diffraction grating, producing angle dependent color. Multiple different colors can be found on one scale alone, when illuminating it from certain angles.

For these reasons, it was assumed, that the color of the moth is mainly based on pigmentation, the iridescent effects of the periodic ridge structure should not be able to

4 Experimental approach

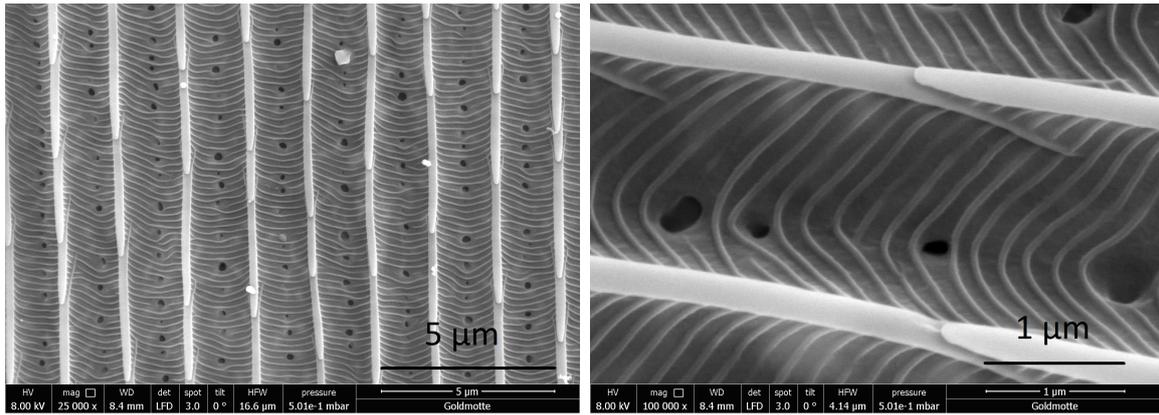


Figure 9: Mongolian moth: Image of the ridge structure on top of the scales. Slightly overlapping lamellae can be seen, with the connecting counter striae in between.

create the observed quality of golden color, especially because the angle dependency of the colors would not allow this. Still, it has to be mentioned, that it can not be ruled out completely, that the coloration does not have structural roots, because no cross section analysis was made.

4.1.2 Common clothes moth

The common clothes moths that were investigated showed practically the same periodic ridge structures on top of the scales as was seen with the Mongolian moth. The difference was that the whole structure was a shrunken version of the Mongolians structure. The distance between two ridges is about $1.1\mu\text{m}$. This is another hint towards pigmentary colors, or at least that the ridge structure does not contribute to the overall color of the moths.

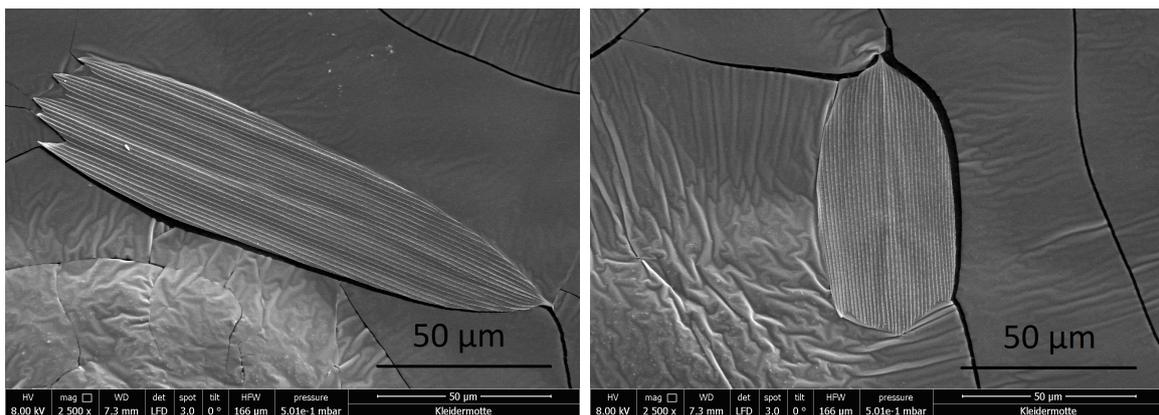


Figure 10: Just as with the Mongolian moth, the common clothes moths' scales show different shapes, but the same colors, when viewed with the optical microscope.

4 Experimental approach

The grating gap difference of about half a micrometer compared to the Mongolian moths structure would change the optical grating effects severely, therefore one would not observe the yellow/golden color, that both moths generally display. While examining cross sections of the scales, two kinds of scales were found. One was composed of a thin membrane, with the striae mounted on top. The other had two membranes, connected with supporting struts. They also had the same ridge structure on top of them. When observing the scales from above with the transmission electron microscope, a network-like structure was revealed to be between the two membranes, showing thick junctions from which small branches emerged. This structure should be the strutting that was found while examining the cross section.

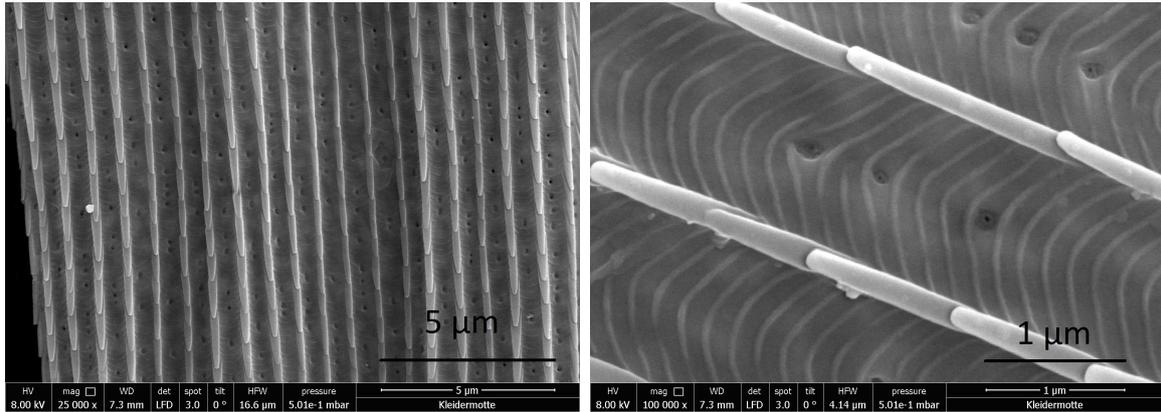


Figure 11: SEM images of the scale surface, with structures very similar to the Mongolian moth, but of smaller dimensions

Measurement with the cathodoluminescence device showed that some areas of the scales, especially the junctions within the scale, produce green and yellow color, while the edges of the ridges lean towards blue. (Shown in figures 12 and 13) Summarizing these facts, it was concluded, that, as far as it is possible to discern at the moment, the color of the common clothes moth derives mainly from pigments. The impact of the inner structure, observed in some scales, is hard to discern, because optical observation did not show the colors, measured with the CL device. So therefore, at least some of the moths color should derive from structure, generated by the knots on the inside or the ridge gratin on the outside or a combination of both.

4 Experimental approach

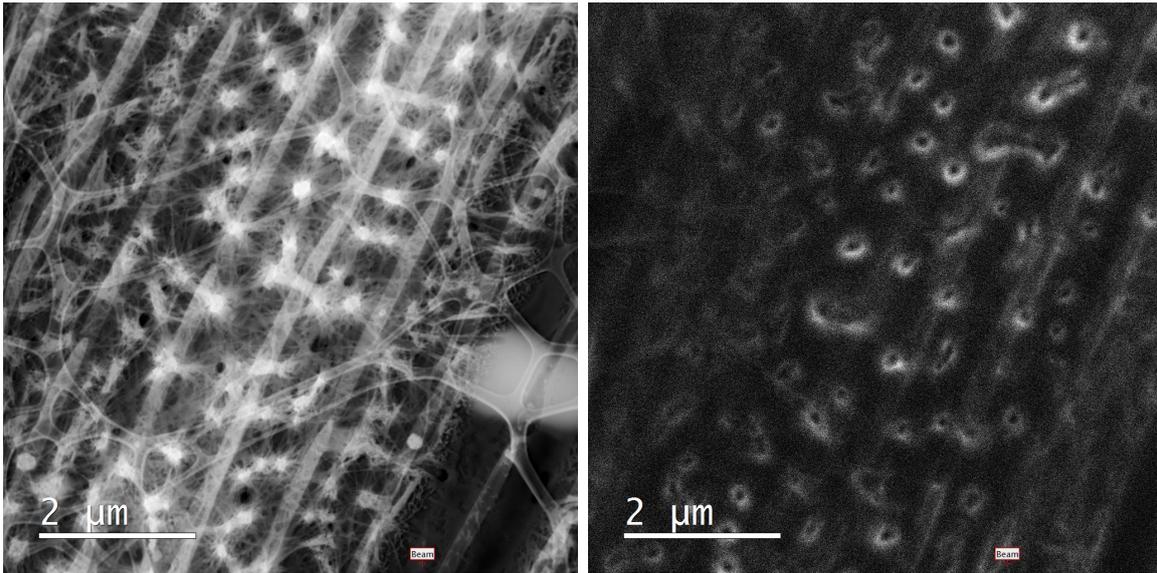


Figure 12: Image of the CL-scanned region, where knots were found (left), high emission intensities were found at the surface of the knots, the white rings indicate, that the emission is due to surface effects (right)

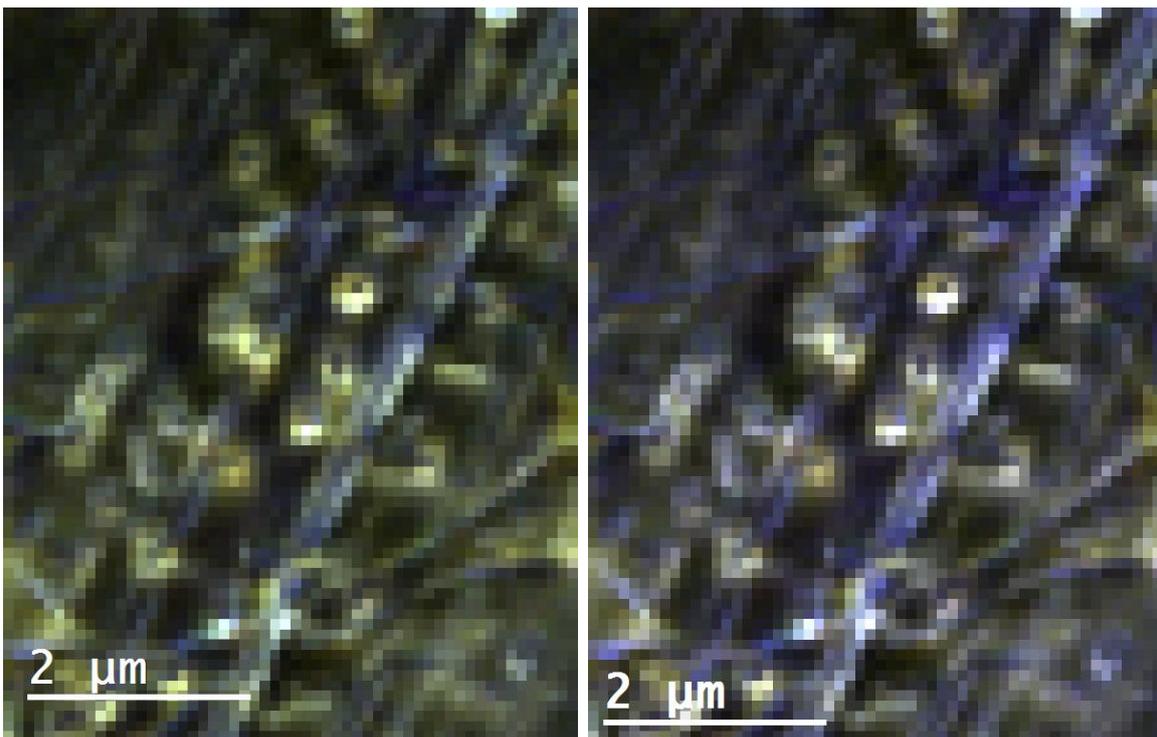


Figure 13: True color representation, before (left) and after (right) corrections, showing, that the knots emit more or less green light, while the ridges tend towards blue

4 Experimental approach

4.1.3 Golden bark

SEM investigation showed three different types of structure layers of the bark. A very thin top layer, with a thickness of only a few nanometers. The surface appeared very smooth, no indications of structuring visible. EDX analysis showed, that apart from the expected carbon and oxygen, small amounts of silicon and calcium are present.

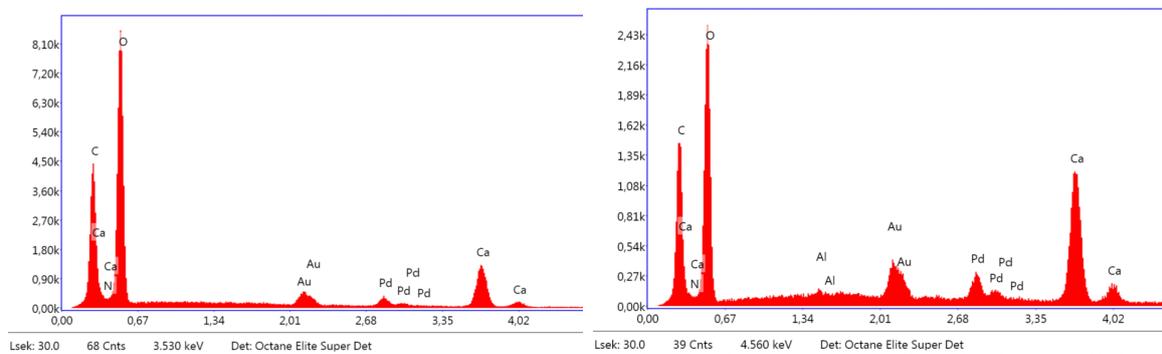


Figure 14: EDX analysis, showing the proportion of calcium in the particles found in the bottommost layer, indicating, that the phytoliths are composed of calcium oxalates

The middle layer seems to be composed of a multitude of homogeneous layers. The middle layer itself has a thickness of about $40 \mu\text{m}$. The thickness of the individual layers was measured to be about 280 nm on average, however the standard deviation amounts to nearly 30%, showing the irregularity of the thicknesses. Displayed in figure 17, in some places, much thinner layers can be seen. It can not be confirmed, whether these are distinct layers or just fracture fragments of the bigger layers.

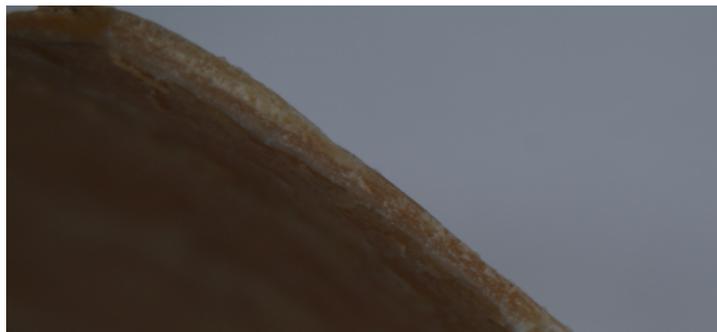


Figure 15: Cross section of the bark with a thickness of approximately half a millimeter.

In figure 15 a cross section of the bark is shown. Here, one can see, that the coloration appears to be the same for the surface and the cross section, with the cross section turning partially whitish only after a depth of about a fourth of a millimeter.

4 Experimental approach

The bottommost structure consists of a multiple layers as well. Some parts of the lower layers showed nearly spherical shapes. Scanning with EDX showed that the particles consist of calcium oxalates. These mineral deposits are called phytoliths and are formed in the tissue of several plants. They consist of either silicon dioxide, calcium oxalates or calcium carbonate and have mainly structural aiding or defense purposes. (Susan C. Mulholland, George Rapp, Phytolith Systematics: An Introduction, 1992)

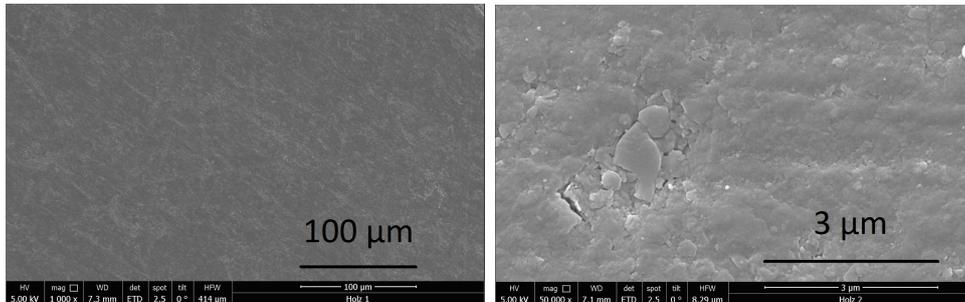


Figure 16: The left picture shows the smooth surface of the bark, one the right side, one can see a closer up picture.

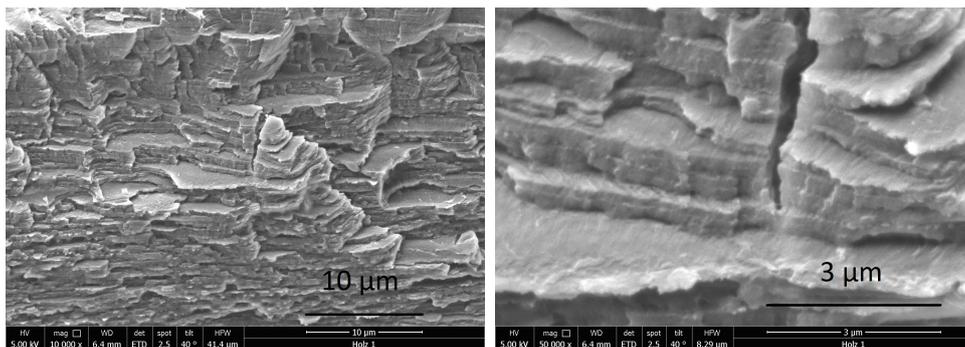


Figure 17: Fractured area, showing the middle layers of the bark.

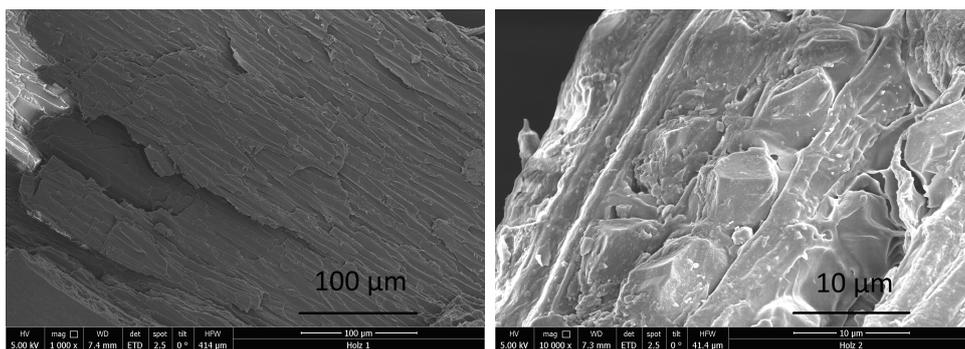


Figure 18: Inner layers, close ups of the liber (left) and incorporated phytoliths (right)

4 Experimental approach

In conclusion, the results of this investigation indicate that the Mongolian bark does not have a color based on structural effects. The equal coloration of the inner side and outer side suggests that the radiant appearance of the surface derives from pigmentation and minimal scattering, due to the smoothness of the surface. Multilayer effects were ruled out, due to the high variance in thickness of the middle structures layers, because no angle dependent color change could be observed and the bark cross section showing the same color as the surface.

4.2 Carved glass structures

4.2.1 Water run off

The water run off tests were conducted in two kinds of ways. First, small water drops, with a diameter of about 4 millimeters, were placed above the pointed end of the V-shape. Then, the glass slide was tilted in a way that the water would flow right in the middle between the two halves of the V-shape. The tilting angles were varied from 30° to 80° . With the greater angles, approximately from 50° to 80° , the water flow shifted slightly in the direction of the ridge structure, but generally ran down the glass slide nearly along the same line as before having contact with the structure. With smaller tilting angles, the direction changing effect improved slightly, the water flow was decelerated, when moving over the structure, and moved longer along the inner side of the V-shapes edges, but did not move onto the carved structure, it eventually returned to moving directly downwards.

Better effects were produced, when the water flow was directed nearly parallel to the ridge lines and the tilting angles were small. Here, the water followed the lines for some lengths, smaller water drops were even impeded by the structure for several seconds, before moving on.

All in all, the glass structures showed little effect on the direction of the water flow. Guidance of water was accomplished, but only to minimal extent. When using larger quantities of water, no clear impact on the moving direction could be observed at all, the same applies for larger tilting angles, where the velocity of the water seemed too high for the structure to take any influence. The hydrophobic effect that *Lepidoptera* wings show could not be recreated. It is expected that the reason for this is the width of the carved grooves. Because the tip of the diamond cutter was rather broad, water drops did not sit on top of the carved structure, but filled up the spacing between the ridges. Therefore, instead of decreasing the adhesive forces, they were increased.

4 Experimental approach

4.2.2 Color enhancement

Another thing that was investigated, was the possible enhancement of structural colors through pigmented backgrounds. As one can see with, for example, the *Morpho peleides* butterfly, if only the structural scales are viewed, a lot of the colors intensity is lost. Only in combination with the pigmented ground scales, the bright blue color can be seen.

To simulate this, the carved glass slides were used as structural color generators. The ridges were sufficiently periodical to act as diffraction gratings, producing angle dependent colors. Colored paper was used as background coloration. Viewed through an optical microscope, no noticeable change in intensity or color could be observed. Whether this is due to the colored background having no effect can not be ascertained. Because the carved structure itself and the angles, at which color can be observed, are so small, there is also the possibility, that an actual enhancement effect was overlooked.

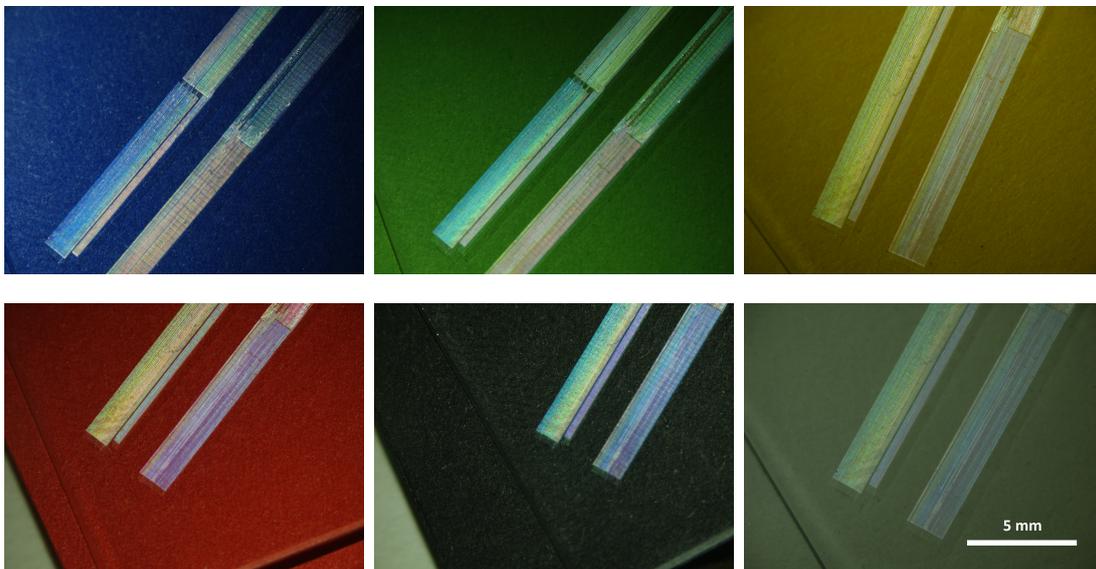


Figure 19: Iridescent colors generated by the ridge structure with underlying colored paper to simulate a pigmented background, similar to pigment scales beneath structure scales.

5 Summary and outlook

The aim of this study was to determine, whether the Mongolian moth, the common clothes moth or the Mongolian bark produce their colors through structures. It has also been tried to simulate the wettability properties of hydrophobic butterfly wings.

Scanning electron microscopy showed that the surface structure of the Mongolian moth and the common clothes moth are really similar. Only the dimensions of the clothes moths scale structures were smaller. The structure did not display any capability to producing interferential color, besides diffraction grating effects. Therefore the color of both moths seems to derive mostly from pigments, as far as it can be determined to this point. The CL true color imaging of the clothes moth showed that the knots emit mainly green light, though errors could have happened, when applying the spectral corrections. It has to be noted that when considering literature descriptions of how structural colors look (shining, bright, metallic), the possibility still remains that some facts were not considered or are unknown and, especially the two moths have some hidden features, that grant color through structural effects. Further results could be achieved, if spectral measurements were made. This was not possible with our current abilities, due to the moth's wings being too small, but may potentially deliver more accurate means of discerning the fact, whether the colors derive solely from pigments.

Regarding the Mongolian bark, the measurements exclude the possibility of structural coloration pretty clearly. Its color should definitely derive from pigments, considering the color of the inner parts of the bark being equally colored. The radiant appearance being caused by the very smooth lacquer-like surface. One thing of interest that could possibly be examined in the future is the toughness of the bark, which seemed rather high, though no actual tests were conducted in this direction, this was just observed during sample preparations. This would make sense: there are few bushes and trees in this part of Mongolia and the few that are there need to protect themselves heavily from being eaten by camels, goats and sheep.

As for the wettability tests and simulations, generated effects were rather insignificant. Better results should be achievable, if a diamond cutter with a sharper tip is used to create a higher number of smaller, thinner grooves. As for examining the impact of pigmented background on the color intensity of structural colors, spectroscopic methods should be used, to grant more sufficient data. Observation with the optical microscope seemed to be too inadequate for accurate evaluation. Also increasing the size of the structured area should help with the assessment. Testing other materials or combining materials may be interesting, like the application of a lacquer coating before the carving process. The combination of hydrophobic and hydrophilic surfaces, which is already a subject of scientific research, can result in fascinating results, for example as guidance structures for liquids. The enhancement of adhesive forces through surface area enlargement, that were observed from the water run off tests with the glass slides, may also be of use in this kind of research direction. For example, guiding microscopic amounts of fluid along predetermined paths without external stimulation.

Looking forward, this field of research will surely develop further and further, proving more interesting every time. The possibilities biomimetics will open up for our society

will hopefully change the way, how we deal with our world, granting better ways for the future in terms of coping with environmental pollution and other issues.

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7 Glossary

biomimetics	imitation of systems found in nature to solve complex technological problems
chrysalis	developmental stage between insect larva and adult insect
counter-striae	connect striae, also called cross ribs
dorsal	backside (of an animal)
electromagnetic waves	coupled oscillating, propagating electric and magnetic fields
field emission cathode	cathode that emits electrons through the application of an electric field
grating constant	spacing between two ridges on a refraction grating
hot cathode	cathode that is heated to emit electrons more easily
hydrophobicity	the property of a material to repel water
interference	superposition of electromagnetic waves, resulting in the extinction or amplification of the wave
iridescence	gradual change of color as the angle of observation changes
lamellae	components of which striae are composed of
Lepidoptera	an order of insects, including butterflies and moths
liber	inner bark of plants
light	electromagnetic radiation with wavelengths between 380 nm and 750 nm
luminescence	emission of light because of supplied energy that is not converted into heat
micromanipulator	device for making small movements in the range of micrometers

7 Glossary

microtome	device used to cut thin sclices of material
multilayer	periodically arranged layers of materials with different refractive indices
photomultiplier	enhances weak electromagnetic signals
phytoliths	microscopic crystals that plants store for stability and defense purposes
polychromatic	radiation composed of at least two different wavelengths
refraction	change of the propagation direction of an electromagnetic wave when crossing a material interface
refractive index	describes how light propagates through a medium and how much its path gets changed when crossing an interface between two different media
striae/ridge	longitudinal lines composed of overlapping lamellae
structural color	color produced by periodic micro- and nanostructures
ventral	frontside (of an animal)

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8 Appendix

Mongolian moth: Dimension measurements

Table 1: Distance between striae [μm]

1.62	1.49	1.51	1.43
1.62	1.49	1.54	1.3
1.52	1.46	1.52	1.62
1.62	1.52	1.54	1.62
1.62	1.43	1.57	1.65
1.62	1.54	1.57	1.68
1.54	1.54	1.68	1.41
1.55	1.57	1.46	1.62
1.68	1.49	1.54	1.32
1.68	1.52	1.49	1.34
1.62	1.43	1.49	1.46

Table 2: Thickness of striae ridges [nm]

232.43	243.24	227.03
249.19	244.24	243.24
243.24	245.24	227.03
254.59	246.24	237.84
243.24	247.24	243.24
259.46	248.24	211.35
254.05	249.24	227.03
264.86	250.24	221.62
237.84	251.24	249.19
243.24	252.24	243.24
270.27	253.24	254.24

Table 3: Distance between counter striae [nm]

113.51	118.92	147.03
124.32	130.27	117.84
130.81	121.62	125.95
135.14	136.76	143.78
130.27	135.68	124.32
125.41	92.97	145.95
129.73	128.65	114.59
129.73	129.73	137.84
129.73	162.16	121.08
122.16	150.81	121.62
128.11	123.24	149.19

Table 4: Diameter of the dents on the scales [nm]

178.38	232.43
194.59	156.76
205.41	194.59
145.95	169.19
145.95	133.51
140.54	210.81
125.41	211.35
200	192.97
153.51	146.49
164.32	195.14

Common clothes moth: Dimension measurements

Table 5: Distance between main striae [μm]

1.11	1.03	1.05	1.08
1.13	0.98	1.08	1.09
1.10	0.97	1.06	1.11
1.10	0.95	1.04	1.12
1.06	0.91	1.04	1.05
1.07	0.88	1.04	1.06
1.05	0.92	1.04	0.96
1.10	0.92	1.04	1.05
1.05	0.98	1.02	1.06
1.03	1.05	1.06	1.11
1.04	1.04		

Table 6: Thickness of striae ridges [nm]

167.84	154.86
162.70	146.49
157.30	146.22
154.32	140.81
155.68	146.22
149.46	141.08
146.76	154.32
171.35	162.70
160.54	159.73
163.51	170.54
162.97	164.86
154.32	

Table 7: Spacing between counter striae [nm]

113.51	103.51
97.84	127.03
111.08	101.08
114.86	123.51
95.95	121.62
108.11	104.32
110.27	110.27
101.62	111.89
100.00	140.54
118.92	108.11
107.84	101.08
112.70	

Table 8: Diameter of the dents on the scales [nm]

122.70	116.22
119.19	129.73
105.68	130.00
127.03	102.97
113.51	145.95
133.24	105.41
108.11	121.89
122.70	120.00
119.46	130.00