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The Colouration of Bird Feathers explained by Effective-Medium Multilayer Modelling

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Chapter 1

INTRODUCTION

The magnificent colouration phenomena that are found in nature have fascinated humans for a long time. Two fundamentally different mechanisms can be distinguished that generate these natural colours: 1) via pigments that selectively absorb light, and 2) via interference effects produced by nano-scaled structures. This thesis focuses on the colouration of bird feathers, specifically on the colours of the barbules, the tiny branches of the feathers (Hill and McGraw, 2006; Kinoshita, 2008). Although the physical understanding of pigmentary colouration in birds is well established (Hill and McGraw, 2006) and the structures that are causing the brilliant colouration of birds have been thoroughly investigated (Durrer, 1977; Kinoshita, 2008), we are still lacking a proper comprehension of the optical effects. These include, for instance, the optics of disordered, short-range and medium-range ordered photonic structures. The focus of this thesis lies in advancing our understanding of one-dimensional medium-range ordered structures that are frequently found in the barbules of bird feathers. These structures can presumably be well-treated as a multilayer. This has indeed been done in various studies already decades ago (see for instance Durrer, 1977; Land, 1972), but these approaches were limited to highly regular multilayer approximations. The application of numerical methods, enabled by modern-day computational processing speeds, greatly enhances the information that we can gather and analyse, and hence allows us to study the effects of non-ideal multilayers in great detail. The effective-medium multilayer modelling technique that I use in this work is not computationally heavy, mathematically simple, and therefore very accessible. It is used in combination with several experimental methods, specifically spectrophotometry and imaging scatterometry, to study feather colours at a micro- and macro-level. An extensive analysis of the measured and modelled spectra, combined with the comparison of different birds with variously coloured feathers, will help us to further understand how the colours and the spectral and spatial characteristics of these particular structures are created.

A brief history about the optics of natural structural colours

The study of the physics behind the colours of bird feathers started three and a half centuries ago when the Royal Society published its first scientific best-seller, Robert Hooke's Micrographia (Hooke, 1665). This wonderfully illustrated book describes meticulously the intricacies of bird feathers, among many other topics. About the feathers of the 'glorious' peacock, Hooke writes: '…their upper sides seem to me to consist of a multitude of thin plated bodies which are exceeding thin, and lie very close together, and thereby, like mother of Pearl shells, do not onely reflect a very brisk light, but tinge that light in a most curious manner […] as arise immediately from the refractions of the light, […] yet by looking on them against the Sun, I found them to be ting'd with a darkish red colour, nothing a-kin to the curious and lovely greens and blues they exhibited'. In other words, Hooke already understood that the peacock feathers are coloured due to the reflection of light from "thin plated bodies" within the feather barbules, a phenomenon we now refer to as structural colour. The 'darkish red' observed upon transmission of sunlight is instead due to the absorption by melanin contained in the feather material and therefore referred to as pigmentary colour.

Although Hooke's physical understanding of the observed structural colouration of the peacock was perfectly correct already in 1665, it took nearly three centuries to confirm the various mechanisms of structural colouration in general. At the beginning of the 20th century, pigments were well understood to be a property intrinsic to the material, but the structures that are responsible for the colouration beyond pigments, were still somewhat debated (Mason, 1923a; Mason, 1923b; Onslow, 1923; Süffert, 1924). While the mathematical description of the multiple reflections/transmissions of light in "regularly stratified materials" or regular multilayers has been formulated by Lord Rayleigh (Strutt, 1917), it was not until the development of the electron microscope in the 1930s that nano-scaled structures were actually revealed in various beetles, butterflies and bird feathers (Anderson and Richards, 1942; Land, 1972; Durrer, 1977; Kinoshita, 2008).

In the past decades, the rising awareness of technological applications of structural colours in the field of photonics has sparked new interest into the particular structural details of materials that cause specific optical effects (Parker and Townley, 2007; Schroeder *et al.*, 2018; Xiao *et al.*, 2020). Also, the readily accessible numerical techniques that are now feasible due to modern-day computer speeds have aided a better understanding of the structural parameters that are responsible for specific spectral phenomena (Kolle *et al.*, 2010; Wilts *et al.*, 2012; Wilts *et* *al.*, 2014).

The basis of colouration

But let us start to discuss the physics of colours in a bit more detail. Generally, if an object is inhomogeneous, with components having different refractive indices, incident light will be scattered randomly inside the medium. In the absence of pigment, an object illuminated by white light will scatter the incident light diffusely to outside the object, so that it will have a white colour. This is for instance the case with white goose feathers, where the keratin matrix contains numerous air channels so that a diffuse white colour is created (Stuart-Fox *et al.*, 2018).

If the feathers contain pigment, it will absorb part of the scattered light, so that the diffusely scattered light obtains a spectral composition complementary to the pigment's absorption spectrum. For instance, the breast feathers of the songbird *Parus major* contain blue-absorbing carotenoid, resulting in a striking yellow colour (Shawkey and Hill, 2005). As the chemical nature of the pigment determines the absorption spectrum, pigmentary colouration is also addressed as chemical colouration.

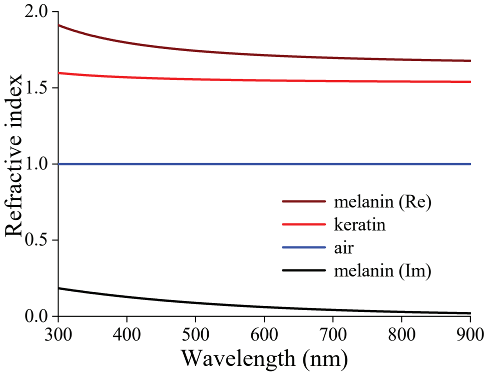

Figure 1.1. Refractive indices of the media of bird feather barbules. The imaginary component of the refractive index, representing absorption, of the main feather material, keratin, is negligible, but this is not the case for melanin. The wavelength (λ) dependence of the refractive index of keratin is given by $n_k = A_k + B_k\lambda^{-2}$ with $A_k = 1.532$ and $B_k = 5{,}890$ nm^2 (Leertouwer *et al.*, 2011) that of melanin by: $\tilde{n}_m = n_m - ik_m$ where $n_m = A_m + B_m\lambda^{-2}$ and $k_m = a_m \exp(-\lambda/b_m)$, with $A_m = 1.648$, $B_m = 23{,}700$ nm^2, $a_m = 0.56$, and $b_m = 270$ nm (Stavenga *et al.*, 2015); the refractive index of air is wavelength-independent, with $n_a = 1$.

The most common pigment of bird feathers is melanin, which is embedded in the keratin matrix of the feathers' barbules in small organelles, the melanosomes (McGraw, 2006; Prum, 2006). These sub-micrometer-sized structures sometimes also contain air. In many bird feather barbules, the melanosomes are not randomly arranged, but rather are structured in regular patterns. The orderly arranged melanosomes then create structural colours, especially because the refractive index of the constituent melanin (and also that of air) distinctly differs from the refractive index of the surrounding keratin (Figure 1.1).

The colouration of the feathers is quantitatively described by their reflectance spectrum. This thesis shows that the feather structures can be treated as dielectric multilayers and that the reflectance spectrum of a feather can be modelled with a matrix transfer function for multilayers. The feather structure therefore is sliced into thin layers for which the effective refractive index can be calculated from the relative concentration of the various material components. It hence is useful to first consider the optics of thin films (Figure 1.2).

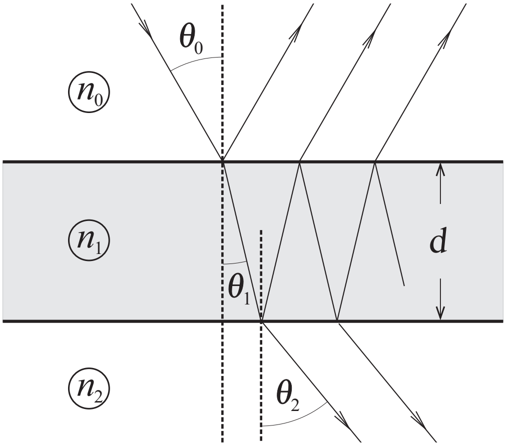

Figure 1.2. A thin film with thickness d and refractive index n_1 in between two media with refractive indices n_0 and n_2. Light incident at angle θ_0 is refracted with angles θ_1 and θ_2 .

Reflectance of a thin film

For a single thin film, the reflectance is derived from $R = |r|^2$, where r is the reflection coefficient of the thin film, which is given for both TE- and TM-polarized light by the Airy formula (Yeh, 2005, Stavenga, 2014)

$$r = (r_{01}+r_{12}\, e^{-2i\varphi}) / (1+r_{01}\, r_{12}\, e^{-2i\varphi}) \qquad (1),$$

where $\varphi = kn_1 d \cos\theta_1$, d is the thickness, and r_{01} and r_{12} are the reflection coefficients of the interfaces given by the Fresnel formulas:

$$\text{TE:} \qquad r_{j-1,j} = (n_{j-1}c_{j-1} - n_j c_j) / (n_{j-1}c_{j-1} + n_j c_j) \qquad (2a)$$

and

$$\text{TM:} \qquad r_{j-1,j} = (n_{j-1}c_j - n_j c_{j-1}) / (n_{j-1}c_j + n_j c_{j-1}) \qquad (2b),$$

with $j = 1, 2$; $c_j = \cos\theta_j$.

For the case where the refractive indices are real, the reflectance is

$$R = \frac{r_{01}^2 + r_{12}^2 + 2r_{01}r_{12}\cos 2\varphi}{1 + r_{01}^2 + r_{12}^2 + 2r_{01}r_{12}\cos 2\varphi} \qquad (3)$$

The reflectance then has extrema (minima and maxima) for $\sin 2\varphi = 0$, or, $\varphi = u\pi/2$, with $u > 0$ and integer, or, for $2d\cos\theta_1 = u\lambda/2$. When the refractive index of the media are $n_0 < n_1 < n_2$ reflectance maxima occur for $u = 0, 2, 4, \ldots$, *i.e.*, for $u = 2m$, with $m = 0, 1, 2, \ldots$. The condition for constructive interference then is $2d\cos\theta_1 = m\lambda$. When $n_0 < n_1 > n_2$ maxima occur for $u = 1, 3, 5, \ldots$, *i.e.*, for $u = 2m + 1$. The condition for constructive interference then becomes $2d\cos\theta_1 = (m+½)\lambda$. So-called destructive interference, causing reflectance minima, occur at the intermediate integer u-values. The spectral composition of light reflected by a thin film depends on the angle of light incidence, and this phenomenon is called iridescence. This colouration is essentially due to the physics of how light interacts with an object, and structural colouration therefore is also addressed as physical colouration.

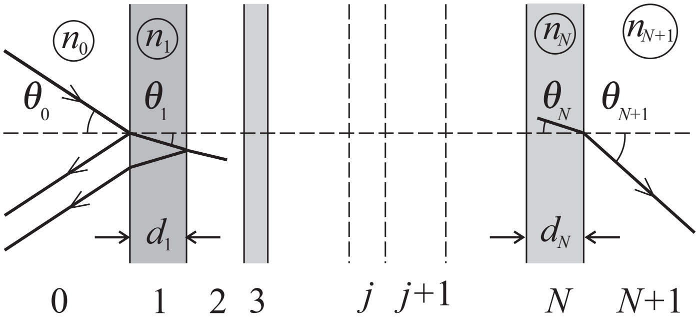

Figure 1.3. Diagram of light propagation in a multilayer consisting of stack of N thin films with thickness d_j and refractive index n_j (j =1, 2,…N) facing media with refractive indices n_0 and n_{N+1}. Incident light entering with an angle θ_0 to the normal, propagates in layer j with an angle θ_j, following from Snell's law.

Reflectance and transmittance of a multilayer

A stack of thin films also creates structural colouration. Let us consider a multilayer consisting of N infinite wide, thin layers of homogeneous dielectric media, separated by parallel surfaces, faced by media with (real) refractive indices n_0 and n_{N+1} (Figure 1.3). The thicknesses of the layers are d_j and the refractive indices are in general complex: $n_j = n_{j\mathrm{R}} - in_{j\mathrm{I}}$ (j = 1, 2,…, N).

The imaginary part of the refractive index is related to the absorption coefficient of the medium, κ_j, by $\kappa_j = 2kn_{jI}$, where $k = 2\pi/\lambda$ is the wave number in vacuum (λ is the light wavelength). The propagation of light through the multilayer is governed by Snell's law:

$$n_j \sin\theta_j = n_0 \sin\theta_0\,, \qquad j = 1, 2, \ldots, N+1 \qquad (1.4)$$

where the angle of incidence θ_j of the light ray at the interface of media j and j + 1 can be complex. The light propagation through the multilayer is described by the transfer matrix (Yeh, 2005)

$$M = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} = D_0^{-1}\left[\prod_{j=1}^{N} Q_j\right] D_{N+1} \qquad (1.5)$$

where

$$D_j = \begin{pmatrix} p_j & p_j \\ q_j & -q_j \end{pmatrix}, \qquad j = 0, 1, 2, \ldots, N+1 \tag{1.6}$$

and

$$Q_j = \begin{pmatrix} a_j & ib_j p_j / q_j \\ ib_j q_j / p_j & a_j \end{pmatrix}, \qquad j = 1, 2, \ldots, N \tag{1.7}$$

with $a_j = \cos\varphi_j$ and $b_j = \sin\varphi_j$, where $\varphi_j = kn_j d_j \cos\theta_j$. For TE-waves p_j and $q_j = n_j \cos\theta_j$, while for TM-waves $p_j = \cos\theta_j$ and $q_j = n_j$. The reflectance and transmittance of the multilayer then are

$$R = \left| \frac{M_{21}}{M_{11}} \right|^2 \quad \text{and} \quad T = \frac{n_{N+1}}{n_0} \frac{\cos\theta_{N+1}}{\cos\theta_0} \left| \frac{1}{M_{11}} \right|^2 \tag{1.8}$$

When the multilayer consists of alternating layers of two material layers with thickness d_a and d_b and refractive index n_a and n_b, the condition for constructive interference becomes

$$2\,(n_a d_a \cos\theta_a + n_b d_b \cos\theta_b) = m\lambda \tag{1.9}$$

where θ_a and θ_b are the angles of refraction in the two media. At normal incidence these angles are $\theta_a = \theta_b = 0^\circ$, or, the principal peak wavelength then is $\lambda = 2(n_a d_a + n_b d_b)$. In a so-called "ideal" multilayer, the optical path lengths of the alternating layers are equal, $n_a d_a = n_b d_b = nd$, giving $\lambda_{\text{peak}} = 4nd$. This relationship allows a quick assessment of the dimensions of the multilayer structure when the reflectance spectrum is known.

This thesis

In this thesis I report on the study of the structural colours in bird feathers. By making use of new methods of measuring and calculating the optical properties of these samples, I have built on pioneering work performed decades ago (Durrer, 1962; Rutschke, 1966; Durrer and Villiger, 1967; Durrer and Villiger, 1970; Durrer, 1977) with the aim to broaden our insights into the physics that governs the appearance of these natural photonic objects.

Macro- and micro-spectrophotometry methods were used to measure the colouration quantitatively by means of reflectance and transmittance spectra. Macroscopic angle-resolved spectrophotometry yielded the reflectance for varying degrees of incidence, while imaging scatterometry allowed to get an image of the spatial distribution of scattering light by the sample. By approximating the various structures with multilayer arrangements and calculating the reflectance and transmittance with a very accessible modelling method, it was possible to describe the measured spectra appropriately. These methods are all described in detail in the respective chapters.

In Chapter 2, I describe the study of biophotonic structures in the feather barbules of the European starling and the Cape starling by treating them as a simple multilayer. Both birds have only a single layer of melanosomes, which are respectively solid and hollow, corresponding to two material interfaces for the European starling and five interfaces for the Cape starling, in both cases without a regular periodicity. Besides providing a more detailed understanding of the melanosome shapes of the Cape starling, my results show that the colour of both birds is coupled to the keratin cortex thickness, which is defined by the spacing of the melanosome layer to the barbule surface.

Chapter 3 is dedicated to the investigation of a very prominent biophotonic structure, namely that of the peacock's blue coloured neck feathers, which have a photonic structure with about 10 melanosome layers, spaced by layers of keratin interlaced with air channels. I found that this truly unique biophotonic structure, which has a very long-range order as well as a two-dimensional arrangement of melanosomes, can be exceptionally well approximated by multilayer modelling. This result is supported by the comparison to a full-scale electromagnetic field calculation using FDTD modelling of the peacock's blue photonic structure, for varying angles of incidence and reflection.

Chapter 4 reports how the approach detailed in chapter 3 can be successfully applied also for the various structural colours of the six different colour regions of the peacock tail feather. Here, the role of short- versus long-range order was clarified for the case of a dielectric multilayer, since the various colour regions comprise between 3 and 12 melanosome layers (7-25 material interfaces). I also describe in detail the crucial role that the uppermost layers of the photonic structure, and especially the cortex layer, play in determining the shape of the spectrum. These results demonstrate that tuning of the structural colours can be based on a very simple mechanism, *viz*. the thickness of the cortex.

Chapter 5 details a study of the changes that the structural colours of bird feathers undergo when subjected to water. I included in this survey both short- and long-ranged ordered structures of both solid and hollow melanosomes that can be found in the European starling, Cape starling, mallard and magpie feathers. Wetting of the feathers causes a bathochromic (toward longer wavelengths) shift of the reflectance spectra, presumably due to expansion of the keratin.

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