Anisotropic Light Transport in White Beetle Scales

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Rather than exploiting selective absorption by pigments, certain insects achieve coloration using complex photonic structures.1–4 For instance, bright and iridescent colors are achieved by butterflies and beetles exploiting optical coherent effects in ultrathin periodic layers of low-refractive-index material.3,5,6 A bright white diffuse appearance is instead more complicated to achieve, since all colors have to be reflected with the same high efficiency. Since broad-band high reflectance cannot be obtained with optical coherent effects (which would lead to wavelength dependencies and iridescence), bright whiteness is usually achieved with thick disordered systems which multiply scatter light. The lower the refractive index $n$ of the scattering elements, the thicker the system has to be to achieve a pronounced brightness. The Cyphochilus beetle is an exception to this, since it obtains a remarkably brilliant white color with extremely thin scales, whose interior is made of a nanostructured random network of low-refractive-index chitin ($n = 1.56$).7,8 In a recent study, we have shown that the multiple scattering of light occurring inside the scales is responsible of the bright broad-band reflectance of this insect and that this scale is one of most strongly scattering low-refractive-index materials known.9 We argued that their internal structure may have evolved, over millions of years,10 toward optimal light scattering employing as little material as possible. We hypothesized that the apparent anisotropy of the chitin network could represent the crucial aspect of such scattering optimization.

In this work, we unequivocally demonstrate that the scales covering the Cyphochilus’ body exhibit anisotropic light transport, which in turn comes as a direct consequence of the anisotropic arrangement and shape of the scatterers. Light transport in the scales thus has evolved to increment the scattering strength along the out-of-plane direction, leading to a pronounced total reflectance, i.e., its brightness. This led, as a trade-off, to a decrease of the in-plane scattering strength, which in any case does not contribute to the brightness. In this respect, our results identify the degree of anisotropy as a key microscopic structural feature in determining a bright white reflectance for thin, low-refractive-index optical coatings. Such discovery can be successfully applied in developing new lightweight, thin optical materials with immediate technological impact in applications ranging from coatings to displays and light-emitting diodes (LEDs)/illumination.

The structure responsible of the extraordinary bright whiteness of the Cyphochilus beetle is shown in Figure 1. The body of the beetle is covered by white scales (Figure 1b), which are about 100 µm wide and 250 µm long. Analysis on scanning electron microscope (SEM) images of 80 different scales of the same beetle, gently dissected with a razor blade, reveals an average thickness of 7 µm with a standard deviation of $\sigma = 1.5 \mu m$ (Figure 2a). The bright whiteness of the Cyphochilus beetle scales angle results from multiple scattering of light by the microstructure present inside the scales (Figure 1d), which is characterized by a cuticular random network of interconnected chitin rods, typically shorter than 1 µm and with a diameter of approximately 250 nm.7 SEM images and analysis reported in literature reveal a very high filling fraction of the chitin network with respect to the volume of the scale ($f \approx 60\%$).7,9

As discussed in our previous work,9 the particular morphology of the internal structure allows a high density of scatterers (which intuitively increases the scattering intensity and, consequently, the brightness) yet reducing the optical crowding effect (which instead tends to lower the overall scattering strength).10 A high filling fraction usually leads to pronounced structural correlations due to the physical size of the scattering elements11,12 which in turn leads to a modulated spectral response, i.e., a coloration.13–16 This is the case, for example, of the blue and green appearance of certain species of birds,17–19 which is determined by the strong degree of spatial correlations in the structural disorder of their feathers. In contrast, the disorder in Cyphochilus scales has evolved in order to avoid spatial correlations, and yet maintain a high filling fraction, by exploiting the anisotropic shape of the scatterers. This comes to a cost: high packing fractions of rods can be reached only introducing a certain degree of angular correlations.9,20 Indeed, from analysis of electron images of the interior of the white scales we can observe that the chitin rods appears to be mainly aligned with a planar orientation. As a consequence of such structural anisotropy, also light transport is expected to be anisotropic,
transmitted through the scale, which is strongly affected by the anisotropy of the system.\textsuperscript{[24–28]} We demonstrated that the results of these two independent experiments are in contradiction when interpreted with isotropic diffusion theory. In contrast anisotropic theory, which accounts for different diffusion coefficients along different axes (see the Supporting Information), can perfectly reproduce the experimental data.\textsuperscript{[26,27]} Since all the scales exhibit similar transport properties, we report here only the results relative to a single reference scale.

In case of negligible absorption, the measurement of the total transmission \( T_{\text{tot}} \) through a diffusive slab is an indirect estimation of the total reflectance \( R_{\text{tot}} \), namely, of the brightness of the system. Isotropic diffusion theory relates the measured total transmission to the thickness \( t \) of the slab and the transport mean free path \( l_t \) through the well-known Ohm’s law for light.\textsuperscript{[9]} By neglecting absorption and ballistic light,\textsuperscript{[9]} it can be written as:\textsuperscript{[29]}

\[
T_{\text{tot}} = \frac{l_t + z_e}{t + 2z_e} = \frac{1 + \frac{2}{3}A}{\text{OT} + \frac{4}{3}A}
\]  

(1)

where \( z_e = 2l_tA/3 \) is the so-called extrapolation length\textsuperscript{[30]} and \( A \) is a coefficient that takes into account the internal reflection due to the refractive index mismatch with the environment.\textsuperscript{[9,31]} The OT is the optical thickness of the system and it is defined by the relation \( \text{OT} = t/l_t \). We calculated \( A \) as done by Burresi et al.,\textsuperscript{[9]} by employing the phenomenological formula proposed by Contini et al.,\textsuperscript{[12]} and applying the Maxwell Garnett mixing rule with a filling fraction of 0.61\textsuperscript{[9]} to calculate the average refractive index of the scale. To measure the total transmission we focused a laser beam (wavelength 550 nm, focal spot diameter of 1.5 µm) on the central part of the scale, orthogonal to its surface, and we used an integrating sphere to collect all the light emerging from the scale (see figure in the Supporting Information). We measured \( T_{\text{tot}} = 0.29 \pm 0.2 \). This value is an average of several measurements made in a relatively small area in the central part of the scale, where it can be considered flat with a good approximation. For this reason, in this experiment, we neglected the actual smooth curvature of the scale toward its edges and we approximated it to a slab with an effective thickness.

We revealed transport anisotropy combining the results of two independent steady state experiments performed on different scales. First, we measured the total transmission through a scale, which is an indirect measurement of the brightness in absence of absorption (this is the case of the beetle scales, as reported by Burresi et al.,\textsuperscript{[9]} Supporting Information). Second, we performed steady state imaging of the diffuse light as observed in many structurally anisotropic materials such as biological tissues,\textsuperscript{[21,22]} nematic liquid crystals,\textsuperscript{[23,34]} or plastic porous fiber samples.\textsuperscript{[25,26]} Therefore, a proper light transport investigation of the scales represents a powerful tool for a noninvasive study of their morphological anisotropy and from this shed light on the mechanism that leads to its bright white reflectance.

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transport properties of the beetle’s scales. Keeping the same experimental conditions as in the total transmission measurement, we focused a laser beam on the front side of the scale while imaging its back side on a CCD camera. Figure 2b shows the geometrical sketch of the experiment, with the image plane parallel to the $xy$ plane and the beam direction parallel to $z$ axis (see the Supporting Information for further details on the setup). The beam impinged in the central part of the scale, and we investigated a region of 40 µm radius centered in the beam focus, where we can consider the scale as a slab with an effective thickness. The resulting image (Figure 2b, inset) is an average of 20 different acquisitions taken moving the beam within a small central area of the scale. This procedure, together with a relatively broadband source ($\Delta \lambda = 10$ nm), makes the investigation statistically accurate, averaging over several realizations of disorder. We compared a crosscut of the imaged profile together with the prediction obtained from the isotropic diffusion equation\[^{[31]}\] for the optical thickness measured with the total transmission experiment, that is, $OT = 5.8$. Thereafter, we also considered various physical thicknesses in a range of $2\sigma$ with respect to the center of the distribution (Figure 2a). This analysis, reported in Figure 2c, clearly shows the discrepancy between the experimental data and the prediction according to isotropic diffusion theory.

The appropriateness of isotropic diffusion to describe light transport in such a thin system must be put to question, since its accuracy rapidly decreases for $OT < 10$.\[^{[33]}\] Therefore we performed isotropic Monte Carlo simulations for a disordered slab with $OT = 5.8$ and the same effective refractive index of the scale. Figure 2d shows the simulated isotropic profile with an isotropic diffusion theory fit of the profile tail. Despite the low optical thickness the transport parameters are retrieved within an accuracy better than 1%, as long as the central points of the profile are excluded. This is necessary since ballistic and low order scattered light, which have a dominant contribution to the peak of the transmission profile in thin slabs, are not taken into account in the diffusion approximation.\[^{[34]}\] We conclude that the large discrepancy shown in Figure 2c is not due to a possible breakdown of diffusion theory, but must be caused by the very nature of light transport in the scale (i.e., transport is not isotropic).

Encouraged by the anisotropy noticed in the electron images, we analyzed the results of the imaging and total transmission experiments in terms of anisotropic diffusion theory.\[^{[26–28,35,36]}\] Anisotropic diffusion equation indeed links the transmitted light through a diffusive slab with its thickness and with the diffusion coefficients, and thus the transport mean free paths, along the different axes (see the Supporting Information).\[^{[27]}\] It is fundamental to note that, in the case of anisotropic transport, the Ohm’s law is still valid, provided that $l_t$ in Equation (1) is replaced with the transport mean free path along the direction orthogonal to the scale surface ($l_{tz}$ with the notations of Figure 2b). Hence, to increase the brightness is only necessary to decrease the mean free path along the z-direction, regardless of the in-plane properties.

Even in the anisotropic diffusion approximation, we can still define the optical thickness as $OT = l_t/l_{t\parallel}$ and determine it experimentally. As before, we considered a set of $l_{t\parallel} = l_t/OT$ values,
The results of the fits for different thicknesses are plotted in Figure 2 e,f for crosscuts along the long axis of the scale. This implies an in-plane anisotropy which seems to be plausible since the “growth” direction of the scales is parallel along the long axis of the scale. However, we cannot make any definitive statement on the subject since the scales have a more pronounced curvature along the y-direction and this might create some reshaping in the transmitted profile. In contrast, the z-direction anisotropy of light transport is clearly evident, demonstrating that the anisotropy of the intrascale structure is optimized to increase the scattering along the direction normal to the scale surface. This come to the cost of a longer in plane transport mean free path, which however does not contribute to the brightness.

In conclusion, we unequivocally demonstrated that the scales of the Cyphochilus beetles indeed developed a dense, anisotropic network compressed along the direction orthogonal to the scale surface. This arrangement is optimized to selectively enhance the scattering strength in the normal direction, increasing the total reflectance, at expense of the scattering strength in the plane, which anyway is not relevant for the brightness.

Notably, optimizing angular correlations in network-like optical materials seems still a largely unexplored strategy in many applications. Considering, for example, the recently growing field of cellulose photonics, it is well known that for highly packed systems, moving from micrometric fibers (like common paper) to sub-µm fibrils leads to increased transparency.

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Experimental Section

Total Transmission: The excitation was provided by a Fianium 1060 supercontinuum source. The beam was focused on the sample with a high NA aspheric lens (spot diameter in the focus ≈1.5 µm). The beam has been filtered with a bandpass filter with central wavelength of 550 nm and bandwidth of 10 nm. The scale was held (by electrostatic forces) on a ND 3 filter, to minimize the background light due to doubly reflected light, at the entrance port of an integrating sphere. The transmission at the output of the sphere was measured using a silicon photodiode and a lock-in amplifier.

Imaging of Transmission Profile: The excitation conditions were the same of the total transmission experiment. The beam was circularly polarized (we used a quarter-wave plate) to avoid polarization artifacts due to single scattering by anisotropic scatterers. The imaging part of the setup was composed by a 20× Olympus objective, a tube lens and an Andor Apogee cooled CCD camera. The scale was held (by electrostatic forces) on a ND 3 filter. The resulting image of the profile is an average of 20 different acquisitions.
Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
The authors wish to thank P. Vukusic for having provided the beetle’s scales used in the experiments and U. Steiner for fruitful discussions. The research leading to these results has received from the European Research Council under the European Union’s Seventh Framework Programme (FP7/2007–2013)/ERC Grant Agreement No. [291349] and the BBSRC David Phillips fellowship [BB/K014617/1].

The licence on this manuscript was changed after publication, on October 20, 2015.

Received: March 30, 2015
Revised: May 16, 2015
Published online: June 24, 2015

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