Brilliant Whiteness in Ultrathin Beetle Scales
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Brilliant Whiteness in Ultrathin Beetle Scales

Pete Vukusic,[1]* Benny Hallam,[2] Joe Noyes[1]

The colored appearances of animals are invariably controlled by pigmentation, highly periodic ultrastructure, or a combination of both (*1*, *2*). Whiteness, however, is less common and is generated by neither of these methods, because it requires scattering processes appropriate for all visible wavelengths. We report the identification of whiteness resulting from a three-dimensional (3D) photonic solid in the scales of *Cyphochilus* spp. beetles. Their scales are characterized by their exceptional whiteness, their perceived brightness, and their optical brilliance, but they are only 5 μm thick. This thickness is at least two orders of magnitude thinner than common synthetic systems designed for equivalent-quality whiteness.

Archetypal brilliant whiteness that is not augmented by fluorescence, such as whiteness from snow or milk, is the result of multiwavelength scattering arising from aperiodic and multiply oriented interfaces between low-absorbance media of appropriately different refractive index (*3*). The whiteness of *Cyphochilus* spp. originates from elongated flat white scales that imbricate its body, head, and legs (Fig. 1A). These scales are about 5 μm thick, 250 μm long, and 100 μm wide. Their interiors are composed of a random network of interconnecting cuticular filaments with diameters of about 250 nm (Fig. 1B and fig. S1).

Two-dimensional fast Fourier transforms (FFTs) of electron microscope images of the scales' interior (Fig. 1C) confirmed an absence of well-defined periodicity. Wave vector space maps produced by this transform [Supporting Online Material (SOM) text] were free from any single spatial component of refractive index variation (fig. S2A). Experimentally this was confirmed by recording the diffraction pattern associated with light incident on individual scales. By mounting specific white scales on separate needle tips and directing low-intensity focused laser light exclusively through the center of each scale, we imaged the reflection and transmission diffraction patterns on spherical screens (fig. S2B). The resulting diffraction patterns closely matched the FFT maps of the scales' interior (fig. S2C) and confirmed the cuticular filament network as the origin of the whiteness. The intrascale cuticle volume occupancy is about 70%. This appears to optimize scattering intensity by maximizing the scattering center number density while avoiding substantial unfavorable optical crowding (*4*). Optical crowding occurs when the radiation fields associated with individual scattering centers overlap. It causes the system to adopt characteristics of fewer and larger scattering centers when the distance between individual scatterers becomes too small. The relatively high void fraction in this *Cyphochilus* beetle's scales appears to be a vital part of the system's ability to scatter light. It is this, as well as the system's aperiodicity and index contrast of about 0.56, that create such intense optical whiteness for very limited thickness.

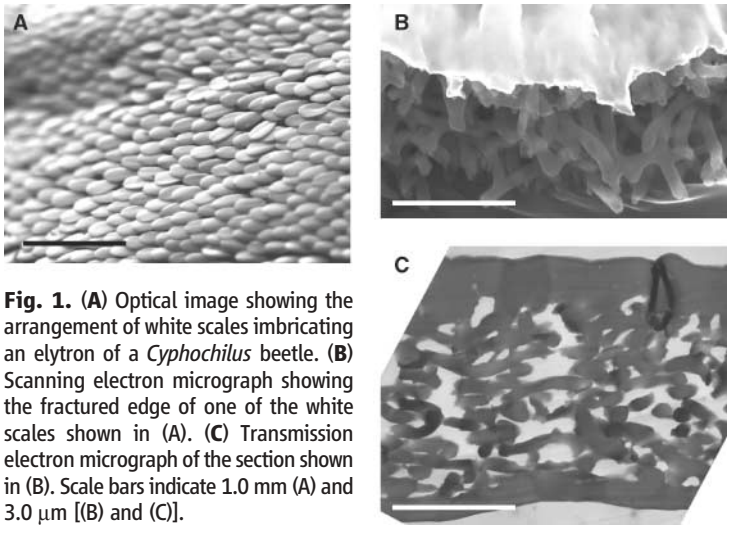

Fig. 1. (**A**) Optical image showing the arrangement of white scales imbricating an elytron of a *Cyphochilus* beetle. (**B**) Scanning electron micrograph showing the fractured edge of one of the white scales shown in (A). (**C**) Transmission electron micrograph of the section shown in (B). Scale bars indicate 1.0 mm (A) and 3.0 μm [(B) and (C)].

The quality of the beetle's whiteness and brightness was quantified according to International Organization for Standardization national standards (SOM text). Its whiteness and brightness values (*5*) were measured to be 60 and 65, respectively, quantitatively indicating remarkable multiwavelength scatter for systems that are only 5 μm thick. In synthetic systems where whiteness is desirable, far more substantial structure is necessary. For example, conventional white uncoated wood-free papers (comprising random networks of bleached cellulose fibers) can be upward of 25 times thicker than these beetle scales but return only an 8% superior brightness. Carbonate or kaolin crystal inclusions and optical brightening agents (blue fluorescing dyes) are added to paper coating formulations to enhance scattering contrast and to improve the perceived appearance of white. However, individual isolated 5-μm-thick calcium carbonate coating layers have a brightness of only 40 to 50, with such poor opacity that its whiteness value is meaningless. Similarly, the whiteness of human teeth is dominated by multiwavelength scattering from packed hydroxyapatite crystals in up to ~2 mm of tooth enamel. Although they are generally considered to be white, their best natural whiteness and brightness are relatively low; typical human milk teeth exhibit a whiteness under 40 and a brightness of about 53, reflecting relatively low index contrast and high absorption at blue wavelengths.

For proportionally insignificant supplementary thickness, the addition of these scales' form of photonic solid would strongly enhance the desirable quality of whiteness in these and many other systems. Additionally, it offers a permeable, flexible, and fault-tolerant ultrathin layer with which to back large-area white light–emitting devices (OLEDs) and control their emission direction.

The phenotypic color of this *Cyphochilus*, credited for cryptism among white fungus, arises from an aperiodic form of structure that might appear to contrast strongly with the highly periodic structure in narrow-band colored weevil scales (*6*). However, they do share several important features: a cuticular filament network with typical filament diameter of the order from 200 to 250 nm, a scale thickness of about 5 μm, and similar extrusion from single epidermal cells into the sealed parcels that comprise each scale. The form of the photonic solid in these white beetle scales confirms that the transition from high-contrast saturated color (SOM text) to optically brilliant whiteness is largely a matter of structural order.

References

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Supporting Online Material

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SOM Text
Figs. S1 to S4

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This PDF file includes:

SOM Text
Figs. S1 to S4

Supporting online material

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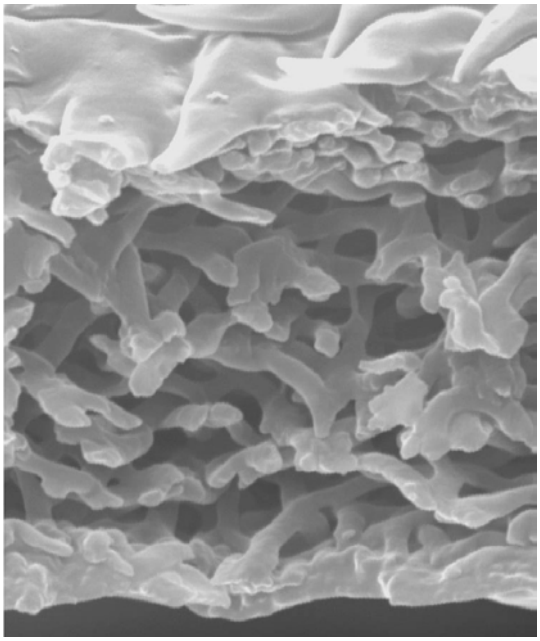

Fig. S1: SEM image (to augment figure 1B) of a fractured edge of a single white *Cyphochilus* scale.

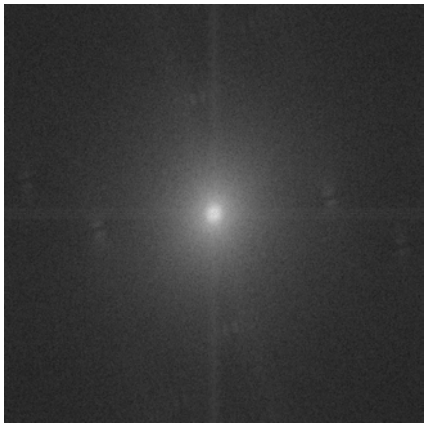

Figure S2A: 2D fast Fourier transform of the filament network within the white scale cross-section shown in figure 1B.

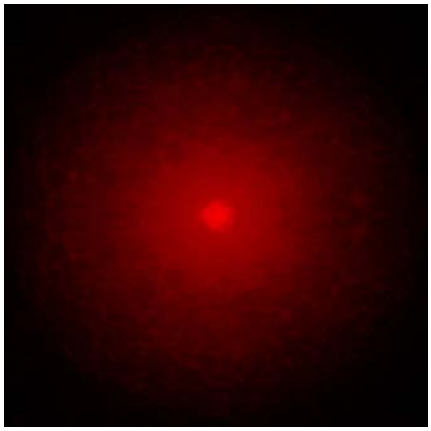

Figure S2B: the experimental diffraction pattern obtained from a laser incident on the single white scale shown in figures 1B and 1C.

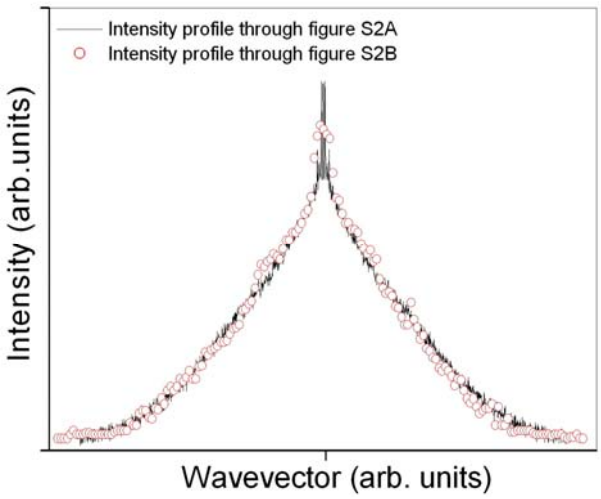

Figure 2C: an overlay of 1D intensity profile data from figure S2A and S2B (the match is excellent despite the experimentally incident beam spot's finite size and the limited structure sample area in figure 1B).

Figure S3: an optical image of a *Cyphochilus* beetle.

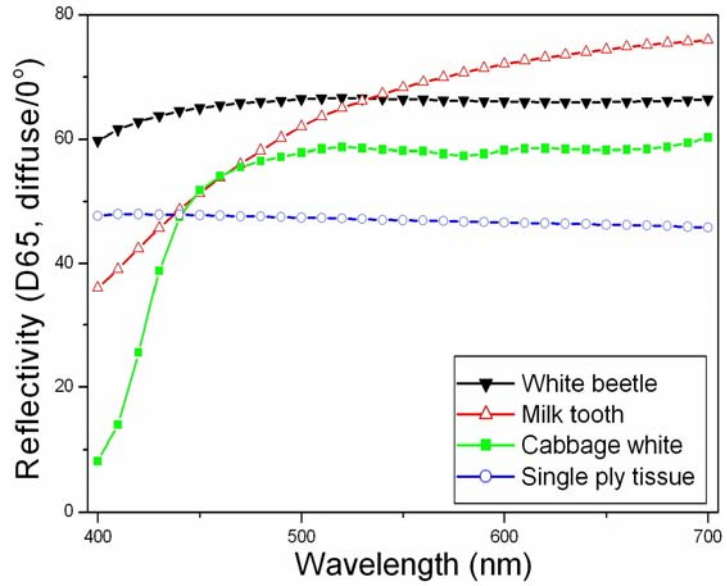

Figure S4: comparison of optical reflectivity data, collected using a Datacolor Elrpho photospectrometer, taken from; a *Cyphochilus* beetle elytron; a human milk tooth; a cabbage white butterfly and a white single-ply tissue.

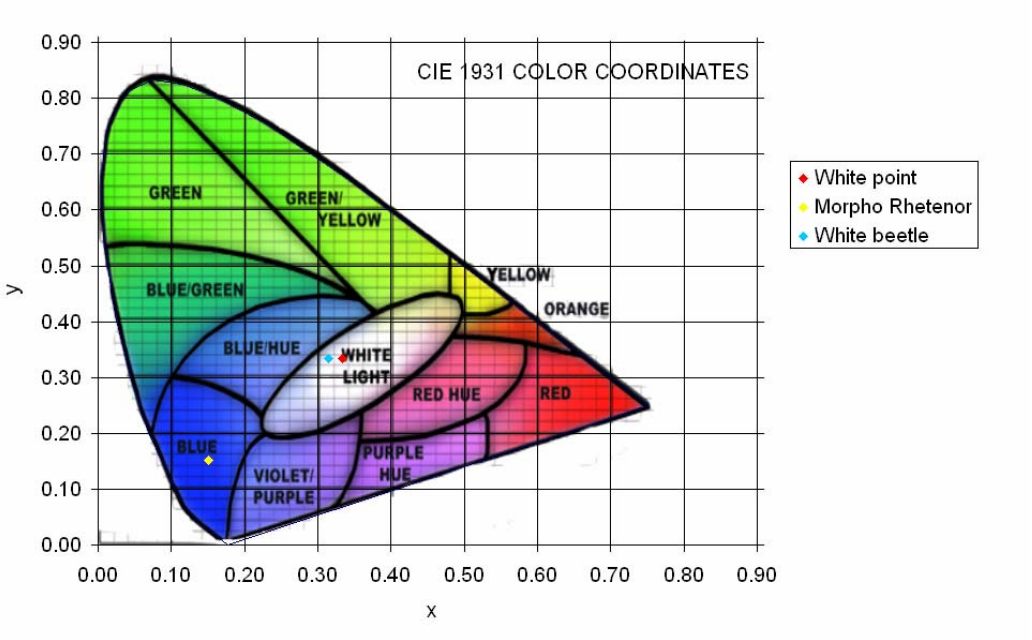

Figure S5: Chromaticity triangle depicting the coordinates of the white *Cyphochilus* beetle and a *Morpho rhetenor* butterfly. The graph shows that the beetle is more than 12 times less saturated than this butterfly.

Supporting References.

S1. Fourier transforms find many applications in the physical sciences. One well-known example is in X-ray crystallography where they are used to interpret or predict diffraction patterns from atomic crystals. In optics too, an understanding of Fourier transformation and of Fourier pairs significantly helps with an understanding of the phenomenon of diffraction.

One such Fourier pair is formed by a diffraction pattern and the physical object that gives rise to it. In this case, the pair is formed specifically between *position-space* (that is, the spatial position of the scattering centres that make up the physical object) and *wavevector-space* (which represents the pattern of light that is diffracted from the scattering centres). With a few added phase considerations, one is the direct Fourier transform of the other.

The Fourier-transform methodology in this article relates to the following. If the 3D structure shown in the beetle scale images in Fig. 1 is the basis of this brilliant whiteness through multi-wavelength scattering, then the optical diffraction pattern experimentally obtained from the beetle scale should be identical to the 2D Fourier transform of the cross-sectional image of this structure. This is because the diffraction pattern should form a Fourier pair with the structure. More explicitly, the diffraction pattern should be the *wavevector-space* representation (or equivalently the *reciprocal-space* or *momentum-space* representation) of the physical spatial distribution of those scattering centres in real-space.

A 2D Fourier transform of the TEM image of the beetle structure was taken to produce a wavevector-space representation of the scattering centres in the beetle. This Fourier transform map (Fig. S2A) is nearly identical to the experimental optical diffraction pattern obtained when a laser spot was focused onto the centre of an individual scale (Fig. S2B). Despite the finite size of this laser spot and the limited TEM image area, the intensity profiles of both images (Fig. S2C) matched closely, providing a strong indication that it is the structure of the imaged cross-section that gives rise to the experimentally-observed scatter. Furthermore, both images were completely absent of any periodic structure (the horizontal and vertical lines are a feature of the finite TEM image size and not of the structure itself), a fact which directly infers the absence of real-space periodicity in the beetle scale structure. This evidence confirmed the scale structure as the source of the bright whiteness.

S2. The quality of the beetle's whiteness was quantified according to ISO 11475, and its brightness to ISO 2470 using a D65 illuminant in a photospectrometer (Datacolor Elrepho) calibrated with an ISO/IR3 standard, traceable to National Standards.

S3. Colour saturation (or excitation purity) is a measure of how close to monochromatic a sample colour is. It may be quantified by calculating the displacement of the sample colour from the neutral point as a percentage of the distance to the locus of the CIE chromaticity triangle that represents the dominant wavelength. A truly monochromatic colour is defined as 100% saturated, while physical white is completely unsaturated with an excitation purity of 0%.

In animate natural systems, one of the most widely quoted examples of significantly saturated colour is that of the iridescent blue wing colour of some species of *Morpho* butterfly. One species in particular, *Morpho rhetenor*, stands out from the other species because of its ultra-high brightness and the significant spectral purity of its blue wing colour (for a description of the *M. rhetenor* structurally coloured system see: Vukusic et al**.** *Proc. Roy. Soc. B*., 266, 1403-1411, 1999). Although its wing colour is not truly monochromatic (i.e. it does not have an excitation purity of 100%), it is among one of the single most saturated structural colours that is exhibited by an animate system. To quantitatively contrast the excitation purity of the *Cyphochilus* beetle's unsaturated whiteness to the high levels of saturation attributed by most researchers to the blue colour of *M. rhetenor*, we measured their dominant wavelengths and excitation purities using ISO National Standard equipment.

Figure S5 shows the chromaticity co-ordinates of both species relative to the white point (physical white). The typical whiteness of a *Cyphochilus* beetle is defined by a dominant wavelength of 492 nm, and a colour saturation of 6.2%. A dominant wavelength of 477 nm and an excitation purity of 79.6% define typical *M. rhetenor* wing colour.