Weak Localization Enhanced Ultrathin Scattering Media

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The brilliant white appearance of ultrathin scattering media with low refractive index contrast and the underlying radiative transport phenomena fascinate scientists for more than a decade. Examples of such systems are the scales of beetles of the genus Cyphochilus, photonic network structures or disordered Bragg stacks (DBS). While previous studies relate the highly efficient scattering in the scales to the anisotropy of the intra-scale network and diffusive light transport, the coherent radiation propagation dynamics remains unaccounted for. Here, we identify the coherent scattering mechanisms for the white scales, which are known to occur for example when weak localization gives rise to coherent back scattering (CBS) or random lasing in disordered active media. For efficient intra-medium scattering, coherent back scattering occurs, when two counter-propagating scattering light paths in the medium, i.e., the illuminating light and collinear back scattered light interfere constructively and give rise to the robust and well known CBS.

Thereby, this framework is often favored over solving Maxwell’s equations, since diffusion theory delivers macroscopic transport properties applicable over the entire structure at much lower cost, while the solution of Maxwell’s equations differs with the specific microscopic realization of the structure. However, diffusive radiation transport neglects the coherent propagation of scattered fields and hence does not account for interference phenomena in disordered media, which are known to occur for example when weak localization gives rise to coherent back scattering (CBS) or random lasing in disordered active media. For efficient intra-medium scattering, coherent back scattering occurs, when two counter-propagating scattering light paths in the medium, i.e., the illuminating light and collinear back scattered light interfere constructively and give rise to the robust and well known CBS.

Indeed, the observation of CBS and modeling of the resulting cone using Monte Carlo simulations is also reported for Cyphochilus scales. However, beyond CBS, the modeling of the brilliant white appearance of Cyphochilus scales neglects any other coherent effects. This can hamper tailoring disordered photonic media since an unambiguously identified scattering mechanism is the basis for nanostructure design for optimized performance. Using ultrafast time-resolved spectromicroscopy of scattered light outside the CBS cone, we here identify the coherent light scattering mechanisms for Cyphochilus scales and disordered Bragg stacks (DBS) and show that weak localization in leaky photonic modes significantly contributes to the brilliant whiteness of these scatterers.

1. Introduction

The animal and plant kingdom is abound with countless colorations ensuring the survival by accomplishing different tasks such as camouflage, signaling, warning, and so forth. These various colorations are mainly achieved by three different mechanisms namely pigmentation, structural colors, and bioluminescence. Especially, structural colors are under frequent investigation since they serve as a great inspiration for engineering of artificial coloration with numerous applications.

While the physics behind some chromatic structural colors is known for more than 300 years, the generation of the brilliant whiteness of ultrathin structures such as the scales of the beetle Cyphochilus, was addressed merely a decade ago. Since being first reported much effort was made to unveil the connection between the structural and the optical properties of the disordered intrascale network focusing on the anisotropy and its connection to diffusive light transport. Furthermore, the fabrication of artificial structures capable to mimic the whiteness was shown using different techniques. However, also in these artificial structures the light transport is treated as a diffusion process only approximating the real scattering behavior.

In strongly scattering media the description of light propagation as ballistic transport breaks down and is commonly replaced by diffusive radiation transport that explains well the observed optical characteristics in many applications.

2. Results and Discussion

To systematically study the impact of coherent transport on the whiteness of the Cyphochilus’ scales, we use the setup shown in
Figure 1. Microscopic and ultrafast time-resolved spectroscopy of light scattered from *Cyphochilus* scales and microfabricated DBS structures. a) Scheme of the spectral interference setup (see explanations in Experimental Section). b) Photograph of *Cyphochilus* (left) and disordered Bragg stacks (DBS, center closeup) with light microscope images of a single beetle scale (right top) and DBS (bottom) as insets. c,d) Spatially resolved time domain amplitude of light scattered from a *Cyphochilus* scale and DBS. The transition threshold between diffusive regime and resonance radiation as identified in (h) are indicated (vertical translucent bar). e,f) Scheme illustrating how incoming light is scattered in the initial diffusion-like regime and later via weakly localized photonic modes indicated by closed pathways. The gray structure is a cross section of a *Cyphochilus* scale (taken from Wilts et al.[11]). The black overlay on the left side shows the disordered Bragg stacks. g) Scattered electric field at a single scan position (white dashed line in (c)) with indication of the short time Fourier transform windows used in (i, red) and (j, blue). h) Wigner distribution function of the scattered field shown in (g). At $105 \pm 10$ fs (black line) the dominating light transport regime changes from diffusion-like to weak localization assisted. i-k) Fourier spectra of the early time window (i) (~30 to 90 fs, red in (g) and (h)), the later time window (j) (285 to 405 fs window, blue in (g) and (h)) and the total measured time window (k).

Figure 1a to perform ultrafast time-resolved light scattering spectromicroscopy on a single scale.[26,27] The observations are confirmed for disordered Bragg stacks (DBS) fabricated via direct laser writing (see Supporting Information) shown in Figure 1b. The DBS mimic the beetle scales, reproduce their known optical properties[28] and allow for realistic scattering light simulations based on finite-difference time-domain (FDTD) Maxwell solvers and Monte Carlo (MC) diffusive light transport simulations.

The identification of coherent scattering is significantly facilitated if the number of interfering pathways is kept small. For example, laser speckles are the most pronounced when only a small area of the scatterer is illuminated. However, if the detector integrates over sufficiently many different interfering pathways, the speckles disappear. In this case with exception of the coherent back scattering peak, the scattering behavior is often well explained by diffusive radiation transport theory,
although the underlying transport is coherent. To reduce the number of interfering pathways, the present investigation relies, both, on the focused illumination and collection of scattered light from a small sample volume. Furthermore, coherent propagation adds a well-defined phase to the scattered fields and thus reconstruction of the temporal evolution of the scattered electric field provides additional information on the scattering mechanism.

To achieve the spatial resolution necessary to observe only few interfering scattering pathways, a parabolic mirror (Figure 1a; M) focuses a pulsed Ti:sapphire laser beam down to a \( \leq 3 \) \( \mu \)m spot on the surface of the sample (Sa) and collects the scattered light under an angle of \( \approx 24^\circ \) relative to the specular direction. To filter for intra-scale scattering, i.e., multiple scattered light components, a cross-polarization configuration is used. The illuminated position is scanned by moving the sample using a piezo stage. Spectral interference between the scattered light pulse (SP) and a reference pulse (RP) allows for the time reconstruction of the field of the scattered light (see Experimental Section). The amplitude of the measured electric field (cf. Figure 1c,d) shows for both samples essentially the same dynamics, i.e., spatially varying exponential decay modulated by distinct beating, indicating interference taking place. As discussed below, two different propagation regimes can be identified in the scattered light signals. Initially, diffusion-like transport (Figure 1e) dominates, whereas for longer times radiation leaking from weakly localized photonic modes formed by randomly closed scattering pathways (Figure 1f) prevails, which gives rise to the observed beating behavior.

To identify the different propagation regimes, we analyze the coherent scattering signal (cf. Figure 1g) in time and frequency domain by means of the Wigner distribution function (WDF, see Experimental Section) exemplarily shown in Figure 1h for the *Cyphochilus* scale. For early times broadband features are present, which reproduce the excitation spectrum when evaluating the short time Fourier transform (cf. Figure 1i). At about \( 105 \pm 10 \) fs, there is a qualitative change in the spectral content of the WDF, i.e., broad spectral features are replaced by fine modulations. This time matches closely to the pulse round trip time (see Experimental Section), i.e., the time a pulse needs to travel back and forth through the layer assuming a homogeneous, effective medium with an effective refractive index, as it is commonly done in diffusion approximation. The spectral modulations stem from multiple sharp resonances, which become better visible in the short time Fourier transform for later times (cf. Figure 1j). The power spectrum illustrates that the signal now contains spectral peaks independent of the original excitation spectrum, whereas the scattered light in the initial diffusion-like phase exhibits no significant modulation. The spectrum for the full measured signal, shown in Figure 1k, exhibits spectral peaks on top of a broadband background and thus reflects the spectral characteristics of both transport regimes. Further examples of spectral intensities and corresponding phases are provided in the Supplementary Information.

While the short time Fourier transforms (Figure 1i,j) allow identifying the contribution of the different light transport mechanisms over time, this spectral analysis of resonances lacks of resolution due to the short time windows. To unambiguously identify the weak localization assisted scattering, the probability distribution of the resonance lifetimes is investigated applying full time Fourier transformations. Figure 2a reveals that the scattered light spectra possess multiple peaks with varying center frequency and width as function of the spatial coordinate. In the incoherent mean of the spectra over the whole scan (Figure 2b, gray shaded area), these narrow spectral peaks average out and reproduce the excitation spectrum (Figure 2b, dashed line), macroscopically resulting in the white appearance. Based on peak fitting (Figure 2b, red curve), we derive the spectral widths of the peaks, which yields a lower limit for the underlying resonance lifetimes. The distribution of these lifetimes is displayed in Figure 2c and follows a log-normal distribution (red curve), deviating from a normal distribution for longer lifetimes as expected when localization effects occur.[31] The tail toward long lifetimes is associated with the rare occurrence of increasingly localized modes, i.e., cases where scattering pathways close inside the structure instead of coupling to loss channels.[32]

This identification of weak localization assisted light scattering is further supported by FDTD simulations based on the known microstructure of the *Cyphochilus* scale[41] (model data provided by courtesy of B. Wills) and the DBS. As exemplified in Figure S7b,c, (Supporting Information) the local spectra recorded inside the structures also exhibit sharp resonances. Statistical analysis of these resonances yields the lifetime distributions shown in Figure 2d,e, which are in excellent accordance with the experimental results. Hence, we conclude that the spectral resonances experimentally observed in the scattered light indeed originate from weakly localized photonic modes occurring in the same way inside the beetle structure and DBS. The corresponding spectral features give rise to the observed beating behavior in scattered light spectromicroscopy (Figure 1c,d).

To further investigate the light propagation inside the structure the spatio-temporal evolution of the local power (in FDTD simulations) and the photon counts (in MC simulations) are recorded on a monitor plane sectioning the DBS perpendicular to the surface (cf. Figure S7a, Supporting information). To avoid artifacts from the lateral periodic boundary conditions (see Experimental Section) a sufficiently large lateral simulation domain of \( 20 \times 20 \) \( \mu \)m\(^2\) is used. This ensures that any potential spectral contribution from this periodicity lies far outside the considered spectral range. In contrast to the rather complex beetle intra-scale structure the DBS consist of simple building blocks and thus is used for further simulations to keep the computation time manageable.

The FDTD simulations (Figure 2f, black curve) reveal a non-exponential decay with lifetimes \( \tau \) ranging from \( \approx 80 \) up to roughly 100 fs. This directly reflects the lifetime distribution (Figure 2e) possessing a mean value \( \approx 80 \) fs, implying that for longer times the longer living photonic modes dominate the decay. In contrast, the MC simulations (Figure 2f, gray curve) show a mono-exponential decay with a decay constant of 65 fs (cf. Figure S8a, Supporting Information), failing to match both the simulated and measured lifetime distributions. Nevertheless, it is possible to find a set of parameters such that the MC simulations reproduce for the same layer
thickness, the properties of the DBS obtained by FDTD simulations, i.e., reflectance, transport mean free path, and the initial shape of the curve. Hence, we conclude that the initial coherent transport inside the structure can be approximated as diffusive transport emphasizing that there is a diffusion-like scattering regime despite interference effects may occur. However, beyond $t \approx 170$ fs modeling as diffusive transport breaks down and the curve obtained by MC simulation starts to deviate from the FDTD result. Assuming propagation in an effective medium approach (as done for the experiment) yields a pulse round trip time of 160 fs for the 100 fs long pulses applied in the simulations (see Experimental Section). This coincides well with the time at which FDTD and MC simulations deviate indicating that the pulse round trip time is indeed a suitable estimation for the upper limit of the time domain in which diffusion-like photon transport dominates. For longer times, the trapping in weakly localized photonic modes takes over, which is only captured in the fully coherent FDTD simulations.

The FDTD simulations provide means to directly visualize the weakly localized photonic modes inside the DBS structure (inset in Figure 2f). The time averaged local power enhancement in a snippet of the FDTD monitor plane averaged over the time span indicated by the blue line (170–650 fs).
almost constant photon count enhancement across the monitor plane (cf. Figure S8c, Supporting information).

Summarizing the observations and model simulations, we conclude that the scattering yield is dominated by photon leakage from weakly localized photonic modes after an initial scattering time window, which can be roughly estimated as the pulse round trip time in the ultrathin scattering layer treated in an effective medium approach. Such modes have previously been identified for systems that exhibit random lasing with coherent feedback,[25,33] but were not yet identified to significantly contribute to the brilliant whiteness of ultrathin scattering media.

While the spectral features observed here resemble signatures often associated with the strong localization regime, the investigated structures do not fulfill required criteria for Anderson localization like the Ioffe-Regel criterion.[34] Despite DBS and Cyphochilus scales exhibit clear signatures of individually localized modes, the light propagation does not come to a complete halt because of the observed diffusion-like regime. In contrast, in the regime of strong localization the complete suppression of propagation is required even for finite size samples as demonstrated in ref. [27]. Thus, we demonstrate here a case where localization effects significantly influence the properties of photonic structures, even before the onset of strong localization.

As shown in Figure 3, scattering via weakly localized photonic modes is responsible for at least ~20% of the total scattering and thus is relevant when the scattering efficiency of ultrathin disordered photonic media are concerned. As indicated in the background shadings of Figure 3, the scales and the DBS would appear rather grayish and not brilliant white if scattering via leakage from weakly localized photonic modes would be missing.

3. Conclusions

In conclusion, we have experimentally shown that the light transport in scattering and brilliant white structures is dominated initially by a diffusion-like transport that is surpassed by scattering via leakage from weakly localized photonic modes after roughly the pulse round trip time in the ultrathin scattering layer. Leakage from weakly localized modes accounts for at least 20% of the scattered light, underlining their significance for the brilliant whiteness of the ultrathin scattering media. This identification of the coherent weak localization assisted scattering mechanisms based on time-resolved scattered light spectromicroscopy could serve, both conceptionally and methodologically, to gain a better understanding of the transport regimes in disordered materials and their time dynamics. This is, e.g., relevant in imaging through turbid media for bioimaging applications or random lasing action in disordered gain media.[35–37] Furthermore, here, we demonstrated weak localization feature of the biomimetic DBS relying on a distorted Bragg reflector design that provides a blueprint for tailoring nanostructures to particularly support random photonic resonances that can enhance light-matter interaction. Therefore, they may find applications as materials for efficient solar energy harvesting[3,26,38] or sensor applications, where resonance enhanced absorption is employed to improve sensitivity.[39] Investigations into whether the, here, observed phenomena might be found in various other scattering media and gaining a better understanding of the structural properties supporting transport modified by weak localization will be the subject of further studies.

![Figure 3](image-url)

**Figure 3.** Spatially averaged time-dependent accumulated scattering yields. The square modulus of the time-resolved scattering fields are averaged over the recorded positions. This incoherent intensity signal is integrated over time to yield the time-resolved accumulated scattering yield. The background shading at $t_{thr}$ indicates the loss of whiteness if weak localization assisted scattering would be absent. a) Accumulated scattering yield experimentally measured for the Cyphochilus scale. The white vertical line corresponds to a threshold time of $t_{thr}=105$ fs, as indicated in Figure 1c,h, from which one weak localization scattering dominates. The scattering yield from weak localization is 35% (white horizontal line). b) Accumulated scattering yield for the simulated DBS, with a threshold time of $t_{thr}=160$ fs, as indicated in Figure 2f. The scattering yield from weak localization is 21%. c) Accumulated scattering yield experimentally measured for the fabricated DBS, with a threshold time of $t_{thr}=190$ fs (see Supplementary Information), as indicated in Figure 1d. The scattering yield from weak localization is 20%.
4. Experimental Section

Experimental Setup: The light source was a mode-locked Ti:sapphire laser (Femtosecond Scientific, Femtolasers Produktion GmbH, Austria) with a center wavelength of $\lambda_0 = 780$ nm and spectral full width half maximum (FWHM) $\Delta \lambda = 47$ nm, filtered in s-polarization relative to the sample. To achieve microscopic resolution, the beam was focused onto the sample by a parabolic mirror (custom fabricated, Jenoptik, Germany). The sample was moved via a piezo stage (M-664.164, Physik Instrumente (PI) GmbH & Co. KG, Germany) in the focal plane to scan the excitation and detection. The parabolic mirror horizontally separated the incoming beam, the specular reflection, and the scattered light under different angles, allowing to select the measured scattering angle via a blocker aperture. To ensure that only light that was scattered multiple times was registered, the scattered light was measured in cross-polarization with a spectrometer (USB 2000, Ocean Optics Inc., USA).

Phase Reconstruction: The time resolution was achieved by phase reconstruction via spectral interference of the scattered light with a reference pulse. Therefore, the incoming pulse was separated into the sample and reference path. The reference path is delayed relative to the sample pulse and rotated into the measured p-polarization. The resulting interference spectrum $|E_r(t) + E_s(t)|^2 = |E_r(t)|^2 + |E_s(t)|^2 + 2E_r(t)E_s(t)\cos(\Delta \phi(t))$ contains the phase difference $\Delta \phi$ between the two beams. Via Fourier filtering of the interference spectrum and after correcting for the phase imbalance of the interferometer, the phase effect of the sample alone could be reconstructed (see Supporting Information).

Monte Carlo Simulation: Monte Carlo simulations were performed using a self-written Matlab code (The MathWorks Inc., USA) based on the well-known algorithm presented in literature.[22,23] To match the FDTD simulation conditions, no absorption inside the slab was applied and in lateral direction periodic boundary conditions were used. As light source (with $\approx$6.8 billion photons), a plane wave was chosen possessing a temporal profile matching the temporal power profile of the impinging pulse in FDTD simulations. An appropriate monitor cross sectioning the slab was placed according to the FDTD setup. The lateral width of the slab was 12 $\mu$m; the height and effective refractive index were equal to the values given above for the simulated DBS model. The applied transport mean free path of $l_2 = 1 \mu$m was equal to the one obtained by FDTD simulations (see Supporting Information). A scattering mean free path of $l_1 = 1 \mu$m was selected reproducing the FDTD results for short times closely (cf. Figure S8, Supporting Information). The anisotropy factor $g$ is defined using $l_1/3(1-g)$ and hence determined by the choice of $l_1$ and $l_2$.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

R.C.R.P. and D.T.M. contributed equally to this work. The authors gratefully acknowledge financial support from the German Research Foundation DFG within the priority program “Tailored Disorder - A science- and engineering-based approach to materials design for advanced photonic applications” (SPP 1839). The authors thank B. D. Wilts for supplying us with a 3D computer tomography model of the beetle scales’ inner structure. We thank the team of the Nano Structuring Center (NSC) at the Technische Universität Kaiserslautern.
for their support with focused ion beam milling and scanning electron microscopy.

Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
biomimetics, efficient scattering media, multiple scattering, random photonic media, tailored disorder, weak localization

Received: March 24, 2022
Revised: May 31, 2022
Published online: July 19, 2022

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