Sources and gain in photonic random media
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Outlook and applications

Based on the work presented in this thesis, several ideas for new research directions and applications are put forward. Some of these ideas are based on the experimental techniques developed in this thesis, whereas others build upon the concept of studying a source inside a random medium. First, the technique used to unravel the spatial structure of random laser modes is shown to be a promising method for analyzing paint layers in greater detail in Sec. 7.1. Second, we propose an apparatus that measures the turbidity of suspensions by using the side imaging technique of Chapter 4. Third, a new line of research is suggested in Sec. 7.3 by combining random lasers with wavefront shaping. Fourth, in Sec. 7.4 studying sinks rather than sources in random media is discussed.

7.1 A new tool for studying paint

Characterization of multiple scattering media is of tremendous importance in the paint industry. The most expensive component in conventional paint is titania. Optimizing the hiding power of a paint, defined as “its ability to hide the color or color differences of a substrate” [208], while using as little pigment as possible therefore significantly reduces the paint’s cost. For coating applications, the paint is also required to prevent exposure of the underlying material to its surroundings, e.g., to make sure that no rust is formed in vulnerable parts of a ship. To analyze the quality of a coating, non-invasive techniques for studying the substrate while covered by the coating are needed. In this section, we introduce Frequency Correlation Imaging (FCI) as a new method for characterizing the opacity of paint. Moreover, this method is potentially able to reveal the structure of a multilayered paint sample and the optical properties of its substrate.

In passive random media, the scattering strength of samples is primarily characterized by angularly resolved techniques such as enhanced backscattering [53, 54] or total transmission and reflection measurements. The width of the enhanced backscattering cone is inversely
Figure 7.1: (a) Illustration of the principles behind Frequency Correlation Imaging. A paint sample is excited by coherent white light. Due to multiple scattering inside the sample, the excitation light diffuses away from the excitation focus. The average path length traversed in the sample is relatively short close to the focus of excitation and becomes longer when moving away from the focus. As a consequence, the spectral speckle collected far away from the focus contains information about light paths reaching deep into the sample. In this case, the spectral speckle collected far from the focus is influenced by another paint layer that is covered by the top layer. (b) Experimental realization of FCI technique.

proportional to the transport mean free path, which makes enhanced backscattering a great tool for studying strongly scattering samples. Information on long paths traversed in the sample accumulates in the tip of the cone, whereas the wings of the cone only convey information about the short paths traversed in the sample. Thus, enhanced backscattering is foremost a sensitive technique for studying short light paths. In the biomedical community, diffuse optical imaging [209] has proven to be a versatile tool for non-invasive imaging inside weakly scattering tissue, but its resolution has been limited to the centimeter scale. In chapter 6, a confocal detection scheme was introduced to measure the spatial structure associated with narrow spectral features in random lasers. The beauty of this technique resides in that it provides spatially resolved spectral information on the micrometer scale. At the same time, the advance of supercontinuum white-light laser technologies in the past decade allows for extending multiple scattering analyzing tools to a large range of wavelengths [59, 210].

The combination of excitation by a supercontinuum laser and spatially resolved spectral detection results in a powerful apparatus for analyzing paint samples. Figure 7.1 shows an illustration behind the idea of the FCI technique. Coherent white light generated by the supercontinuum laser (Fianium SC-450) is tightly focussed onto the surface of a paint sample. Multiple scattering results in diffusion of light. The diffuse light escaping the sample is collected in reflection. The average path length traversed through the sample depends on the distance between the point of excitation and the point of detection. Close to the focus, the average path length is short, whereas far away from the focus the average path length is long. This difference in path length changes the spectral intensity speckle. These changes in spectral speckle are measured by using our confocal detection scheme that allows for measuring the spectrum at different regions on the surface of the sample. Speckle due to long light paths in the sample decorrelates stronger than speckle due to short light paths, because the longer the traversed path in the medium the larger the phase
7.1. A new tool for studying paint

Figure 7.2: (a) Spectrally averaged contour plot of the diffuse spot measured in reflection. The contour lines are logarithmic. (b) Intensity versus radius for a diffuse spot in reflection. Black disks: experimental data points for parallel polarized light. Black line: fit to diffusion theory. Gray triangles: experimental data points for perpendicular polarized light. Gray line: fit to diffusion theory. (c) Experimental spectra around $\lambda = 604 \pm 25$ nm for different radii. (d) Experimental spectral intensity-intensity autocorrelation for different radii.

Figure 7.2 shows some first results obtained with the FCI technique on a 14 ± 1 μm thick layer of dried white paint. The experimental apparatus that was used is sketched in Fig. 7.1(b). The spatial scan covered 54 × 77 microns and the spectrum was measured from 518 to 724 nm. In Fig. 7.2(a) the iso-intensity contour lines of the diffuse spot in reflection are shown. Spatial speckle has been averaged out by averaging over a frequency interval. The focus of the white-light light source is smaller than 2 μm. By fitting the diffuse spot profile with diffusion theory, as shown in Fig. 7.2(b), the transport mean free path ($\ell = 2 \pm 0.1$ μm), the extrapolation ratio ($z_e/\ell = 2.4$), and the absorption length ($\ell_a = 11 \pm 1$ μm) are retrieved. In addition, the spatial profile of the diffusive spot turns out to be strongly dependent on the measured polarization channel. Close to the focus of the white-light source, the intensity of the parallel polarization channel is higher than the perpendicular polarization channel due to a strong single scattering contribution. Far
Figure 7.3: Turbidimeter based on the method of extrapolation. Light from an incoherent light source is incident on a turbid sample. A detector placed at 90 degrees with respect to the incident beam forms a conventional nephelometer. This part of the apparatus is used to measure the turbidity of weakly scattering samples. For multiple scattering media (samples with high turbidity), the turbidity is measured by imaging the side of the sample onto a CCD camera. Extrapolation of the diffuse intensity to zero intensity, as illustrated in the graph, returns the extrapolation length indicated by the gray dot. From the extrapolation length the transport mean free path is deduced which is a direct measure for the turbidity of the sample.

away from the focus, both polarization channels have equal intensity, because multiple scattering inside the sample scrambles the input polarization and thus results in a complete depolarization. Examples of measured spectra at different radii of the diffusive spot are shown in Fig. 7.2(c). Clear speckle intensity fluctuations are visible. By calculating the intensity-intensity spectral autocorrelation $C(\delta \omega)$ in Fig. 7.2(d), we conclude that the speckle indeed decorrelates faster on a spectral scale for larger radii.

These promising initial results show that FCI is a great way to characterize paint samples. In future experiments, the sensitivity of the technique to absorbing layers buried underneath a white paint layer needs to be investigated. The fact that our setup has a good signal-to-noise ratio for radii up to 15 times the transport mean free path indicates that the apparatus is capable of measuring effects of absorption on relatively long light paths. Depending on the sensitivity of the technique, FCI might even be able to recover images obscured by a white layer of paint, for example by performing the above mentioned experiment for different positions of the focus spot.

### 7.2 Turbidimeter based on the method of extrapolation

The clarity of liquids is often used as an indication for their quality. The industrial method for measuring the turbidity of suspensions relies on so-called nephelometers or turbidimeters. The measured turbidity is expressed in terms of the Nephelometric Turbidity Unit (NTU), which is a measure for the turbidity of the sample with respect to a standard solution of Formazin in water. The basic design of a nephelometer consists of an incoherent white light source and a detector placed at 90 degrees from the beam path that measures the light scattered out from a sample placed in the beam path. These type of nephelometers return accurate results for weakly scattering suspensions, but become useless for multiple scattering media. More complicated designs involving more than one detector exist that increase the turbidity range. Yet, dilution of samples remains required for strongly scattering samples [211, 212].
In the framework of this thesis the turbidity of a sample is expressed by the inverse of the transport mean free path rather than in terms of the NTU. These units are however in principle inversely proportional to each other. In chapter 4, we showed that knowledge of the extrapolation ratio and a side imaging technique enabled us to retrieve the transport mean path of a strongly scattering suspension. We propose a new type of turbidimeter based on this method of extrapolation that is able to measure both weakly and strongly scattering samples.

A sketch of the apparatus is shown in Fig. 7.3. The design is an extension to a conventional nephelometer, in which the diffuse intensity is imaged onto a CCD camera. By extrapolating the diffuse intensity at the edge of the sample to zero intensity, the extrapolation length is found. This extrapolation length is related to the transport mean free path via the extrapolation ratio. Since this ratio is well-known for water based suspensions in a cuvette, the transport mean free path can be deduced. The method of extrapolation is based on diffusion of light and is therefore limited to multiple scattering media. By combining this method with a conventional nephelometer however, a wide range of turbidity samples can be covered. This measurement device therefore reduces the need for dilution of samples.

7.3 Controlling random lasers by wavefront shaping

The spectral output of a random laser has been one of the main topics in this thesis. Introducing an absorber was shown to lead to more control over the emission wavelength in chapter 5. Our approach of tuning a random laser by absorption has recently been extended to the domain of random lasers with narrow spectral features [213]. Yet, controlling the number and spectral position of these spikes in the spectrum has remained very much out of reach for experimentalists. We have seen in chapter 6 that the spectral position of spikes is often chaotic and differs from shot-to-shot. The situation is remotely similar to ordinary speckle patterns where the spatial position of an intensity maximum is difficult to predict a priori. Wavefront shaping of the incident light, that is modulating the amplitude and phase profile of a plane wave, has led to incredible control over the scattered light from a passive random medium [36]. Here, we suggest that wavefront shaping the pump beam in a random laser enables engineering of the spectral narrow features in random lasers.

A sketch of the proposed experimental configuration is given in Fig. 7.4. Pump light is guided towards a spatial light modulator (SLM). Typically, a beam expander needs to be put in the beam path to match the size of the beam with the size of the SLM. The light reflected by the SLM is incident on a random laser sample. In wavefront shaping experiments, one either focuses the excitation light directly onto the sample or one images the separate SLM-pixels onto the surface of the sample [214]. The latter method has the advantage that it becomes possible to compare the wavefront shaping experiment with our work on the spatial structure of random laser modes in chapter 6, where we have measured the spatial distribution of emission light at the surface of the sample. The emission light is analyzed using a high-resolution spectrometer. Depending on the sample, spectral narrow features might appear in the emission spectrum. The spectral intensity of one of these features is selected by the experimenter as a feedback signal of the spatial light modulator. An optimization algorithm is then employed to modulate the amplitude and phase of the SLM, in order to increase the intensity of the selected feature in the spectrum.

Ideally, this pumping scheme leads to the enhancement of one of the narrow spectral
features at the cost of others and to an increased stability of random laser spectra. A serious complication is formed however by the fact that random laser spectra have the tendency to fluctuate strongly from shot-to-shot. These fluctuations stand in sharp contrast with wavefront shaping experiments in passive random media, where the used speckle patterns are stationary. To circumvent this complication, one might reduce the number of lasing modes from the start. For example, by using amplification of light by stimulated emission outside the gain medium [181, 193] or by taking only small samples into consideration [178]. After such a preselection of modes, averaging over multiple shots increases the output stability and hence the possible success rate of the optimization routine.

Our proposal for employing wavefront shaping in random lasers does not only lead to better control over the output of random lasers, it also promises to give more insight in the working principles behind a random laser. When the SLM is imaged onto the sample surface, the amplitude profile of the SLM after optimization corresponds with the excitation structure of a single random laser mode. Using our confocal detection scheme then allows for a direct comparison between the excitation and emission structure of a random laser mode. Moreover, a wavefront shaping experiment on a random laser sample very likely contains quantitative information about how much of the gain medium is shared between the different modes. To that end, the presence and intensity of narrow spectral features other than the one chosen for optimization need to be monitored as well. A complete suppression of the other spectral features is strong indication for little mode overlap inside the random laser sample. Successful implementation of the above brings random lasers closer to the realm of applications as it allows for the selection of a very narrow emission line at a random frequency.

### 7.4 Sinks in random media: the black shades of white

Researchers in the field of multiple scattering of light have shown a strong preference for nonabsorbing systems in the past [215]. This thesis considered systems in which radiation was not just conserved, but also created. We have seen how the interaction of sources with a multiple scattering random medium leads to fascinating phenomena such as random lasing and infinite range intensity correlations. The mathematical symmetry between gain and absorption has recently inspired a team of physicists to construct a so-called coherent...
7.4. Sinks in random media: the black shades of white

The perfect absorber (CPA), in which interference of two coherent light beams results in almost complete (99%) absorption of light in a slab of silicon [166, 167]. In essence, a CPA is the time-reversed analogy of a laser: in a conventional laser a standing wave pattern inside a leaky cavity filled with gain leads to the creation of coherent radiation outside the cavity; in a CPA two sided coherent illumination leads to a standing wave pattern in an absorbing cavity and subsequently to annihilation of the excitation light. The technique of CPA relies on resonances inside the cavity and the coherence of illumination. The technique is therefore intrinsically narrowband. A broadband strongly absorbing sample is one of the holy grail in the field of photovoltaics as such a material promises to drastically increase the efficiency of solar cells. Spherical scatterers placed on top of a silicon wafer show great potential for photovoltaic applications as they guide light into the wafer and thereby lead to enhanced absorption [216].

These studies illustrate how the focus in the field of nanophotonics is gradually enlarged towards including absorption. Three subjects follow naturally from the work described in this thesis when the role of sources is replaced by sinks. First, absorption might lead to intensity correlations that are similar to the ones as induced by a classical dipole source in a random medium. The source in the diagrammatic expression for the $C_0$-correlation is then replaced by an absorber located in close vicinity to a scatterer. In this view an absorbing particle is essentially a miniature version of a detector. If the analogy with the $C_0$-correlation holds, certain configurations of disorder lead to more extinction of incident light than others. In fact, in might well have been this $C_0$-correlation induced by absorption that has led to the observation of intensity fluctuations from a source in the literature [116]. Second, introducing local absorbers in our transverse localization experiment might enable a further selection of participating modes and thereby diminish the observed beating effects. Third, weakly absorbing random media might be made strongly absorbing when the amplitude and phase of the input light are altered using wavefront shaping. The feasibility of such an extension of CPA to the field of random media has recently been evaluated theoretically [217]. Our discovery that diffusion in random lasers make a multiple scattering medium act as an integrating sphere might be of use here since it reduces the need to shape the wavefront over a $4\pi$ solid angle.