Determination of the spectral refractive index of spherical microparticles from optical extinction measurements

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A method is presented to infer the wavelength-dependent real part of the refractive index (RI) of microspheres from extinction (or collimated transmittance) measurements in the wavelength range from 260 nm to 1050 nm.

1 Introduction

The real part of the refractive index (RI) that is, in general, a complex material constant, describes the refraction of a beam of light at a (macroscopic) interface between any two materials. Often times, these materials cannot be condensed into a homogeneous bulk sample, like in the case of microparticles. Media containing particles with sizes comparable to the wavelength of visible light (e.g., a suspension of cells or a cloud) are turbid, i.e., light is scattered in a complex process instead of propagating in straight rays. Nevertheless, the interaction of light waves with such materials is still determined by the optical and geometric properties of the constituting particles. Spherical polystyrene microparticles ("beads") are widely used in biomedical research, e.g., as a calibration material for cell measurements in optical and impedance flow cytometry. However, their high refractive index (n_D=1.59) gives them optical properties much different from most biological samples ($n_D \leq 1.4$), which can be problematic for applications such as optical size determination. Microparticles from lower-RI materials like silica (n_D=1.46) or Poly(methyl methacrylate) (PMMA) (n_D=1.49) are suitable alternatives in these cases. However, they are typically less well-characterized than highly standardized polystyrene particles.

In this work, we present a method to infer the wavelength dependence of the real part of the RI ("real RI" in the following), from a single measurement of the spectral extinction cross section for spherical particles in the Mie scattering regime, i.e., the size is comparable to the wavelength. We exemplify the method for suspensions of PMMA beads to demonstrate the capabilities for low-RI particles.

2 Experiment

The optical setup for spectral transmittance measurements is shown Fig. 1. It is a modified version of a setup previously employed for polystyrene particles and red blood cells [3,5].

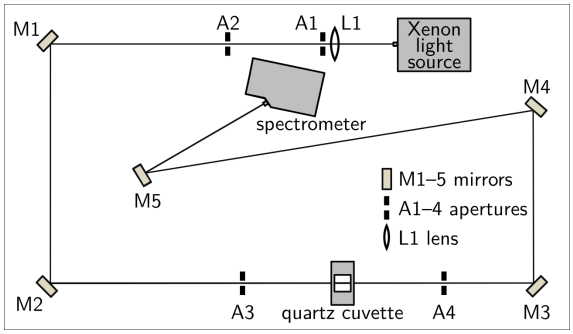

Fig.1 *Optical layout to measure extinction spectra.*

A high-power, continuous HPX-2000 Xenon light source is employed to irradiate the sample in the wavelength range between 185 nm to 2000 nm. Spectra are recorded between 200 nm and 1100 nm with a Maya2000 Pro spectrometer (Ocean Optics, Inc., USA). We use free-beam optics instead of connecting the components via fibers to achieve the very low divergence angles required for optical extinction measurements. With the help of 5 mirrors M1– M5 the long light path is folded and guided through the sample cuvette into the spectrometer. The lens L1 is used as condenser to obtain an approximately parallel light beam. The apertures A1 – A3 serve to reduce the size of the beam to a diameter of about 1 mm corresponding to a divergence of 0.25 mrad (half angle). The sample (aqueous particle suspension) is filled in a quartz cuvette (type 110-QS, Hellma GmbH& Co. KG, Germany) with *d* = (10 ± 0.01) mm optical path length. The cuvette is mounted in a temperature-controlled sample holder (Quantum Northwest, inc., USA), set to 23°C. Aperture A4 blocks the light scattered in the non-forward direction by the sample to suppress background light. The spectrometer's entrance opening of 1.0 mm diameter corresponds to an observation angle of 0.3 mrad (half angle). The low divergence illumination and detection suppress light scattered at small angles into the spectrometer's aperture. This allows to measure the directed transmittance (exact forward direction) and analyze the measurements based on optical extinction cross sections. The experimental setup allows to measure the same quantity as a previous monochromator-based

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setup used by Gienger *et al*. [1]. However, the setup in Fig. 1 offers a significant advantage, since due to the parallel detection of the spectrum in contrast to the previously used wavelength scanning, the measurement time is reduced from typically 20 min to about 10 s.

When measuring a sample, the suspension of PMMA microspheres was pre-diluted appropriately and pipetted in a water-filled cuvette in several steps to allow for measurements at multiple, increasing particle concentrations. These concentrations were selected such that the transmittance $T(\lambda)$ ranged from roughly 95% down to 30% for each sample. We compared the log-transmittances [cf. Eq. (1) below] at the different concentrations to ensure that multiple scattering effects are negligible. From the measured transmittance $T(\lambda)$ we compute the ensemble-averaged extinction cross section $C_{ext}(\lambda)$ of the particles in the sample according to Eq. (1)

$$C_{ext}(\lambda) = -\ln[T(\lambda)]/d\,c, \qquad (1)$$

where $d = 10\ mm$ is the thickness of the cuvette and c is the number concentration of the particles. The latter is estimated from the known initial concentration and volumetric dilution and the value is later refined during data analysis.

3 Results

Mathematically, the experiment can be described using the Lorenz-Mie Theory (LMT) [2], which is the analytical solution of the light scattering problem for a single spherical particle and a plane wave. The measured $C_{ext}(\lambda)$ curves were therefore then fitted using a mathematical model based on the LMT. This model is based on a series representation of the real RI $n(\lambda)$ using specific, spectrally localized basis functions [1, 3]. The expansion coefficients, as well as the particle concentration and parameters of the particle size distribution (mean and standard deviation) are free parameters of the model. Their values are obtained from the measured curves by nonlinear optimization.

We measured the collimated transmission spectra of two samples of PMMA microspheres with different sizes. The corresponding extinction cross sections $C_{ext}(\lambda)$ are shown in Fig. 2 (left) for diameter 1.3 µm particles (blue curve) and 2.0 µm particles (green curve). The analysis is limited to the region $\lambda \geq 260\,nm$, where PMMA is quasi-nonabsorbing.

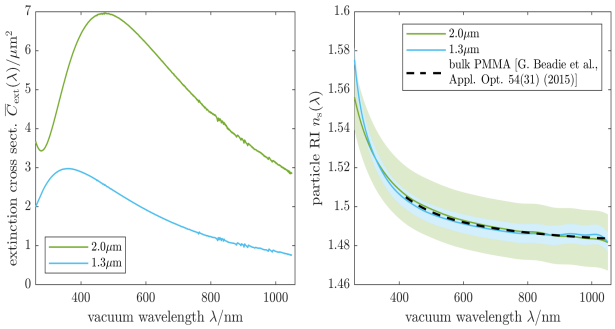

***Fig.2** Measured extinction coefficient (left) and calculated refractive index of PMMA microparticles (right).*

The two samples exhibit significant variation of the extinction cross sections, because the spectral positions of the resonances due to light scattering depend on particle size. For example, the maximum of the extinction is red-shifted for the larger particles. At the IR end of the spectrum, both samples feature a similar decay of $C_{ext}(\lambda)$ with λ. The resulting real RI obtained by nonlinear optimization is shown in Fig. 2 (right). Our results are in good agreement with values measured on bulk material [4]. Additionally with our method we were able to expand the data set of the RI for PMMA well below 400 nm, where no literature values are available.

4 Summary

We presented a method to infer the wavelength-dependent real refractive index (RI) of microspheres from extinction measurements in combination with an adapted mathematical model in the wavelength range from 260 nm to 1050 nm. We have demonstrated its potential for low RI materials using PMMA particles.

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Literatur

[1] J. Gienger et al., Appl. Optics 57 (2), 344-355 (2018)

[2] M. Daimon and A. Masumura, Appl. Opt. 41, 5275–5281 (2002)

[3] J. Gienger, Dissertation 2019, in PTB-Bericht Opt-85 (April 2019)

[4] G. Beadie et al.: Appl. Optics 54 (31) 139-143 (2015)

[5] J. Gienger et al., *Sci. Rep.* **9**, 4623 (2019)