Sizing of single evaporating droplet with Near-Forward Elastic Scattering Spectroscopy

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ABSTRACT

- An optical setup and numerical models to analyse spectral properties of single evaporating microdroplet was developed [1].
- A new approach combines the advantages of the electrodynamic trapping with the broadband spectral analysis with the supercontinuum laser illumination.
- The elastically scattered light within the spectral range of 500 – 900 nm is observed by a spectrometer placed at the near-forward scattering angles between 4.3°and 16.2° and compared with the numerically generated lookup table of the broadband Mie scattering.
- The solution has been successfully applied to infer the size evolution of the evaporating droplets of pure liquids (diethylene and ethylene glycols) and suspensions of nanoparticles (silica and gold nanoparticles in diethylene glycol), with maximal accuracy of ±25 nm.
- Newly developed Near-Forward Elastic Scattering Spectroscopy has been compared with the previously developed sizing techniques: (i) based on the analysis of the Mie scattering images - the Mie Scattering Lookup Table Method [2] and (ii) the droplet weighting.

EXPERIMENTAL SETUP

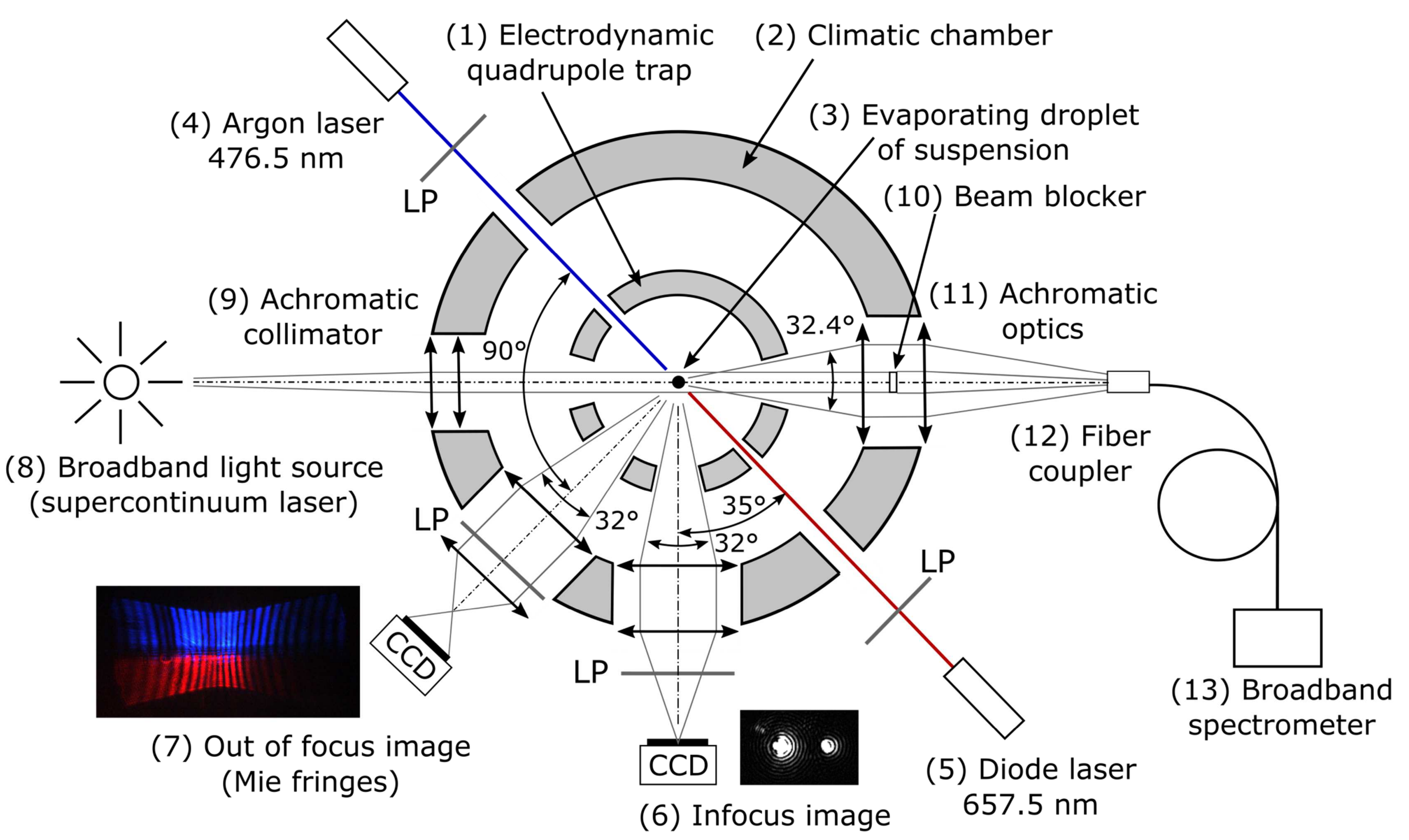

Fig. 1. Schematics of the experimental setup: electrodynamic trap equipped with optical characterization devices and enclosed in the climatic chamber [1].

NEAR-FORWARD ELASTIC SCATTERING SPECTROSCOPY (NFESS)

Principles of the NFESS

The NFESS technique measures the scattering (extinction, when absorption is present) spectra of a droplet or a solid particle (aggregate) illuminated by a collimated, polychromatic light beam. Thus, this optical technique requires a beam with spectral range λ_i and intensity $I_0(\lambda_i)$ to be scattered (absorbed) by the droplet (aggregate) under analysis. The scattered spectral intensity $I(\lambda_i)$ is collected by an achromatic optical system and analysed via a spectrometer. The beam scattered spectral intensity recorded by the spectrometer is given by the following equation:

$$I(\lambda_i) = I_{sca}(\lambda_i)\, I_0(\lambda_i)$$

To infer scattering spectra of the analysed object $I_{sca}(\lambda_i)$, the reference spectra of the supercontinuum laser $I_0(\lambda_i)$ must be known in advance. An example of the experimental dataset processing for a droplet of diethylene glycol is shown in Figure 2 (see figure caption).

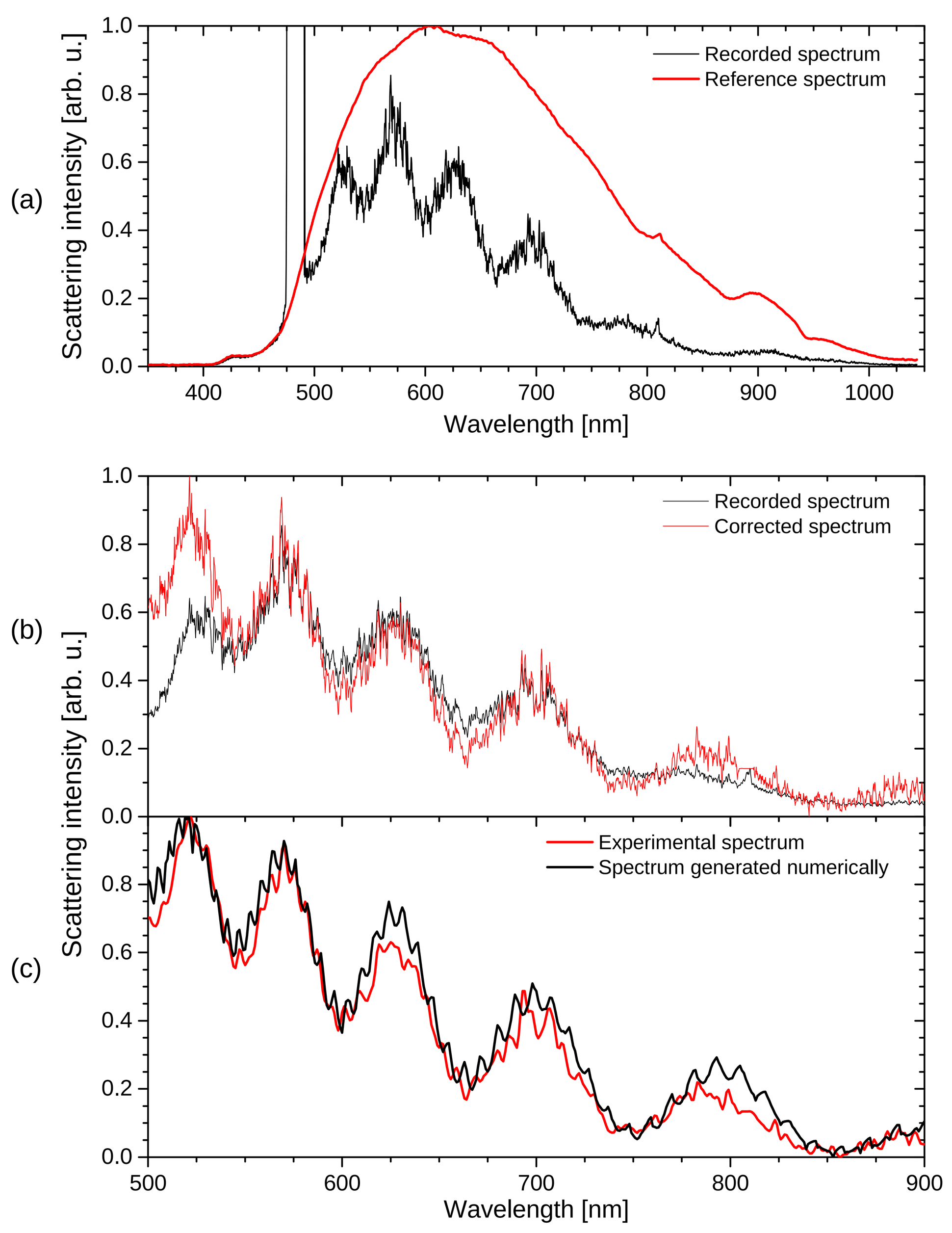

Fig. 2. Example of the experimental dataset processing:

(a) the supercontinuum reference spectrum (red solid line) and the original intensity of light scattered by the droplet (diethylene glycol with radius ~6.35 μm, black solid line),

(b) the same spectrum narrowed to the region of interest (black solid line) and after the background correction and dividing by white light reference (red solid line),

(c) spectrum filtered with FFT filter (black solid line) and numerically generated spectrum corresponding to the experimental results (red solid line).

Inversion model and data post-processing

Numerical procedure used for inversion of the experimentally recorded data is based on the fitting of the Mie theory predictions to the experimentally obtained scattering spectra. It seeks the least squares solution x that minimizes the norm:

$$Min\{\|b - \boldsymbol{A}x\|\},$$

where b is the m-length vector containing experimentally recorded spectra for the given wavelengths and **A** is the m×n matrix (lookup table) with numerically generated scattering patterns for the same number of wavelengths m spanning the n-length range of droplet radii.

RESULTS AND DISCUSSION

The results obtained with Near-Forward Elastic Scattering Spectroscopy (NFESS) have been compared with the previously developed sizing techniques:

(i) **Mie Scattering Lookup Table Method (MSLTM)** [2]. Based on the analysis of the Mie scattering images solution provides high accuracy of ±10 nm in favourable cases and has been applied to several studies performed in our laboratory.

(ii) **Droplet weighting** [2] - method based on the analysis of the constant electric signal U_{DC} used for the droplet stabilisation. The temporal evolution of droplet mass-to-charge ratio is used to derive a mathematical formula describing droplet radius.

Evaporation of pure liquids

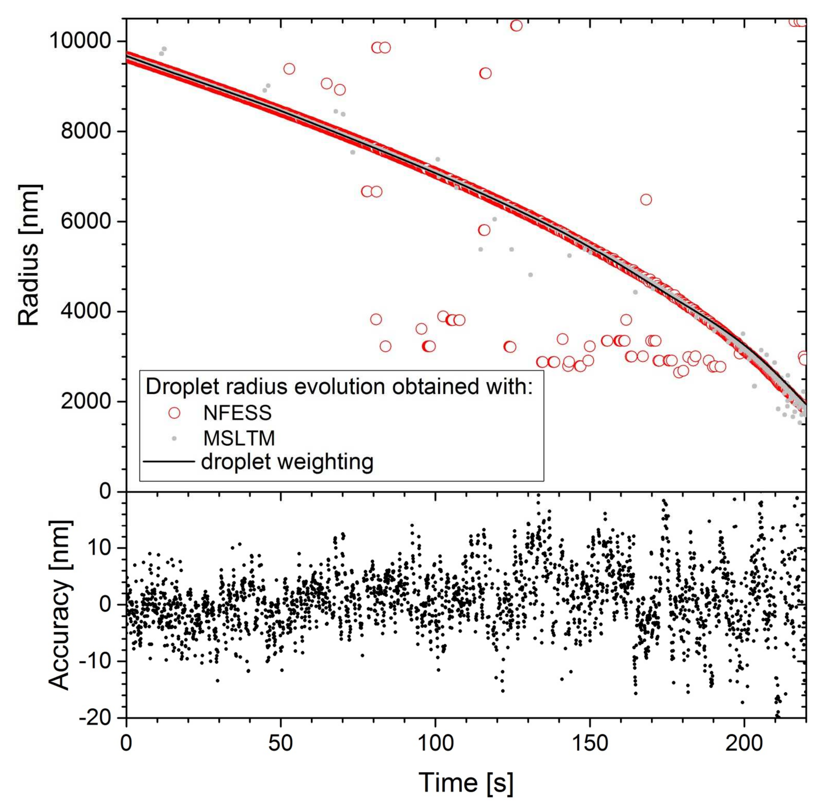

Fig. 3. Evaporation of droplet of **diethylene glycol (DEG)**. Top figure: Droplet radius evolution obtained with: (i) NFESS - open red circles, (ii) MSLTM - grey dots and (iii) droplet weighting - solid black line. Bottom figure: Relative error (difference) between the NFESS and the MSLTM obtained after removal of the outlying points.

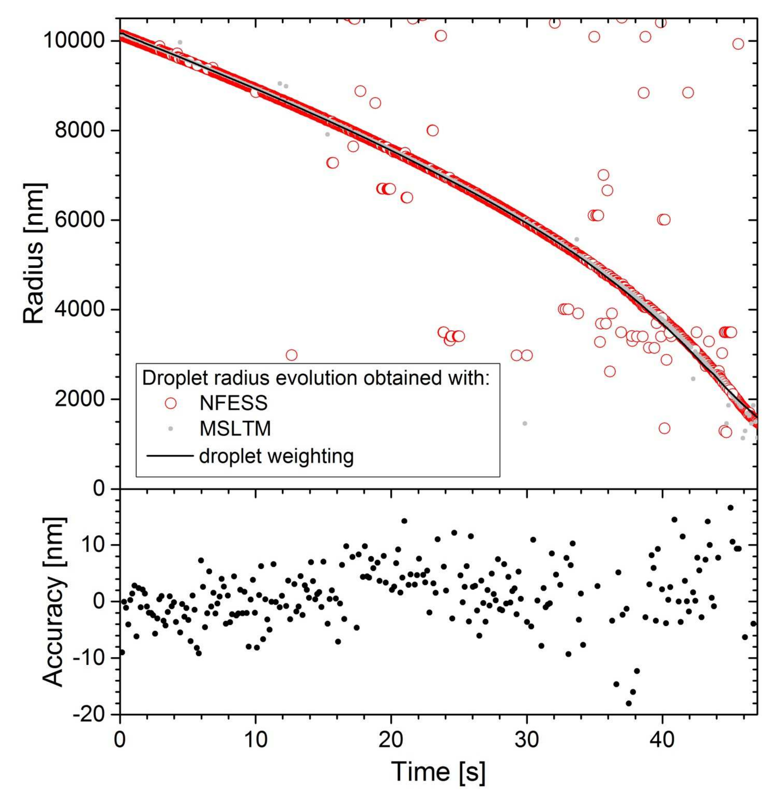

Fig. 4. Evaporation of a droplet of **ethylene glycol (EG)**. Top figure: Droplet radius evolution obtained with: (i) NFESS - open red circles, (ii) MSLTM - grey dots and (iii) droplet weighting - solid black line. Bottom figure: Relative error (difference) between the NFESS and the MSLTM obtained after removal of the outlying points.

Evaporation of nanoparticle suspensions

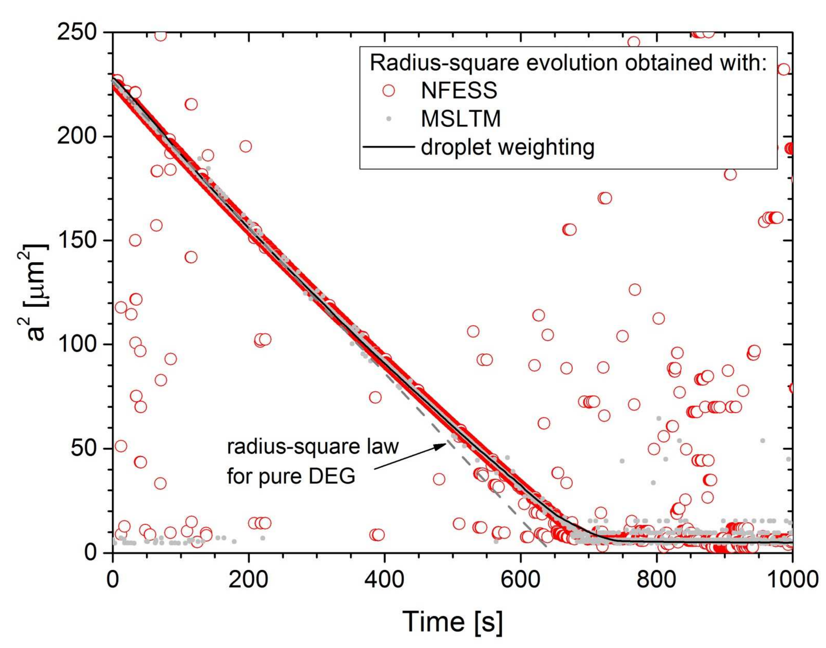

Fig. 5. Evolution of the evaporating droplet of suspension of 250 nm **silica nanoparticles** in **diethylene glycol** with 1:50 mass fraction obtained with: (i) NFESS - open red circles, (ii) MSLTM - grey dots and (iii) droplet weighting - solid black line. Dashed dark grey line (iv) shows the radius-square law for a droplet of pure DEG of the same initial radius.

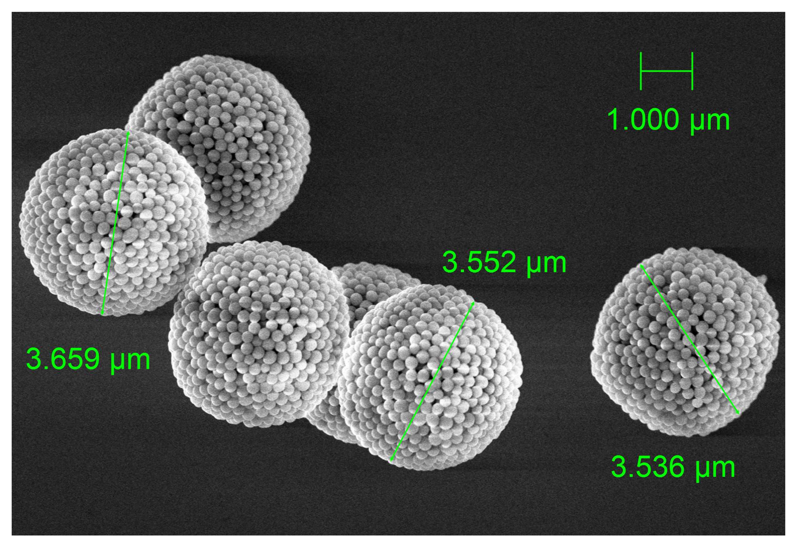

Fig. 6. SEM image of the experimentally obtained highly-ordered aggregates with external diameter of 3.5–3.7 μm built of approx. 4300–5000 of 250nm-diameter silica. Results obtained in the parallel experiment to the one presented in this work (see [3] for more details).

ACKNOWLEDGMENTS

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