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Dynamic light scattering investigation of single levitated micrometre-sized droplets containing spherical nanoparticles

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Abstract

We applied the dynamic light scattering (DLS) technique for studying single microdroplets of dispersions of SiO\textsubscript{2} and TiO\textsubscript{2} nanoparticles. The evaporating microdroplets were levitating in an electrodynamic quadrupole trap. Exponentially decaying autocorrelation functions (ACFs) were obtained for the light scattered by evaporating microdroplets as well as for microaggregates of nanoparticles formed at the end of the evaporation process. It was found that the temporal variation of the ACF generally reflects the evolution of the microdroplet of dispersion. At the initial stage of the evaporation, apart from the optical morphology resonances of the droplet (dielectric sphere), it was possible to identify the characteristic times corresponding to the Brownian motion of the dispersed nanoparticles. At the end of evolution, when the drying microdroplet transforms into a (non-rigid) microaggregate, the Brownian motion of the dispersed nanoparticles was masked by the rotational Brownian motion of the microaggregate as a whole.

Keywords: Dynamic light scattering, autocorrelation function, nanoparticle, microdroplet, electrodynamic trap

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1. Introduction

Nowadays, a wide variety of micro- and nanoparticles is produced for applications in technology and biosciences (see e.g.: [1, 2, 3, 4] for reviews). The diversity concerns shape, size, material and its distribution. Dispersions of such particles gain additional functionality at the expense of their complexity. Furthermore, dispersions are commonly encountered as droplets and microdroplets in particular. Such shape of the gas-liquid interface additionally imposes spherical symmetry onto the system modifying its properties. Therefore, efficient characterisation methods are pursued, both for bulk materials and droplets. On the other hand, droplets of complex suspensions are formed in the atmosphere via the process of so-called “scavenging” [5] – water droplets of clouds (also often condensing on aerosols), fog and rain absorb gases and various atmospheric aerosols, both soluble and insoluble. The complex droplets of suspension/mixture undergo further evolution, in particular, evaporation, which leads to self-organisation of inclusions. Inclusions such as micrograins of sand or spores can aggregate into fairly regular structures. Since aerosols have a significant impact upon (radiative) energy transfer in planetary atmospheres (see e.g. radiative forcing but also visibility impairment), their optical properties have utmost importance (see e.g. [6] and references therein) and require in-depth studies.

In order to effectively investigate evaporating composite droplets it is necessary to use a technique that provides non-invasive and non-contact means of droplet characterisation. There are rather few methods that might enable characterisation of the internal or at least the surface structure of a composite droplet (compare e.g.: [7]). For instance, static light scattering-based methods were investigated: white light scattering by Matic et al. [8], rainbow refractometry by Saengkaew et al. [9] and Li et al. [10] and digital in-line holography by Coëtmemellc et al. [11]. The applicability of dynamic light scattering (DLS) was investigated e.g. by Bronk et al. [12] and Krieger et al. [13]. However, the conclusion of [13] was rather discouraging. Since our preliminary studies [14, 15] suggested otherwise, in this work we’ve revisited the issue.

For bulk fluid dispersions, DLS method is predominantly used for measuring particle size and size distribution, studying relaxation and aggregation processes, indirect viscosity measurement etc. (see e.g. [16, 17] for review). The DLS technique is based on the analysis of the intensity fluctuations of the scattered light, caused by Brownian motions of the dispersed particles.
It requires measuring the autocorrelation function (ACF) of the fluctuations [18, 19]. A standard procedure of the ACF analysis for polydisperse samples is the application of the method of cumulants [18, 20]. However, it must be kept in mind that the results obtained with the DLS technique depend to a large extent not only on the data quality but also on the data processing method. The reliability of the results depends on the completeness of the applied model of the observed processes.

In this paper we adapted the DLS technique, usually used for macroscopic samples in cuvettes, to study single levitated micrometre-sized droplets of liquids containing spherical nanoparticles. Bronk et al. [12] reported the application of ACF technique to characterisation of the levitating droplets of liquid/dispersion under similar experimental conditions. However, the results were obtained for non-evaporating droplets, only for one order of magnitude in the ACF time scale. Krieger et al. extended the idea proposed by Bronk et al., though they concluded that “diffusional motion of an inclusion within the aqueous solution of a host droplet is masked by rotational Brownian motion”. In our previous research [14, 15], we found that we are probably capable of finer measurements. We found some features of the DLS signals that could be connected with the properties of a microdroplet of dispersion. In this work we further follow this track and carefully study the relationship of the measured ACF to the changes in the size and composition of the evaporating droplet.

2. Basic relations and experimental setup

2.1. Autocorrelation function in DLS method

The random motion of particles in dispersion with the temperature $T$ and the viscosity $\eta$ is characterised by the diffusion coefficient $D$, which by means of the well-known Stokes-Einstein relation $D = k_B T/(6\pi \eta R_H)$ is related to the so-called hydrodynamic radius $R_H$. In order to obtain the hydrodynamic radius with the DLS method it is necessary to measure the time dependence of the autocorrelation function $G(\tau)$ of the scattered light intensity $I(t)$. For a delay time $\tau$, the normalised ACF is given by [17, 21]

$$G(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2}.$$  
(1)
For a monodisperse system of particles with Brownian motion the ACF exhibits the following exponential decay [19]:

$$G(\tau) = A_0 + A_1 \exp(-2Dq^2\tau),$$

(2)

where $A_0$ is the baseline of the ACF for $\tau \to \infty$, and $A_1$ is the amplitude of the ACF at $\tau = 0$. The scattering vector

$$q = \frac{4\pi n}{\lambda} \sin(\theta/2)$$

(3)

is connected with the refraction index of the dispersion medium $n$, vacuum wavelength of the incident light $\lambda$ and with the scattering angle $\theta$. In many cases the intensity correlation function (2) can be written in the form of the so-called Siegert relation [18, 22], in which $A_0 = 1$ and $A_1 = \beta$ is the coherence factor, which is determined by the experimental conditions. The exponential time constant $\tau_0 = 1/(2Dq^2)$ can be easily found from the $G(\tau)$ curve for the monodisperse sample. This is the characteristic time of the considered diffusion process. However, in most cases the samples are polydisperse and the ACF becomes a sum of single exponential decay functions with different decay time constants (characteristic times) $\tau_i$ [23].

2.2. Experimental details

In this work, single droplets of diverse dispersions of nanoparticles, levitating in an electrodynamic quadrupole trap, are investigated with light scattering techniques. We used two types of nanoparticles: SiO$_2$ and TiO$_2$. The first represents natural nanoparticles - fine grains of sand, while the other represents nanoparticles in technological application. Water, diethylene glycol (DEG) and tetraethylene glycol (TetraEG) were used as the dispersion media. The droplets, with initial diameters in the $20 - 30\mu m$ range, were introduced into the electrodynamic trap by means of a droplet-on-demand injector [24, 25]. The trap was placed in a small chamber with dry nitrogen atmosphere at room temperature ($\sim 22^\circ C$) and atmospheric pressure. The dry nitrogen is obtained from LN boiling freely in a vessel. On the way to the chamber, it passes through the heat exchanger in the thermostatic bath in order to equalise the gas temperature with the temperature of the chamber. The chamber is flushed with dry nitrogen before each droplet injection and the dry nitrogen flow is stopped only for the duration of the droplet measurement. The droplet carried an electric charge (gained by charge separation in the electric field on passing through the injector nozzle) which
enabled stabilisation of its position using a combination of the static (DC) and oscillating (AC) electric fields. There are several mature droplet levitation techniques (see e.g. [26] for a review). We’ve chosen electrodynamic levitation rather than optical or acoustic (compare [7]), since it enables trapping of (sub)microdroplets and causes smallest droplet deformation [27, 28]. The AC field was in a quadrupolar arrangement, so in the very centre of the trap, the AC field strength (ideally) went to zero. In consequence, the micromotion (with the frequency of the AC field) of a particle kept in the centre of the trap is minimal. Furthermore, a particle in the ground state of the trap performs no secular motion (macromotion). The DC field is used to balance the particle weight and actively stabilise the particle in the centre in a vertical direction. The levitated droplet was illuminated with a collimated vertically polarised blue Ar\textsuperscript{+} laser beam (497 nm, 50 mW) and/or horizontally polarised red semiconductor laser beam (654 nm, 20 mW). The in-focus image of the droplet, registered with a CCD camera, was used to determine the droplet position and measure its distance from the centre of the trap (reference position) in arbitrary units (pixels). This measurement provided the error signal for feed-back loop (operating at 50 cycles/s), which

Figure 1: The optical scheme of the dynamic light scattering experiment in the electrodynamic trap.
controlled the DC voltage source and stabilised the particle at the reference position. It should be noted that the applied DC voltage is proportional to the droplet mass-to-charge ratio and with proper calibration can be used for finding the (effective) droplet mass/radius (electrostatic weighting). See e.g. [25] for the details of our trap arrangement.

The optical scheme for the scattered light detection consisted of a two-lens objective (using aspherised lenses) and an optical fibre (Fig. 1). The droplet was in the focus of an objective and a diaphragm of 1-mm-diameter limited the numerical aperture to ensure the scattering angle very close to 90°. For the detection of the fluctuations of the light scattered by a single micron-sized droplet, we used a fast photon-counting PMT (R649, Hamamatsu) with a thermoelectric cooler and an amplifier-discriminator (F-100T, Advanced Research Instruments Corporation) that sends the TTL-level 10-ns pulses to the real-time digital correlator (Photocor-FC, Photocor Instruments) [29].

The autocorrelation functions were analysed with Alango DynaLS software [30] to obtain the corresponding characteristic times by discrete component analysis. Although only a small part of the 2 mm laser beam was scattered by a microdroplet, the sensitivity of the DLS setup is sufficient to get the signal level of $10^5 - 10^6$ cps. As a result, we could obtain a high-quality autocorrelation function after a time of only 10 s. However, in order to expand the temporal range of the ACF, accumulation time sometimes reached $t_a \sim 10^3$ s.

The light scattered by the droplet was also collected with a CCD camera as out-of-focus images. For (nearly) homogeneous and spherical microdroplets, such as usually encountered at the beginning of their evolution in the trap, the Mie-scattering patterns [31] could be used for very accurate droplet radius measurement [25]. Such measurement was used for precise calibration of the droplet electrostatic weighting. Then the electrostatic (DC field) weighting enables measuring the droplet/aggregate (effective) radius during the whole evolution (evaporation), in particular for the rigid aggregate.

A conventional DLS setup with a cuvette was used for reference particle size measurements for the given dispersion. It utilised a diode laser working at 405 nm wavelength [14, 15]. These experiments were also performed at room temperature.

The coherence factor $\beta$ of the measured ACF is strongly dependent on the geometry of the experiment, in particular on the number $N$ of coherence areas $A_{coh}$ that is defined by the following relation [17, 32]:

$$NA_{coh} = \frac{\lambda^2 l^2}{\pi d^2}.$$  (4)
Here $l$ is the distance of the detector from the scattering volume, and $a$ is the scattering radius (i.e. the microdroplet radius). It is important for a good signal quality that the radius of detector $b$ is chosen to have $N \approx 1$ and $A_{coh} = \pi b^2$. Increasing the number of coherence areas above unity leads to a decrease of the ACF amplitude [32, 33] that is associated with the decrease of the coherence factor. The geometry of our experimental setup (Fig. 1) meets the condition of $N \approx 1$. The optical fibre of $2b = 400 \mu m$ core diameter defines a much smaller viewed-volume and at the same time increases the measurement sensitivity [34, 35].

![Figure 2: The optical morphology resonances manifesting in the temporal dependence of the intensity of light scattered by a very slowly evaporating droplet of tetraethylene glycol with titanium dioxide nanospheres. The structure of several consecutive resonances in 3000-4100 s range is shown in detail in the right panel.](image)

At the initial stage of a microdroplet’s evolution, the optical resonances of a dielectric sphere (also known as Morphology Dependent Resonances) [36, 37] dominate in the temporal changes of the scattered light intensity (see Fig. 2). The analysis of these resonances enables an accurate relative or even absolute measurement of the temporal evolution of the droplet radius [38, 39]. Alternatively, as mentioned above, it is possible to analyse the angular distribution of the scattered light using a high-resolution CCD camera with a wide dynamic range [24, 25, 40].
3. ACFs for levitating microdroplets of diverse dispersions of nanospheres

3.1. ACFs for the evaporation stage of microdroplet

Figure 3 shows representative autocorrelation functions that were obtained over a broad (nine orders of magnitude) temporal range for a tetraethylene glycol (TetraEG) dispersion of the 250 nm titanium dioxide ($\text{TiO}_2$) nanospheres. Here and in the following figures, the ACF is presented in normalised form as $(G(\tau) - 1)/\beta$ according to the relation (1), and in semi-log scale, unless specified otherwise. The coherence factor $\beta$ is determined in our setup by the real-time correlator.

The autocorrelation function for the accumulation time $t_{a1} = 10000$ s (Fig. 3, curve (1)) exhibits a periodic character (with period $T = 296$ s). The corresponding light intensity oscillations are presented in the right panel of Fig. 2 in a ten times shorter time-scale, to show the structure of the morphology resonances in greater detail. In order to identify the Brownian motion of TiO$_2$ nanospheres we used a short-time part of this ACF obtained after subtraction the long-time $A_0$ level (according to relation (2)) and normalisation. Such procedure was possible since the periodic part of the ACF changes very little over the first few orders of magnitude on a time scale.

The short-time part is represented in Fig. 3 by the curve (2). There are several characteristic times that can be identified in this part of the full ACF. The longest characteristic time of about 1 s is the remnant of the long-time part and should be associated with the optical resonances, which can be seen in Fig. 2. The shortest characteristic time ($\tau_1 = 0.034$ s) corresponds to the Brownian motion of titania nanospheres in the microdroplet.

The small local maxima that are present on the curve (2) correspond to the frequency of the AC field of the electrodynamic trap. In our experiment, the AC frequency varied from 200 Hz at the beginning of the droplet evolution to about 700 Hz at the final stage, when most of the liquid has evaporated and the mass of the resulting aggregate was significantly smaller. The presence of such maxima in the ACF confirms the high sensitivity of the technique to small changes in the intensity of scattered light, such as those connected with the small droplet’s oscillations at the AC frequency, relative to the equilibrium position – so-called micromotion. Since the droplet/aggregate was in the ground-state of the trap, there was no droplet macro-motion at the eigenfrequency of the trap.
Figure 3: Normalised autocorrelation functions for 250 nm TiO$_2$ nanospheres in TetraEG. (1-4) pertain to the microdroplet and wavelength $\lambda = 497$ nm, while (5) pertains to the bulk sample in a cuvette and wavelength $\lambda = 405$ nm. (1, 2): ACF with the accumulation time $t_{a1} = 10000$ s, (3, 4): subsequent ACF with $t_{a3} = 20.5$ s and $t_{a4} = 7.1$ s respectively. The inset shows a typical snapshot of the Mie-scattering pattern for wavelength $\lambda = 497$ nm at the beginning of the droplet evaporation.

In order to eliminate the effects of long-time signal changes upon the ACF, smaller accumulation times could be used (see curves (3) and (4) in Fig. 3). On the other hand, the Brownian motion of the titania nanospheres in TetraEG is very slow and a rather long measurement is required. Therefore, the accumulation time $t_{a4} \approx 7$ s (see curve (4)) seems not sufficiently long to get a good quality ACF.

The ACF labelled as (5) was obtained with the conventional DLS setup (in a cuvette) for the same dispersion. Taking into account the difference in the wavelength of the scattered light, we found $\tau_5 = 0.023$ s and got a good agreement with the results obtained for a microdroplet. This indicates that, in principle, in our DLS experiment with the levitating droplet it is possible to detect the Brownian motion of nanoparticles in a droplet of micrometre size. However, it must be kept in mind, that this may not be possible for quickly evaporating microdroplets (dispersion media of higher volatility) since relatively long ACF accumulation times are necessary.
3.2. ACFs for microaggregates

In order to prevent multiple light scattering, the concentration of nanoparticles in the microdroplet should be small. We used various initial concentrations from 0.25 down to 0.005 wt% of silica and titania particles dispersed in water. Knowing the density of silica and titania in nanoparticles, as well as of water, it can be found that, for example, for 0.25 wt% concentration of the 250 nm nanospheres there are respectively $\sim 300$ of TiO$_2$ and $\sim 500$ of SiO$_2$ nanospheres inside a 20 $\mu$m microdroplet.

![Figure 4: (a) Experimental in-focus image of a levitated, pure TetraEG, $\sim 40$ $\mu$m droplet and (b, c, d) consecutive in-focus images of an aggregate - at the final stage of evolution - obtained from a water droplet containing $\sim 500$ TiO$_2$ spheres of 250 nm diameter. Image (a) was obtained with additional back-illumination with unpolarised white light, in order to visualise the droplet boundaries.](image)

Assuming a uniform distribution of nanoparticles in the volume, the average distance between them is $\sim 3 \mu$m then. Thus, the number of scattering centres in the microdroplet is small in the optical path (image of such droplet is similar to that of a pure liquid droplet, see Fig. 4a), which explains the small amplitude of the ACF at the initial stage of temporal droplet evolution. In the case of water as the dispersion medium, a sensible ACF accumulation
time significantly exceeds the interval between the optical morphology resonances. As mentioned above, since the autocorrelation of a periodic signal from the photon counter system is also periodic with the same period, the ACF analysis is significantly hindered at this stage as longer accumulation times are not accessible. Thus, for a quickly evaporating droplet (e.g. of aqueous dispersion) the ACF with exponential decay can be obtained only at the end of the droplet evolution.

The evaporation of the dispersion medium leads to an increase in the particles’ concentration and ultimately to the formation of (non-rigid) aggregates. The images in panels (b), (c) and (d) in Fig. 4 correspond to an aggregate consisting of ~ 500 titania particles, after the water evaporation (the initial concentration of nanoparticles of 250 nm diameter was 0.025 wt%). The roughness of the surface of the aggregate can be inferred from the images. It was shown in the SEM study of the aggregates deposited on a substrate [41] that their overall shape is regular - close to spherical - and the irregularities of the surface are minor.

The small size of the aggregate precludes the application of 2D-autocorrelation analysis, as it was done in [42]. We obtained the ACFs of diverse aggregates using different initial concentrations of silica and titania spherical nanoparticles. Systematic observations have shown that the ACF almost always had an exponential decay like in the case of a monodisperse scattering bulk medium. The regular rotational motion of a rigid aggregate, if present, is also expected to introduce a periodicity into the autocorrelation function for the large accumulation times. However, we have never found such periodicity. Small maxima visible on curves (1), (2) in Fig. 3 and 5 correspond to the frequency of the AC field of the trap and were by default eliminated by the proper selection of the DC voltage and AC frequency as described in Section 2.2. It is however expected that the frequency of free aggregate rotation would be independent and different from the AC frequency.

As the reference, the ACFs was also found in the standard DLS setup in a cuvette for the laser wavelength of 405 nm (curve (5) in Fig. 5). It confirmed the monodisperse character of the studied dispersion, with the characteristic time $\tau_5 = 6.1 \times 10^{-4}$ s. In accordance with relation (3), the characteristic times obtained from standard DLS experiments for the same conditions should be scaled with the wavelengths of the illumination source. For instance, for 497 nm and 654 nm, the exponential ACF decays with the characteristic times of $9.2 \times 10^{-4}$ and $16.3 \times 10^{-4}$ respectively can be expected. Then, $\tau(405) : \tau(497) : \tau(654) = 1 : 1.5 : 2.7$. 

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Figure 5: A series of the ACFs that were measured for 0.004 wt% aqueous dispersion of 250 nm TiO$_2$ nanoparticles in droplets and in cuvette. Curves (1), (2) correspond to ACFs for a larger droplet, while curves (3), (4) correspond to a smaller droplet. Curves (1), (3) correspond to the wavelength of 654 nm, while (2), (4) correspond to the wavelength of 497 nm. Curve (5) corresponds to the measured in a cuvette at the wavelength of 405 nm. The inset presents the curves (3), (4) and (5) in ln-lin scale in a short range of delay time.

However, in our DLS experiment with microdroplets (curves (3) and (4) in Fig. 5) we obtained $\tau_3(654) = 1.23 \times 10^{-3}$ s and $\tau_4(497) = 0.61 \times 10^{-3}$ s, which corresponds to the ratios of 1 : 2.2 : 4.4, significantly differing from the theoretical prediction for the Brownian motion. It can also be noticed in the inset in Fig. 5 (in ln-lin scale) that curves (3) and (4) exhibit a somewhat different character (curvature) than curve (5). As expected, the ACFs obtained for microaggregates exhibit properties, which do not correspond to the standard assumption of the Brownian motion of the nanoparticles.

Fig. 6 shows the ACFs corresponding to final stages of evolution of four different droplets that contained silica nanospheres dispersed in DEG and TetraEG respectively. First of all, the opposite shift in the distribution of the characteristic times versus the diameter of the nanospheres (100 and 450 nm) can be noticed. Secondly, despite the significant difference of viscosity between DEG and TetraEG the obtained characteristic times hardly differ for the same nanoparticle diameter (see the characteristic time distributions in the insets in Fig. 6).
Figure 6: The ACFs obtained at the wavelength of 497 nm for four droplets of SiO$_2$ nanoparticles dispersions at the final evolution stage – (nearly) dry microaggregates. The left panel corresponds to DEG as a dispersion medium, while the right panel corresponds to TetraEG. Curves (1, 3) correspond to nanoparticle diameter of 100 nm, while (2, 4) correspond 450 nm diameter. The insets (1$'$, 3$'$) and (2$'$, 4$'$) show the characteristic time distributions for each of the four functions respectively.

Naturally, the rotational Brownian motion of a non-spherical aggregate should be considered as a source of the observed ACF properties. There is a lot of theoretical research on this topic (see e.g. [43, 44, 45]). Usually however it is applied to the liquid medium. For example, the rotational Brownian motion of isolated micrometre-sized ellipsoidal particles in water was experimentally studied by Han et al. [46] in the framework of 2D random walk trajectories. Application to gaseous medium – droplets trapped with electric fields in air – was implemented experimentally by Krieger et al. [13]. The authors claim that in their experiment the characteristic time of rotational Brownian motion was on a time scale of seconds. We estimate that in our experiment it was on a time scale of tens of milliseconds, as visualised in the real-time, 50 fps movie of a dry aggregate [47]. The distributions of characteristic times corresponding to different dispersions (1$'$, 2$'$, 3$'$, 4$'$ in Fig. 6) show peeks at large values (10$^{-1}$ – 100 ms), while displaying a significant variation in this region. The characteristic time of rotational Brownian motion calculated with Einstein–Smoluchowski relation with the rotational frictional drag coefficient for a sphere at viscosities comparable to that of air seems to be in agreement with these results. It must be kept in mind that microaggregates are not (perfectly) spherical and their surface is highly structured.

Based on the in-focus images (see the movie [47] and snapshots in panels
(b) – (d) in Fig. 4), it can also be inferred that the random rotations of the microaggregate in the electrodynamic trap induce/facilitate changes of its shape. We haven’t noticed any dependence of the rotational Brownian motion of aggregates upon the AC frequency. Thus we do not expect that they are significantly assisted by the AC field. We expect that ACFs corresponding to different levitating aggregates (Fig. 5 and 6) reflect primarily the changes in the distribution of mass and charge in the aggregate and its random rotation rather than the parameters of the original dispersion.

4. Conclusions

We used the dynamic light scattering technique adapted to study single levitating microdroplets of dispersion. Three laser wavelengths, different sizes of silica and titania nanoparticles and their concentrations in different dispersion media (liquids) were used. We registered ACFs at the evaporation stage of microdroplets and at the final stage of evolution corresponding to microaggregates.

At the initial stage of the microdroplet temporal evolution it seems possible to find the characteristic times corresponding to the Brownian motion of the dispersed nanoparticles, though the optical morphology resonances of a dielectric sphere manifest strongly as periodic changes of the scattered light intensity.

At the final stage of the droplet evaporation, the formation of non-rigid aggregates of nanoparticles is confirmed by the in-focus images. We obtained the ACFs of the aggregates using different sizes of silica and titania spherical nanoparticles in water, DEG and TetraEG. The autocorrelation functions measured with the DLS technique for microdroplets exhibit exponential decay. However, the temporal dependencies of these ACFs are significantly different from those found for the nanoparticles dispersion in bulk. It follows from the rotational Brownian motion and shape changes of the final non-rigid aggregates in the AC electric field of the trap.

The analysis showed that the dynamics of a levitated droplet of dispersion is rather complex and, apart from ACF, it requires simultaneous tracking of additional parameters such as size, weight, number of nanoparticles, electric charge (distribution), etc. The difficulties encountered in experiments on single levitating droplets and analysing the obtained results call for further detailed research. Also, the theoretical basis for dynamic light scattering on a single microdroplet awaits an additional consideration.
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• Autocorrelation function reflects the evolution of the microdroplet of dispersion
• The rotational Brownian motion of the aggregate in the trap can be detected
• The autocorrelation functions are different from those for the dispersion in bulk
CRediT author statement

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: