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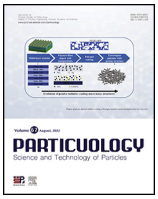

A new method for precise optical measurements of the sub-micron height of levitation of droplet clusters

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ABSTRACT

Droplet clusters levitating over the locally heated water surface are considered a very promising phenomenon of microfluidics for the potential use of the droplets as unique microreactors for microbiological experiments. A new optical method is suggested for precise measurements of the sub-micron levitation height of single droplets. The method is based on the analysis of variable colors of the interference halo around the droplet. For the first time, it is possible to measure the extremely low downward velocity of droplets, which grow due to steam condensation. This velocity was found to be 5–8 nm/s. The height of the levitation of various droplets just before their coalescence with a layer of water was also determined. Such measurements might be used for the verification of sophisticated models for the droplet levitation to be developed for a very thin layer of humid air under the droplet when the Knudsen effect should be taken into account.

1. Introduction

A new method for measuring small vertical displacements of droplets near the surface of a water layer is applied by the authors to study the behavior of the so-called levitating droplet clusters. It makes sense to first briefly explain how these droplet clusters are formed and look like. Small droplets can be easily observed above the surface of hot water. These droplets are formed by condensation of water vapor on sub-micron particles of dust suspended in air and the droplets usually move chaotically near the water surface (Ienna et al., 2012; Schaefer, 1971). It turns out that local heating of the water surface produces a relatively stable regular structure of much larger droplets, tens of microns in diameter. This self-assembled group of droplets is called a droplet cluster. Conditions and mechanisms of the formation of various droplet clusters are described in (Fedorets et al., 2002).

The lifetime of a droplet cluster is usually about 1 min. The size of the levitating droplets increases as a result of continuous condensation of vapor from the humid air rising from the surface of the evaporating water layer. The growing droplets descend lower and finally coalesce with a layer of water. However, the use of water droplets in chemical and microbiological laboratory research requires droplets of a constant size, steadily levitating for long periods of time. In (Dombrovsky et al., 2016) external infrared heating was proposed to suppress the condensation and stabilize the cluster. Somewhat later, a method for generating clusters consisting of a given number of nearly identical droplets was found, which made it possible to consider applications of droplet clusters in microbiological experiments (Fedorets et al., 2019). Interest in such experiments is related, in particular, to the known effect of significant acceleration of organic chemical reactions in small droplets (Wei et al., 2020; Zhang et al., 2021). A brief overview of twenty years of research on droplet clusters can be found in a recent article (Fedorets & Dombrovsky, 2023). The main results concerning levitating droplet clusters are detailed in a recent monograph (Fedorets, Dombrovsky, et al., 2023).

However, some features of the behavior of droplet clusters are poorly described theoretically. Therefore, more accurate measurements of the levitation height of large water droplets would be very useful to continue the research. Photographs of the cluster along the water surface clearly show reflections of the droplets in the

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Nomenclature

d	Droplet diameter
N	Frame number
T	Temperature
t	Time
u	Velocity
Greek symbols	
Δ	Path difference
δ	Height of levitation
λ	Wavelength of light
$\Delta\lambda$	RMS deviation of λ
Subscript	
surf	Water surface

water layer, which allows one to measure the height of levitation of the droplets (Fedorets et al., 2020; Zaitsev et al., 2021). The accuracy of such measurements is limited by the capabilities of optical microscopes. In our experiments, the image pixel size was about 0.6 μm which is insufficient to measure the low levitation heights of large droplets. In the present work, a new method of precise measurements is proposed, which is proven to be effective for such important measurements.

2. Experiments

We used the experimental setup described in (Fedorets et al., 2022, 2023b). The details of the experimental procedure can be also found in these papers. The cluster was formed over a layer of pure water with an admixture of a surfactant which suppresses thermocapillary flows. The water layer was locally heated by a laser beam from below through an opaque solid substrate. Low-power sources of infrared radiation were used to stabilize the cluster. Experiments have shown that when a cluster is illuminated by white light, thin color halos can appear around the droplet images. This phenomenon is observed only at low levitation heights of the cluster, and the color of the halo changes as the distance of the droplets to the water surface decreases.

The diameter of the droplets is almost two orders of magnitude larger than the wavelength of visible light, but the levitation height, understood as the distance between the lowest point of the droplet and the water layer, can be comparable to the light wavelength. A very thin gap between the levitating cluster droplets and the water layer is responsible for the halo. As in interference in thin films, the maximum light intensity is observed at the following values of the path difference of the light reflected from the bottom surface of the droplet and from the surface of the water layer (Born & Wolf, 2020):

$$\Delta = 2\delta = m\lambda, m = 1, 2, 3 \quad (1)$$

where δ is the droplet levitation height and λ is the wavelength of the halo light. Eq. (1) is written for the normal incidence of the white light beam.

It is known that when illuminating a transparent spherical particle, light is not only reflected and refracted, but one observes rather complicated wave effects described by the known Mie scattering theory, which gives an exact analytical solution for the scattering problem (Bohren & Huffman, 1998; Van de Hulst, 1981). In particular, the so-called surface waves propagate along the particle surface. Calculations show that in some cases this effect should be taken into account even when determining the integral characteristics of light absorption and scattering by particles (Dombrovsky, 1974, 1996; Irvine, 1965; Liou et al., 2010). Usually, interference of surface waves and diffracted radiation also takes place. In the considered problem, the light reflected from the water layer propagates from the lower surface of a droplet to its side surface and forms the observed halo. The width of the halo ring is comparable to the wavelength, but the microscope used makes the halo quite distinguishable.

The exact solution for the problem of interference of surface waves, when a droplet of water is illuminated from two sides (by a light source from above and by light passing through the droplet and reflected from the surface of the water layer), is a much more complicated task than the classical Mie solution. Note that scattering problems similar to the considered one have been solved for particles on the surface of a medium (Moreno et al., 2007), as well as for closely placed particles of material with arbitrary optical properties (Mishchenko et al., 1995). However, in the literature known to the authors, there is no solution for our specific problem. Fortunately, to interpret a periodical change of color of a droplet halo it is not necessary to solve such a complicated problem, because we are interested not in the intensity of a halo glow or in the polarization of the halo light, but only in the periodicity of appearance of the same color when the droplet levitation height changes. Therefore, a simple formula (1) is sufficient for our study.

It should be noted that optical interference is used to solve problems similar to the one considered in this paper. In particular, in (Barton et al., 2012; Celestini & Kirstetter, 2012) special schemes of interference measurements of the vapor gap thickness at Leidenfrost levitation of a single droplet were developed. In contrast to the more sophisticated measurements of (Barton et al., 2012; Celestini & Kirstetter, 2012), we do not use additional equipment and consider only the colors of the halos of the continuously descending droplets of the cluster. Of course, we cannot determine the current absolute value of the levitation height, but the change in the halo color of droplets as they move towards the substrate water layer enables us to determine the droplet velocity.

It is also important that a droplet cluster consists of nearly identical droplets and the difference between the colors of the halos of these droplets makes it possible to estimate the difference in the levitation height of droplets of an almost flat cluster.

In other words, the relatively simple interference method of this paper provides useful information on the gradual decrease of a drop cluster of nearly identical water droplets. As for the absolute height of the droplet levitation during the whole process (up to the coalescence of the cluster with the water layer), it can be obtained only approximately, based on the fact that the luminous halo becomes colorless when the droplet approaches the water layer at a distance smaller than an average wavelength of visible light.

A computer code used in regular experiments to process the images of droplets was modified according to a method suggested in (Afanasiev et al., 2015; Glassner, 1989) to analyze the halo color. The wavelength $\lambda = 580$ nm was assigned to white (colorless) pixels, whereas the wavelength of interest was determined as the average of the twelve brightest colored pixels of the halo image. Analysis of data from four radial cross-sections of the droplet image (the orientation of the cross-sections is shown in Fig. 1(c), with three pixels each in the vicinity of the luminance maximum) allowed for significant smoothing of the inevitable variations in the determined halo color associated with color noise. Experiments with clusters of a small number of droplets were carried out at laser power providing surface temperature of the water layer of $55 \le T_{\text{surf}} \le 70$ °C with an error of ±1 °C. The resulting diameter of the cluster droplets varied from $d = 15$ μm to 60 μm.

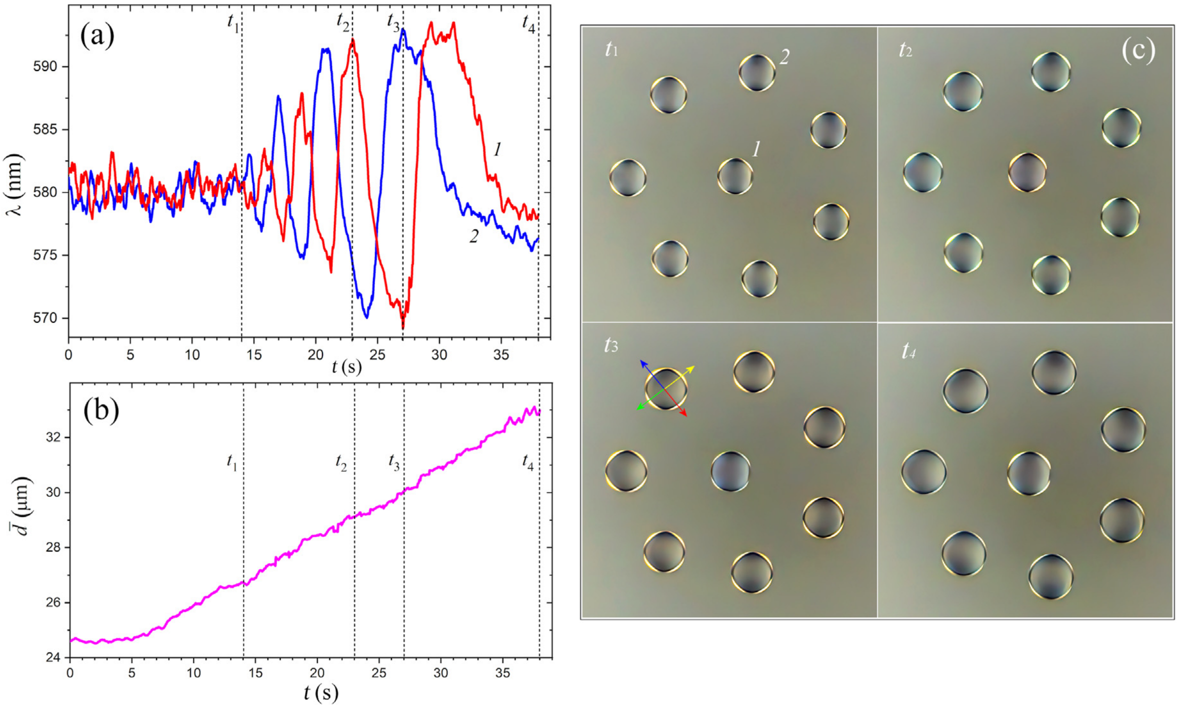

Fig. 1. Changes in the color of the halo around the droplets of the levitating cluster as the droplets grow and approach the water surface: (a) wavelength of the colored halo of the droplets 1 and 2, (b) average diameter of the cluster droplets, (c) cluster images at characteristic times, shown in panels (a) and (b) (the scale of the four images is the same, the lack of a scale bar is compensated for by the data in panel (b)). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

To observe the slow growth of the droplets due to condensation, we started with an equilibrium cluster obtained by prolonged infrared heating (Dombrovsky et al., 2020). Once the infrared heating was started, the infrared heating was switched off and the droplets began to grow. Some of the results obtained are shown in Fig. 1. The halos of the equilibrium cluster droplets (at $t < t_1$) were colorless as the droplets levitated at high altitudes. The difference between the curves in Fig. 1(a) for the central and peripheral droplets increased with time, and a few seconds before the cluster coalesced with the water layer, when the levitation height was less than the emission wavelength, the halos of all droplets became colorless again.

To estimate the error in determining the wavelength of the halo and the associated droplet levitation height it is convenient to consider an equilibrium cluster (at $t < 5$ s, Fig. 1) when the droplet size, droplet levitation height and halo color do not change with time. Fig. 2 shows the root-mean-square (RMS) deviation of $\Delta\lambda$ for a set of successive frames of video recording (N is the frame number). In our case, the standard deviation (SD) is equal to 0.54 nm, i.e. with a confidence interval (CI) of 0.90 the halo wavelength error is ±0.9 nm. This value is very small and has practically no effect on the accuracy of the results obtained.

The difference in the color change of the halos of the central and peripheral droplets in the cluster is a result of the slightly different sizes of these droplets. The central droplet surrounded by other droplets receives less vapor and therefore its size does not increase as rapidly. It is interesting that the maximum difference of diameters of droplets 1 and 2 found is less than 1 μm, i.e. it is at the limit of possibilities of optical microscopy with a pixel size of about 0.6 μm. Note, that at the time about t_3 the height of levitation of these droplets differs only by 23 nm. Interestingly, for clusters of more droplets, a regular difference in color of the halos around the droplets is observed (Fig. 3). This result further confirms the exceptionally high sensitivity of the proposed interference method to determine the levitation height of droplets. Note that with a colored halo, the entire droplet also takes on a hue of the same color. This does not help or hinder measurements but makes the image of the droplet cluster even clearer and more appealing.

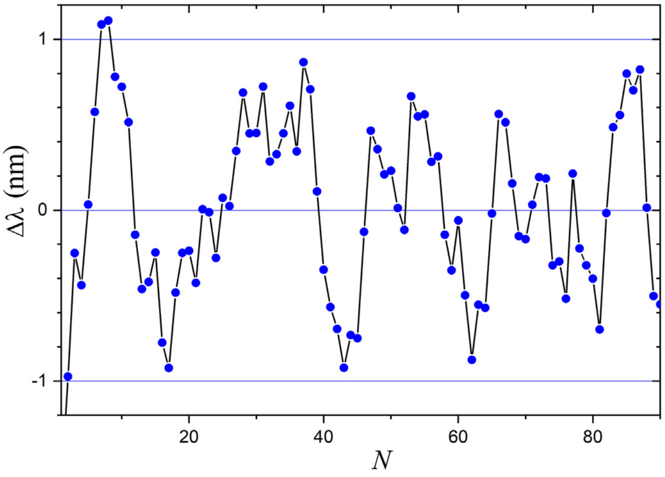

Fig. 2. The root-mean-square deviation of the measured wavelength of light for the stabilized droplet cluster.

Fig. 4(a) shows the halo color curves at different temperatures of the water layer. The maximum diameter of the droplets at $T_{\text{surf}} = 55$ °C was 25 μm and at $T_{\text{surf}} = 70$ °C was 60 μm. Even with this significant difference in diameters, the corresponding curves in Fig. 4(a) are similar to each other. The result obtained is explained by the fact that the color of the halo does not depend on the droplet size but is determined by the height of its levitation. The change in color of the halo over time makes it possible to determine the downward velocity of the droplet (see Fig. 4(b)). The change in the

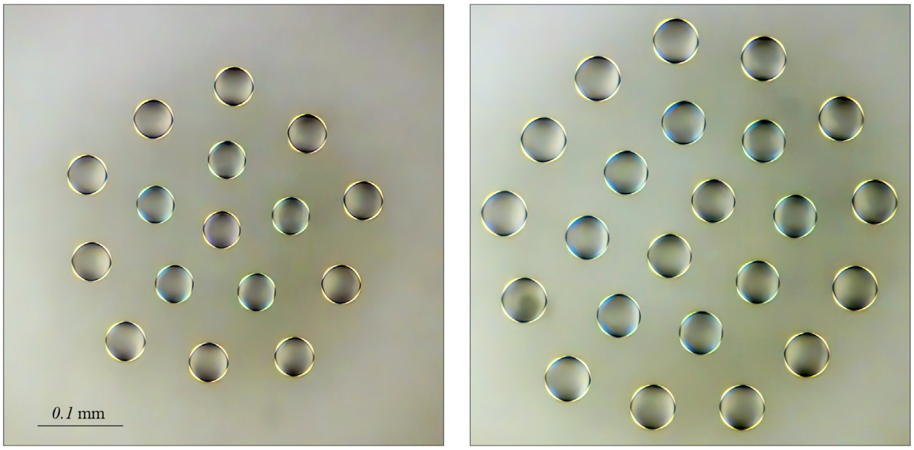

Fig. 3. Typical color halos at the droplets of stabilized clusters. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

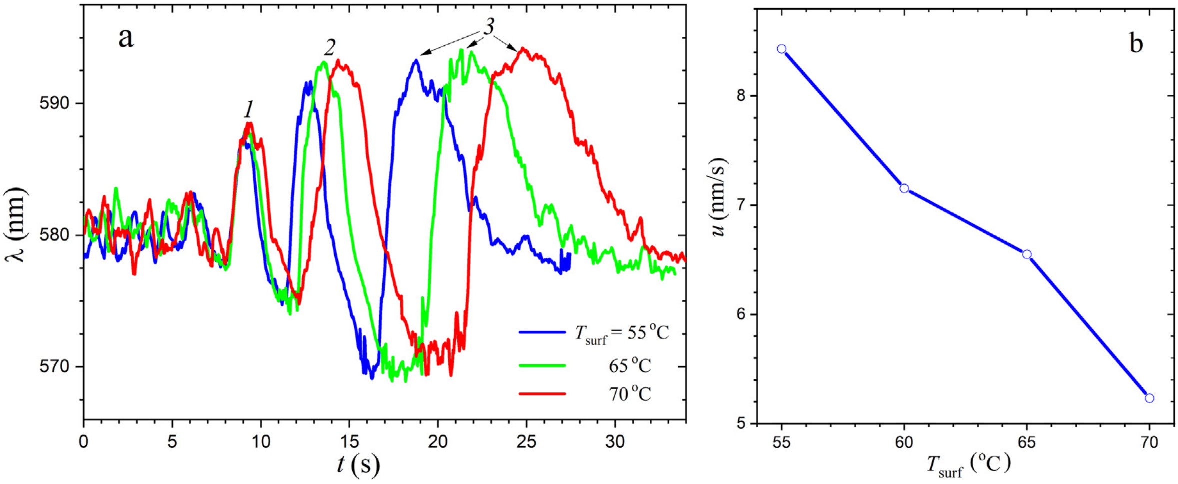

Fig. 4. (a) Color change of the droplet halo over time; (b) Water surface temperature's effect on the droplet's descending velocity. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

wavelength between local maxima 1 and 3 showed that this velocity decreases approximately linearly from $u = 8.4$ nm/s to 5.2 nm/s as the temperature increases from $T_{\mathrm{surf}} = 55$ °C to 70 °C.

It is also interesting to consider the comparably fast downward motion of droplets. In a special experiment, even before the droplet sizes of the stabilized cluster were equalized, the laser heating power was drastically reduced. At the same time, the droplets practically fall down. The rapid change in color of the halos observed in Video 1 (Shimmering droplets of a falling cluster) is discernible by the tenfold slowing down of the video playback.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.partic.2023.08.005

The color change of the halos when a cluster containing droplets with diameters from $d_1 \approx 45$ μm to $d_1 \approx 37$ μm falls is shown in Fig. 5. At the moment of switching off the laser heating, the large droplet 1 was slightly lower than droplet 2 (its halo was already colored). The first color maximum of the large droplet was ahead of the corresponding maximum of the small droplet by about 0.04 s. In 0.12 s, the large droplet passes through two color maxima and its

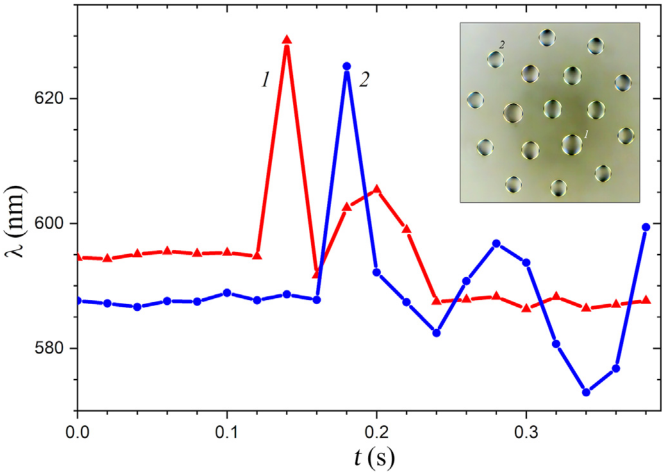

Fig. 5. Changing the colored halo of the droplets 1 and 2 as the cluster falls.

halo becomes colorless about 0.14 s before the cluster collapses. The color of the halo of small droplet changes several times before the collapse of the cluster.

The droplet falling velocity is found to be approximately the same for droplets of initially different sizes: 1.8 ± 0.5 μm/s, which is about 300 times greater than the velocity of slowly growing droplets. If the minimum levitation height of a large droplet is neglected, the initial (just before switching off the laser heating) levitation height of this droplet is found to be 104 nm and the corresponding value for the second droplet is 145 nm. These values do not contradict images of a side view of the cluster, in which even at maximum microscope magnification the levitation height of the droplets does not exceed one pixel. The suggested interference method is another example of the use of optical methods in microfluidics (Choi et al., 2016; Zhou et al., 2022). The measurements carried out gave new data for the low height of levitation of droplets as well as for the downward velocity of growing microdroplets.

The motion of droplets near the water surface cannot be calculated in the usual way when the gas flow is considered in the continuum approximation and the Knudsen lifting force is ignored (Joung, 2011; Kelling & Wurm, 2009; Roy et al., 2022). The experiments with the use of the suggested optical method are expected to be important for the verification of theoretical models for the levitation of droplets at a small distance from the water surface.

3. Conclusions

A new method for precise optical measurements of the submicron levitation height of droplet clusters over the water surface was suggested. The method is based on the analysis of the interference halo around a single droplet. The extremely low downward velocity of droplets, which grow due to steam condensation was measured for the first time. At the surface temperature of the water layer from 55 °C to 70 °C, this velocity was found to decrease from about 8 nm/s to 5 nm/s. The height of the levitation of various droplets just before their coalescence with a layer of water was also determined. This value is too small to be directly measured with the use of an optical stereomicroscope. The authors believe that the results obtained will be useful for the verification of advanced theoretical models for droplet levitation in a specific case when the Knudsen effect should be taken into account.

Declaration of competing interest

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.

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