Thermal conditions for the formation of self-assembled cluster of droplets over the water surface

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Abstract. The effect of temperature profile of the water layer surface on the formation and structure of a levitating droplet cluster is studied in the paper. The laboratory experiments indicate that a local temperature maximum of water is a necessary condition for the formation of a cluster. A quantitative criterium of transformation of a monolayer of randomly positioned microdroplets to a self-assembled cluster of relatively large droplets is obtained.

1. Introduction

A layer of microdroplets over a uniformly heated surface of water \cite{1} and a droplet cluster \cite{2} levitating in an air-vapor flow over a locally heated area of the surface differ significantly in their dynamics and structure. In the first case, the droplets are positioned randomly, while in the cluster the droplets are larger and they form a regular structure. The ordering of the droplets can be described by the so-called Voronoi entropy. With fixed maximum water temperature and ambient air properties, the condition for the formation of a droplet cluster is a small area of a hot spot of the water surface. The effect of this area is studied in the paper. The critical condition is determined under which a droplet cluster is observed.

2. Laboratory set-up and experimental procedure

The traditional schematic of the experiment \cite{3, 4} is presented in figure 1. A droplet cluster (1) is formed over a locally heated area of a layer of distilled water (2) containing the surfactant impurity (sodium dodecylsulfate at a concentration of 0.5 g/L \cite{5}). A sitall substrate 400 μm thick (3) was glued to the metal bottom of the cuvette. The cuvette body has channels (4) connected to the external circuit of the CC 805 cryothermostat (Huber, Germany), which allows stabilizing the temperature of water layer in the range of \( 9 \leq T_0 \leq 60 \)°C. The water layer is also locally heated by a laser beam (5) directed to the lower blackened surface of the substrate. The semiconductor laser BrixX® 808-800HP manufactured by Omicron Laserage (Germany) is used. A displacement of the defocusing (concave) lens (6) made it...
possible to vary the diameter of the heating spot. In all experiments, the thickness of water layer was equal to 400 ± 2 μm. The axisymmetric temperature field of the water surface was recorded with an A655sc thermal imager (FLIR, USA). Video recording of the cluster was carried out using an AXIO Zoom.V16 stereomicroscope (Zeiss, Germany) equipped with a PCO.EDGE 5.5C high-speed camera (PCO, Germany).

3. Experimental results

The parameter $K = T_{1/2}/R_{1/2}$ (where $T_{1/2} = (T_{\text{max}} - T_0)/2$, $R_{1/2}$ is shown in figure 1a) was used as the characteristic of the temperature profile (see figure 1a) in the analysis of experimental results. Figure 1b shows a typical image of a cluster with automatically recognized droplets. The experimental study included two stages described below.

3.1. Transition from a monolayer of droplets to a droplet cluster

It is known that a monolayer of very small droplets condensed from the surrounding humid air can be formed near a hot water surface [1, 6]. In this case, the effects typical of a droplet cluster [7] are not observed: the self-assembly of an ordered structure, condensation growth of droplets, as well as displacement of the cluster as a whole while maintaining its structure. The fundamental importance of local heating for the formation of a cluster is well known [7–9], but the transition from a monolayer of chaotically moving droplets to a cluster remains insufficiently studied.

In a series of experiments, the parameter $K$ decreased due to an increase in the thermostating temperature $T_0$ at a constant temperature $T_{\text{max}} = 60^\circ\text{C}$. In each heating mode, the time required for cluster formation was maintained. Due to the limited volume of the paper, figure 2 shows only two typical images of clusters and the corresponding Voronoi tessellation illustrating the ordering of droplets in the cluster. The transition from a monolayer of droplets to a cluster was observed in the range of $1 < K < 2.5$ K/mm, after which the Voronoi entropy, $S$, decreased linearly with the parameter $K$ (figure 3). Ordering of the cluster also may be quantified with the continuous measure of symmetry [10].

3.2. Effect of the hot spot area on the density of droplets in a cluster

The experimental set-up enabled us to change parameter $K$ at constant temperatures $T_0$ and $T_{\text{max}}$. For this, vertical displacements of the lens were used (see figure 1a), and a small change in $T_{\text{max}}$ was compensated by adjusting the laser heating power. In the series of experiments at $T_0 = 29 \pm 0.2$ °C and $T_{\text{max}} = 70 \pm 0.2$ °C, five different lens positions were used, corresponding to the range of $15.5 \leq K \leq 16.6$ K/mm, when water droplets form a classical hexagonal cluster. The images of the cluster were processed by a special program that allows measuring the position and diameters of the droplets, as well
as calculating the average values of the diameter $D$ and the distance $L$ between the centers of neighboring droplets in the selected group. The averaging was performed over a group of seven droplets in the central region of the cluster (these droplets are marked by a red ring in figure 1b). The ratio of $L/D$ was used to evaluate the density of droplets in a cluster.

![Figure 2](image)

**Figure 2.** Evolution of droplet cluster with the parameter $K$: a, b – $K = 17.2$ K/mm, c, d – 2.9 K/mm; a, c – images of clusters, b, d – Voronoi tessellations.

The results for clusters with the number of droplets from 60 to 80 and the average diameter of central droplets from 31 to 33 μm from two video recordings for each cluster are shown in figure 4. Obviously, the density of droplets in a cluster is sensitive to changes in $K$ even in the case of a constant temperature of the water surface under the cluster. As one can expect, a decrease in the area of the heating spot leads to an increase in the gas flow rate and to a decrease in the density of droplets in the cluster.

![Figure 3](image)

**Figure 3.** Effect of local heating of water surface on ordering the levitating droplets.
4. Conclusions

The effect of temperature profile of the water layer surface on the formation and structure of a droplet cluster has been experimentally studied. It was shown that the local temperature maximum is a necessary condition for the formation of a self-assembled cluster. The Voronoi entropy, which characterizes the degree of ordering of the hexagonal structure of the cluster, increases in proportion to the temperature difference between the center of the hot spot and the periphery of the water layer. The laboratory experiments show that the distance between the droplets of the cluster increases with a decrease in the area of the hot spot at the water surface. The results obtained are important for understanding and further practical use the droplet clusters as promising micro-reactors for bio-chemical studies.

Acknowledgements

The authors gratefully acknowledge the Russian Science Foundation (project 19-19-00076) for the financial support of this work.

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