Laser-induced jetting and controlled droplet formation

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Abstract: The article reports, in the general context of developing techniques to generate microjets, nanojets and even individual nanodroplets, a new method to obtain such formations by interaction of a single laser pulse at 532 nm with an individual/single mother droplet in pendant position in open air. The beam energy per pulse is varied between 0.25 and 1 mJ, the focus diameter is 90 μm, and the droplet's volumes are either 3 μl or 3.5 μl. Droplet's shape evolution and jet emission at impact with laser pulse was visualised with a high speed camera working at 10 kfps. Reproducible jets and/or separated microdroplets and nanodroplets are obtained which shows potential for applications in particular in jet printing. It is demonstrated that it becomes possible to play with the geometrical symmetry of both laser excitation and liquid in order to manage the number and the orientation of an induced microjet and consequently to actuate the orientation and the production of nanodroplets by light.

1 Introduction

Generating microjets and droplet formation by interaction of laser beams with a liquid layer is currently a subject of outmost interest due to both basic research on laser-fluid interaction [1], [2] and appealing printing applications based on wall-free direct-write strategies [3]. Based on recent experiments [4], the present article aims at generalizing laser-induced micro-jets from classical flat liquid layers to finite size objects of spherical symmetry by using instead suspended microdroplets. The reported experiments are conceived to use the unresonant interaction of pulsed laser beams with droplets hanging in air in open atmosphere, without particular protection with respect to room atmospheric variations, in order to generate microjets and nanodrops. The dimensions and propagation speeds of these newly produced "objects" depend on beam's energy, focus shape and position with respect to droplet's surface or center of symmetry, as well as droplet volume and shape.

Interaction is always produced and effects are measured for one single droplet on which one single laser pulse is directed, when it is focused on the droplet's input surface, or in its symmetry center, or even on its output surface.

The results show that although the optical alignment of the experiment is quite critical, a good reproducibility of effects is obtained from pulse to pulse and droplet to droplet, provided the droplet's volume/dimensions are the same and the focus position and dimensions are kept unchanged. This is in agreement with data reported in [5] and [6].

This investigation, developed during the lifetime of COST MP1205 network, illustrates the interplay between symmetry and focusing conditions.

2 Experimental set-up

The general structure and characteristics of the experimental set-up is shown in [4] were water droplets are generated in pendant position in open air using a computer controlled droplet generator dual syringe system. The resolution of the system is 0.05% out of the utilized syringe volume, i.e. 2.5 nl), for a 50 μl total syringe volume; the accuracy in generating a given volume is ±1 % (when droplet volume is 10% of syringe volume) and precision is ±0.2 % (indicating variation of generated volume from one droplet to another). The capillary material is hydrophobic so that droplet is tangent to capillary tip and does not wet the capillary outer surface. The evolution of the droplet

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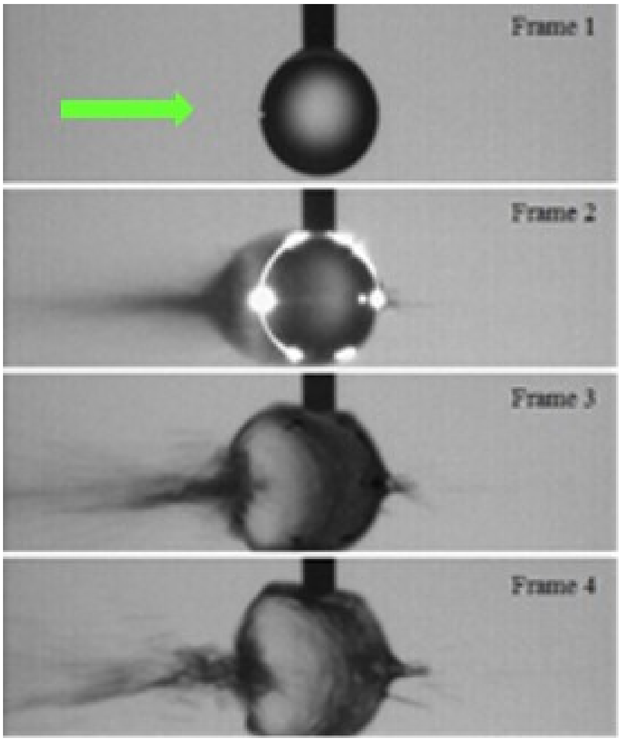

Figure 1: Recording of effects produced on droplet by laser beam sent in the equatorial plane and focused on its front face. Droplet volume V_2 = 3.5 µl; beam energy E_4 = 1 mJ, frame rate 10 kfps. The green arrow indicates laser beam incidence.

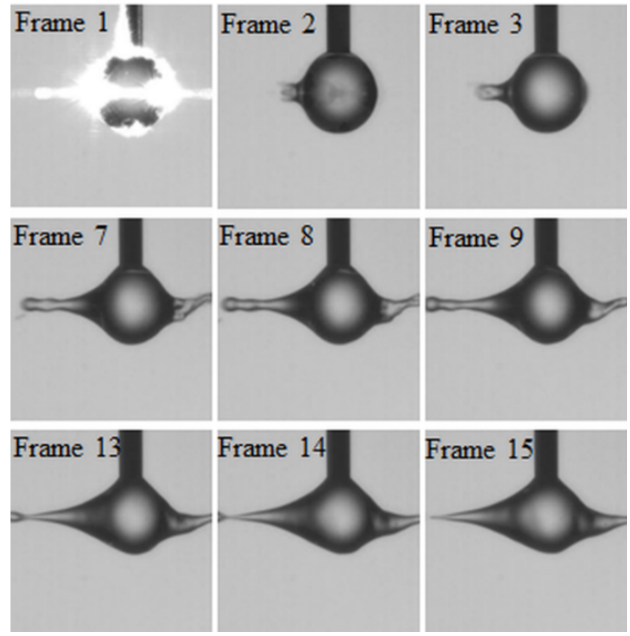

Figure 2: Directional jetting and droplet generation. Laser beam sent in droplet equatorial plane and focused on front face. Energy is E_1 = 0.25 mJ; the sequence shows frames as follows: Frame 1 contains data harvested during the first 10 µs after impact and in each frame starting with Frame 2 events for 100 µs are recorded, so that in Frame 15 data between 1410 µs and 1510 μs are displayed. Droplet volume V_1 = 3 µl; frame rate 10 kfps.

shape was recorded in real time using a high-speed camera working at 10 kfps and further characterized with a drop shape analysis method. Droplets with two volumes, 3 and 3.5 µl, labeled V_1 and V_2, respectively, were generated [4, 7]. Laser pulses of 6 ns full-width half-maximum were emitted at 532 nm, with the average energies per pulse: E_1 = 0.25 mJ, E_2 = 0.4 mJ, E_3 = 0.7 mJ or E_4 = 1 mJ monitored pulse by pulse. The beam was focused at different positions on droplet interface or within its volume, by moving it with respect to focus point; the laser beam diameter at focus is 90 µm, i.e. quite small compared to mean drop radius which is 900 µm. The precision of ±0.2 % in generating a droplet, implies a variation of droplet volume and, consequently, the radius and diameter of the droplet vary within certain limits that may influence the characteristics of the laser beam effect on droplet. As an example, for 3.5 µl, the droplet diameter is 0.9417 mm and the radius varies between 0.9411 mm and 0.9424 mm i.e. 1.3 µm. This is enough to explain variations of the effects of the laser beam on droplet by unresonant effects when the irradiation is made in the same optical arrangement conditions and the beam is focused on droplet/air interface.

3 Results

In Fig. 1 is presented a sequence of four frames recording the effects produced on droplet by a laser pulse sent in the equatorial plane and focused on its front face. For 3.5 µl and 1 mJ beam energy, aerosols (pico- and nano-volumetric droplets) are generated in the backward direction.

Frame 2 shows the superposition of images recorded at laser pulse impact with the droplet and mechanical effects that occur at much later time. The halo around droplet is due to reflection and guiding of 532 nm beam at the droplet interface [8, 9].

In Fig. 2 are also shown the results of laser beam interaction with a 3 µl droplet for a lower energy per pulse E_1 = 0.25 mJ. A water jet with nanodroplet formation at the tip is induced backward with respect to laser radiation propagation direction; the same happens with some delay in the forward direction at output from droplet. At later stage, surface tension forces jet recoil towards the droplet, leaving back a nanodroplet as a result of the Rayleigh-Plateau instability [10] and the subsequent breakup whose dynamics is clearly identified by the conical ends [11].

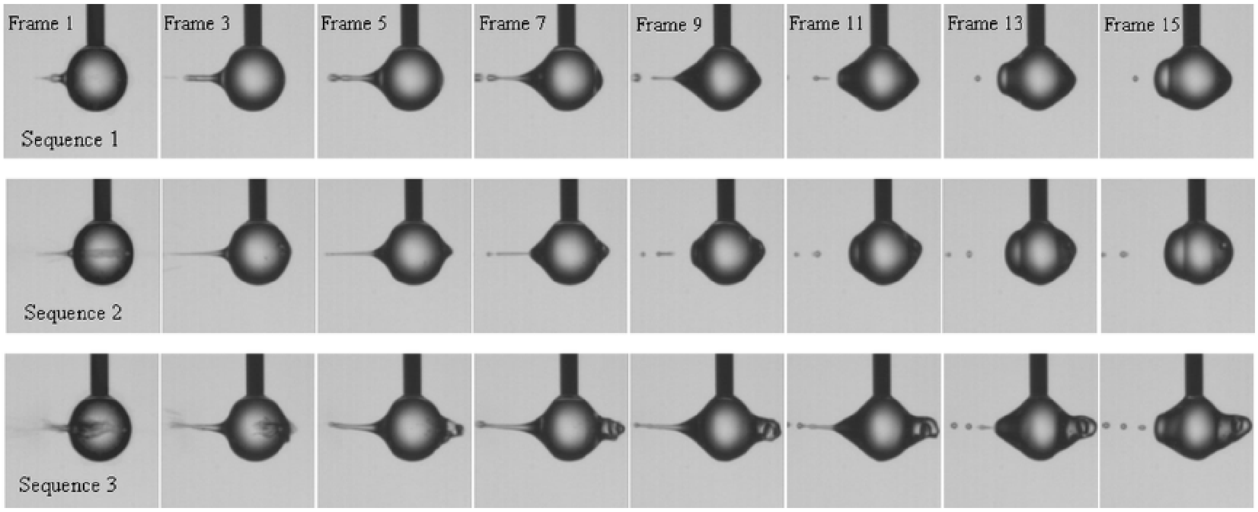

Figure 3: Backward generation of directional conical microjets emitting nanodroplets as a function of deepness of focus point on droplet front face. Each sequence is recorded for the same geometrical conditions but on different droplets. The variable deepness results from the reproducibility of the volume from droplet to droplet which is within ±0.2 % for expected volume $V_2 = 3.5$ µl. Sequences show the odd consecutive frames from 100 to 1500 µs obtained in the same experimental conditions. Pulse energy $E_1 = 0.25$ mJ; frame repetition rate 10 kfps.

Depending on the deepness of focus point into droplet after penetrating through the front face, laser beam may induce electrostriction [12–14] which leads to formation of well-defined bulge and jet (Fig. 3) formations; electrostriction, related to a local pressure increase at the center of the beam occurs for time scales smaller or around the time to propagate a sound wave over the droplet diameter, which here is 1 µs. Individual jets are generated with a conical pedestal that ends up with a cylinder of cross section of the order of a few tens of micrometers and breakup occurs at the tip due to the Rayleigh plateau instability [10]. A droplet with a diameter of few tens of micrometers is detached from the tip of the microjet whereas the remaining long cone-cylinder retracts to the mother drop due to surface tension effects (Fig. 3). This process may take from a few hundreds of µs to some ms [7].

Alternatively, in the case of laser beam focused in droplet geometrical centre, scattered light is angularly distributed around droplet (Fig. 4). Normally, Stimulated Brillouin scattering (SBS) takes place in bulk samples in the backward sense of the beam if the sample is long enough [15].

Fig. 4 illustrates the jetting morphology when both increasing the pulse energy and focusing the beam right at the center of the drop. Even if the jet at the incidence point is the fastest (Frame 3 in Fig. 4 which records events along 100 µs), the significant increase of electrostriction in the drop center produces, by symmetry, radial jets almost isotropically. Their final breakup offers this unique opportunity to produce nanodroplets in 4π directions (Frame 7).

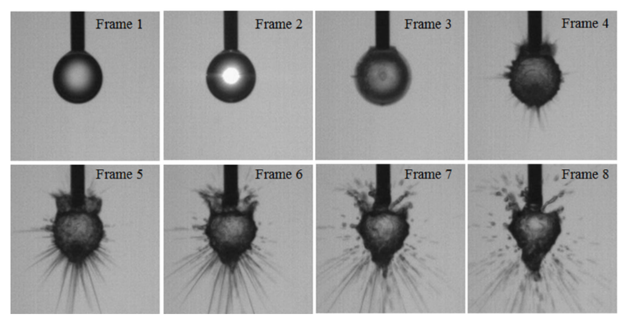

Figure 4: Multi-directional microjets generation. Laser beam is sent in droplet equatorial plane and focused in its geometrical centre. Sequence shows consecutive frames from 1 to 8 ending with the breakup into a nanodroplet assembly. Energy $E_4 = 1$ mJ; droplet volume $V_2 = 3.5$ µl; frame repetition rate 10 kfps.

As production and control of very small droplets remains an incredible challenge, in particular for printing inks, functional nanomaterial, or cells, the static and dynamic dimensions of nanodroplets/microjets were determined by comparing them with the overall diameter of the capillary tip (510 µm) used as a benchmark for calibrating the actual size of a pixel in the frames. Examples are illustrated in Fig. 5a and b. Alternatively, the velocity of a microjet is calculated by determining the propagation length during a frame exposure from two consecutive frames, one used as reference as in Fig. 5c; with frames recording rate 10 kfps, two images are separated by 100 µs. Considering the experimental conditions, the jet formation is extremely fast as the tip velocity deduced from Fig. 5c is 25 m/s. Con-

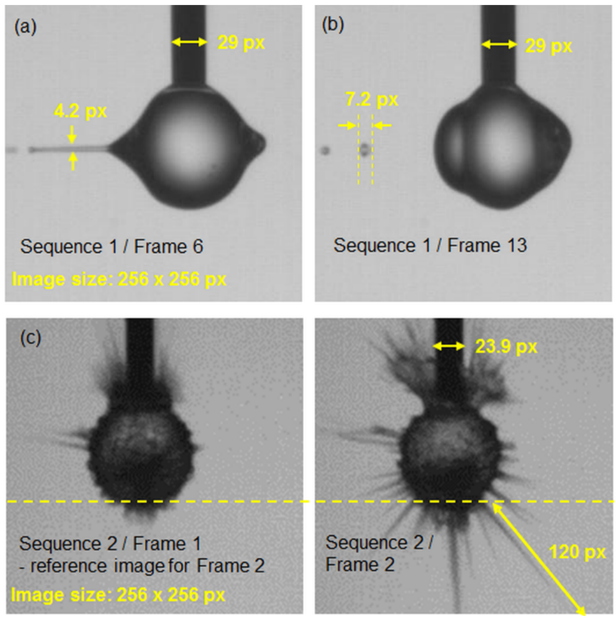

Figure 5: Dimensional characteristics of liquid structures ejected from droplet by a laser pulse sent in droplet's equatorial plane and focused on (a) – (b) its front face and (c – 2 Frames) in its centre. Pulse energy: E_1 = 0.25 mJ in (a) and (b); E_4 = 1 mJ in (c); droplet volume V_2 = 3.5 µl; frame repetition rate 10 kfps; dimensions are specified in pixel units.

sequently, jet diameter easily reaches about 70 µm (Fig. 5a) and the emitted droplets may have a diameter smaller than 60 µm (Fig. 5b) roughly corresponding to a resolution of 500 dpi.

The obtained emissions are reproducible from one droplet to another and from pulse to pulse, provided the experimental conditions are strictly reproducible, namely: the droplet's volume is generated with a precision of ±0.2 % (which means a reproducble shape of the droplet), the laser pulse energy is reproducible within ±1% and the beam structure is single mode and stable from one pulse to another [4].

4 Discussion

While light-induced forward transfer, the so-called LIFT [2], is performed in thin liquid layers or close to flat interfaces, this communication reports on light-induced jetting by suspended mother droplets in order to explore the effect of symmetry in the generation of microjets and nanodrops. On one hand, at low fluence and/or when focusing is set at the front interface of the mother drop, the axial symmetry of the incident laser beam governs the jetting. On the other hand, at large fluences and for beam focusing right at the center of the drop, the jetting pattern seems to be driven by the spherical symmetry of the mother drop, since interaction gives birth to a large set of radial microjets. As a first conclusion, it is demonstrated that it becomes possible to play with the geometrical symmetry of both laser excitation and liquid in order to manage the number and the orientation of induced microjets and consequently to eventually actuate the orientation and the production of nanodroplets by light. These conclusions need nonetheless to be densified using a larger recording frame rate in order (i) to set the right mechanism leading to jetting from ns pulses (bulk bubble nucleation and/or electrostriction as a function of the fluence) and (ii) to access to the early stage dynamics of the induced fluid flow which build the liquid jet.

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