Applied Surface Science 302 (2014) 303–308

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Film-free laser printing: Jetting dynamics analyzed through time-resolved imaging

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ARTICLE INFO

Article history:
Received 28 June 2013
Received in revised form
19 September 2013
Accepted 20 September 2013
Available online 1 October 2013

Keywords:
Laser printing
Laser-induced forward transfer
Cavitation bubble
Liquid jet
Time-resolved imaging

ABSTRACT

The film-free laser-based microprinting technique allows high-resolution printing of transparent liquids without the need for the preparation of the liquid in thin-film form. Its operating principle relies on the tight focusing of ultrashort laser pulses in the liquid free-surface proximity producing upon absorption a rapidly expanding cavitation bubble that generates the ejection of micrometric liquid jets.

While the technique proves feasible for microprinting, a deeper understanding of the influence on the printing process of its most relevant technological parameters is required. Therefore, in this work we analyze through time-resolved imaging the laser pulse energy influence on the bubble–jet dynamics of a film-free liquid ejection event. We simultaneously image the evolution of both cavitation bubble and ejected liquid, showing that for all the analyzed energies the transfer mechanism is mediated by the formation of two liquid jets which originate during the successive expansion-collapse cycles that the cavitation bubble undertakes close to the liquid free-surface. We find that the evolution of both bubble and jets depends strongly on the energy. The different bubble geometries that appear are interpreted in terms of the counter-jet interaction with the bubble, which in its turn depends on the energy.

1. Introduction

In recent years there has been an increasing interest in the digital manufacturing of small-scale patterns of diverse materials. Among the various techniques capable of printing high-quality micropatterns, laser-based direct-writing techniques stand out as excellent alternatives, offering high-resolution, fast speed, and contamination-free deposits, being in the same time capable of printing a large spectrum of liquid rheologies in a quite straightforward way [1]. Laser-induced forward transfer (LIFT) is probably the best-known laser-based printing technique, allowing a wide range of material printing [2–14] through a liquid transfer mechanism whose understanding has prompted considerable research [15–21]. Its great printing potential made the technique quite attractive for the industrial printing market, and this recently triggered the development of cost-effective LIFT processes capable of achieving remarkable high printing throughputs, both in terms of printing speed and printing area [22].

However, despite its printing versatility LIFT requires additional pre-printing steps, such as depositing the laser absorbing coating of the donor substrate, which is time-consuming, or preparing the liquid of interest in a thin-film form, which can result in a decrease of reproducibility, especially for high-resolution droplet printing [23–25].

Nevertheless, these difficulties can be sorted out through the use of film-free laser printing, a versatile yet simple printing method which has been demonstrated to be feasible for high-resolution liquid microprinting [23–26]. In this technique, a short pulse laser beam is tightly focused in the subsurface of a liquid that is transparent or weakly absorbant to the used laser radiation. Upon absorption, a laser-generated cavitation bubble is produced and rapidly expands close to the liquid free-surface, leading to the ejection of a high aspect ratio liquid jet that may contact a nearby substrate to generate the deposition of a droplet. The desired micropatterning is achieved by translating the substrate relative to the fixed laser-liquid container assembly [23–26].

The optimization of the performance of the technique requires, however, a deep understanding of the influence of the main technological parameters on the morphology of the printed features. In previous works [24,25] we demonstrated that given a fixed laser focusing depth there is a range of laser energies for which circular droplets are obtained, whose diameter shows a linear dependence with the laser pulse energy. In addition, time-resolved images of the evolution of the liquid surface taken during a transfer event revealed that liquid ejection is mediated by two different types of jets, and that the individual contribution of these jets to the finally deposited droplet depends on a well-defined energy threshold [24].

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0169-4332/$ – see front matter
http://dx.doi.org/10.1016/j.apsusc.2013.09.119

Although understanding the energy influence on the deposits was important from a technological point of view, it would be of great interest as well understanding that influence from a more fundamental perspective related to the bubble–jet interactions. In a previous work [27], we investigated such bubble–jet interplay for a single laser pulse energy through the simultaneous visualization of both cavitation bubble and liquid jet, and we showed that the two types of jets mediating the liquid transfer are strongly correlated with the dynamics of the laser-generated bubble that develops close to the free-surface of the liquid. In the present work we continue that study by extending it to the analysis of the influence of the laser pulse energy on the bubble–jet dynamics through the use of similar time-resolved imaging techniques. Such understanding may prove valuable not only for the optimization of the liquid transfer conditions, but it may also help elucidating more fundamental aspects related to the bubble–jet interactions.

2. Experimental

The experimental configuration has been described in detail elsewhere [27]. Briefly, it consists of two main parts: the film-free liquid printing setup and the time-resolved imaging setup.

The liquid microprinting setup consists of an ytterbium diode-pumped laser, an optical system and a container holding the liquid to be printed. The laser (1027 nm, 450 fs), is operated in single-shot mode for all the pulse energies used in this study (4, 8 and 14 μJ), and its beam follows a mirror-guided optical path toward a microscope objective (50×, 0.55 NA), whose purpose is to deliver the tightly focused laser pulse inside the liquid of interest and close to its free-surface. The liquid is a 50% (v/v) water and glycerol solution (1.1 g/cm^3 density, 6 mPa s viscosity, 67 mN/m surface tension), which completely fills a relatively small container fitted with thin, transparent walls that assure an optimum illumination inside the liquid bulk. This solution represents a simple and transparent model for many of the inks currently used in printing. The reservoir is supported on a xyz high precision positioning stage which makes possible the controlled focusing of the beam at the desired depth.

The time-resolved imaging setup is a stroboscopic shadowgraphy system that is aligned at grazing incidence relative to the edges of the container, being capable of imaging the relevant processes that take place around the liquid-air boundary. It is composed of a 18 ns pulse strobe lamp, a Köehler homogenizing system, and a 2 Mp CCD camera coupled to a microscope objective. Images of a single liquid transfer event at controlled delay times can be obtained by simply strobing the flash lamp over the area of interest in a synchronized way with both laser pulse and CCD camera. While the CCD integrates for 500 ms, the image is only exposed over the 18 ns strobe of the lamp, which sets the time resolution of the imaging system.

3. Results and discussion

Three complete stop-action movies showing transfer events generated under different energy conditions (4, 8, 14 μJ) are presented in Fig. 1. The images depict the most representative jetting situations that arise when the laser pulse energy is varied, with all the other controllable experimental parameters kept fixed. In all the frames, the laser beam is impinging downwards, from the ambient air toward the liquid bulk, being focused at a constant depth of about 80 μm below the liquid free-surface. Such working conditions would lead to the deposition of relatively large droplets in an eventual printing experiment, as the enlarged features significantly improve the overall time-resolved imaging visualization conditions. In addition, it should be noted that the dynamics is axisymmetrical, and therefore most of the out-of-axis elements in the images must be disregarded from the analysis, as they are most probably bubble remnants resulting from previous jetting events.

The overall view of the dynamics displayed in Fig. 1 shows that regardless of the initial laser pulse energy, liquid ejection follows a sequence of well-defined steps: cavitation bubble generation, rapid bubble expansion, thin-jet formation, bubble collapse and re-expansion, and finally, thick-jet formation. The same features were also observed in a previous work [27], where we identified the fundamental role of the cavitation bubble expansion-collapse cycle in the jet formation process. Essentially, it was found that the rapid expansion of the cavitation bubble induces the formation of a stagnation point at the bubble top-pole which drives the formation of two opposing jets: one propagating outwards (thin-jet), and one toward the liquid bulk (counter-jet), not visible in the images, but which strongly interacted with the bottom of the collapsing bubble. The interaction resulted in the split of the original bubble into two separate bubbles, with the former one acquiring a torus-like shape. The emergence of the initially cylindrical thick-jet was attributed to the subsequent re-expansion of the toroidal bubble close to the liquid surface.

Following this description, it is possible now to build an interpretation of the laser pulse energy influence on the observed bubble–jet dynamics. According to the images displayed in Fig. 1, the magnitude of bubble expansion clearly increases with laser pulse energy, which also raises the bubble capacity of displacing more liquid upwards. As the pressure increases in the bubble top-pole, the protrusion above the liquid free-surface gradually collapses from the sides, in a thinning process that leads to the formation of a thin-jet, whose aspect ratio and speed clearly increase with energy. It is, however, quite remarkable that for all the analyzed energies the jet always maintains a good stability in its propagation away from the liquid surface.

In all cases, the bubble collapses in a highly asymmetrical way, and as a result its top-pole gets severely flattened, as opposed to the lower pole, which maintains a quite round profile. The subsequent moments, however, show a completely different evolution that critically depends on the laser pulse energy. Thus, for the lowest energy (Fig. 1a), and in contrast with our previous results [27] as well as the other analyzed energies, the collapse proceeds without the emergence of any secondary bubble from the bottom pole of the collapsing cavity. This can be understood considering that an initially low energy pulse can only give rise to a relatively low overpressure zone in the bubble top-pole, and therefore, to a slow thin-jet and counter-jet pair. In this way, the inwards propagating counter-jet does not have sufficient kinetic energy to detach the secondary bubble at the possible impact. However, despite this lower energy, the overpressure is still strong enough to generate a net push on the bubble, which as a result slowly sinks toward the liquid bulk in a process that lasts until the final stages of the recorded dynamics. At around 15 μs, the bubble reaches its minimum dimensions, and starts to expand again up until 23 μs, after which it re-collapses while continuing to sink inside the liquid.

Increasing the laser pulse energy results in a dynamics analogous to the one described in Ref. [27]. In these cases (Figs. 1b and c), the counter-jet is sufficiently energetic to promote the detachment of a second, smaller bubble coming out of the bottom pole of the first one. Depending on the energy, the bubble may emerge very close to the moment of the complete collapse of the initial bubble (intermediate energy), or on the contrary quite early, during the first moments of bubble collapse, in which case a prolonged interaction is seen to last for almost 3–4 μs (highest energy). The two bubbles detach from one another, and continue to collapse until reaching their minimum size almost at the same time. Then they start to expand again, co-interacting much more strongly for the intermediate energy case, when they contact and even squeeze each other boundaries as they reach the maximum expansion size,