Supplementary Information

Thorough Small Angle X-ray Scattering analysis of the instability of liquid micro-jets in air

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1. Micrographs of the exit cross section of the stainless steel tubes producing jets

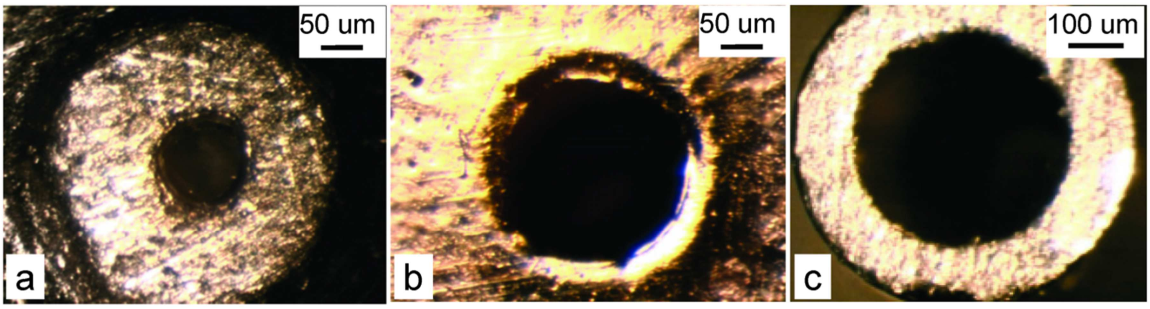

Figure S1. Optical microscope images of the exit opening of the three stainless steel tubes employed to produce free liquid jets in air. (a) tube with 100 µm diameter, (b) 250 µm diameter, (c) 450µm diameter.

2. SAXS pattern for region I: scanning across the width of the jet

The SAXS curves are relative to a water jet exiting from a circular nozzle with diameter 100 µm and flow rate 2.235 ml/min.

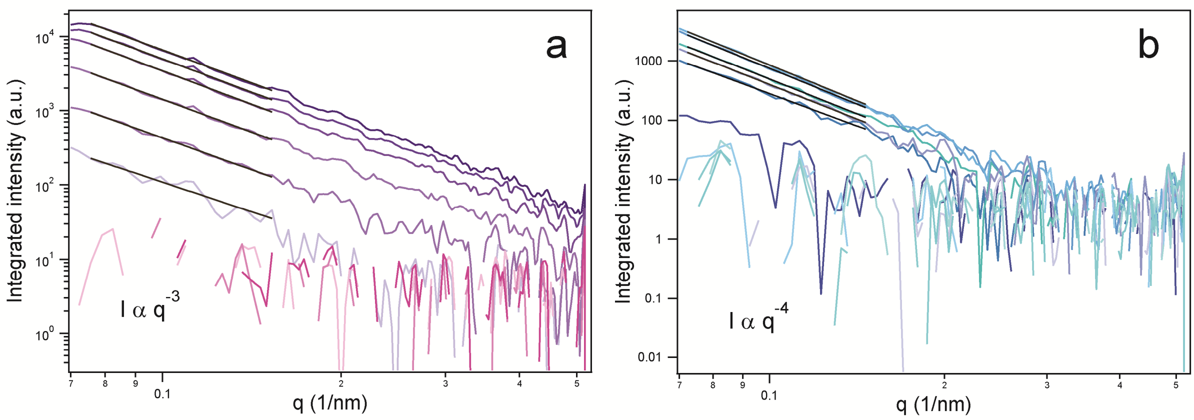

Figure S2. Intensity curves obtained with a horizontal scanning of the cylindrical jet corresponding to the position of peak 1 of figure 4 (a) and to the position of peak 7 of figure 4 (b), that is before (a) and after (b) the onset of the instability. The sampling distance of the horizontal scan is 200 µm. The curves have been fitted with a power law as explained in the main text of the article. It can be observed that, independently from the position across the jet, the fitting in figure S1(a) presents always an exponent of -3 following equation (4) of the main text. The fitting of the curves of figure S1 (b) gives always an exponent of -4 following equation (8) of the main text. Therefore, the fitting results are only dependent on the position of the beam along the jet, and not on its position across the jet.

3. Experimental high speed video of microjet breakup

3.1 Experimental conditions

In order to visualize the jet breakup and the shape change of the droplets immediately after jet breakup we have used high-magnification optics mounted on a high-speed CCD camera which has allowed independent control of the frame rate and the exposure time per frame (model HISPEC, FASTEC Imaging Inc, San Diego, CA, USA). The experimental setup had a spatial resolution of approximately12 µm/pixel. A water jet exiting from a tube with 100 µm diameter and a flow rate of 2.235 ml/min has been investigated.

To achieve a good image contrast an acquisition time of 5 µs has been selected and the light source (a halogen lamp with an isotropic diffusor) has been placed directly behind the jet and pointing into the camera. With this setup, the background features a radial gradient, with a bright area at the center of the light and becoming darker towards the borders of the image. As a result of this non-uniform illumination, the contrast between the jet and the background also changes along the jet trajectory. The position of the jet has been adjusted so that the highest contrast region is the jet breakup zone, guaranteeing satisfactory contrast for at least 3 mm after jet breakup, as shown in Fig. S3a.

3.2 Video image analysis

The objective of the video analysis has been both to measure the breakup length and to establish whether the oscillations detected with SAXS in the breakup region are compatible with shape changes in the droplets. Given that SAXS provides a time and space averaged measure of the sample, we have decided to extract the “average jet” from the video. Due to the high speed of the jet, a given droplet moves during the exposure. The examined jet had an average velocity of 4.74 m/s, i.e. a displacement of 4.74 µm in a microsecond. Limiting the exposure time to 5 µs has ensured that the blur was contained to about 2 pixels along the jet trajectory and the images were sharp enough to perform the analysis in the illuminated region (see Figure S3). The average jet has been computed by giving, in each frame, a value of 1 to the pixels containing fluid (jet or droplet), and of 0 otherwise, adding all frames. This has involved subtracting the bright background first and applying a threshold on the resulting image to get a binary representation. The background has been modelled by a two-dimensional Gauss distribution, which has been fitted to each frame, masking out the region occupied by the jet and the droplets, and the center of the bright spot from the lamp, which saturated the detector resulting in an artificial plateau. The high values of the fit result have been clipped to the maximum detector value and subtracted from the original data, resulting in an image with a uniform light grey background as shown in Fig. S3b.The threshold level has been set to 20% of the difference between the maximum and minimum pixel values of the frame, to take into account the slight oscillations in the overall image intensity. This criterion has produced droplet shapes that are slightly larger than those perceived in the original frame or the background-subtracted image, due to the motion blur along the jet trajectory (Fig. S3c).

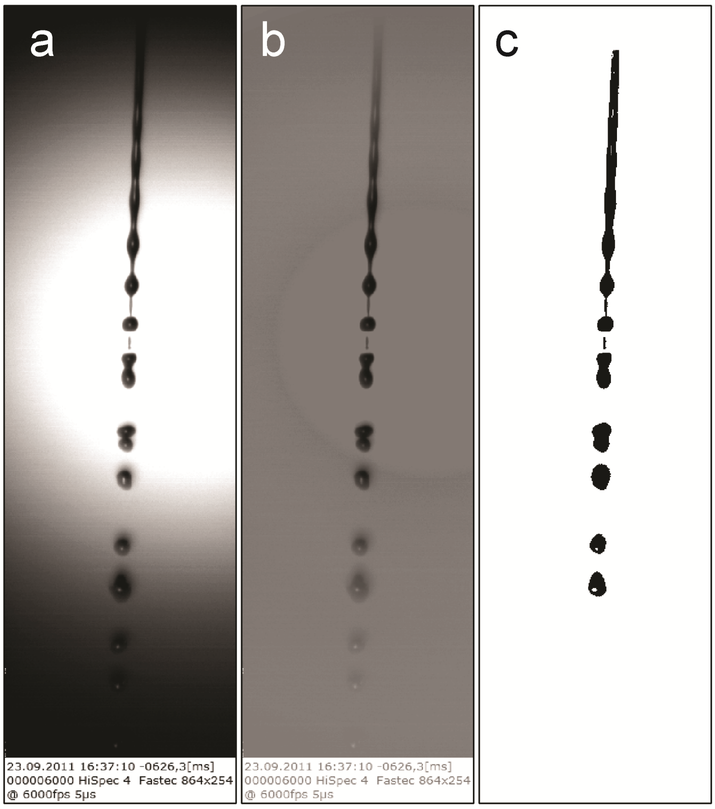

Figure S3. : (a) Radial-gradient image, 5 µs, 6000 fps: raw image. (b) Background-subtracted image. (c) Binary image after threshold.

3.3 Results

After the elaboration, the jet has been averaged, as shown in Figure S4a. It clearly shows alternating wide and narrow regions at a distance of approximately 0.5 mm from each other, which result from the change in the droplet shape from oblate to prolate and back. The absence of droplets is due to the variations in their position over the duration of the video acquisition, due to random oscillations in the experiment (environment, vibrations, oscillations in the pumping rate etc). These variations can be clearly seen in Figure S4b, which shows three non-consecutive frames from the series. The rectangle in the center of the image represents the approximate X-ray beam footprint in the SAXS experiments.

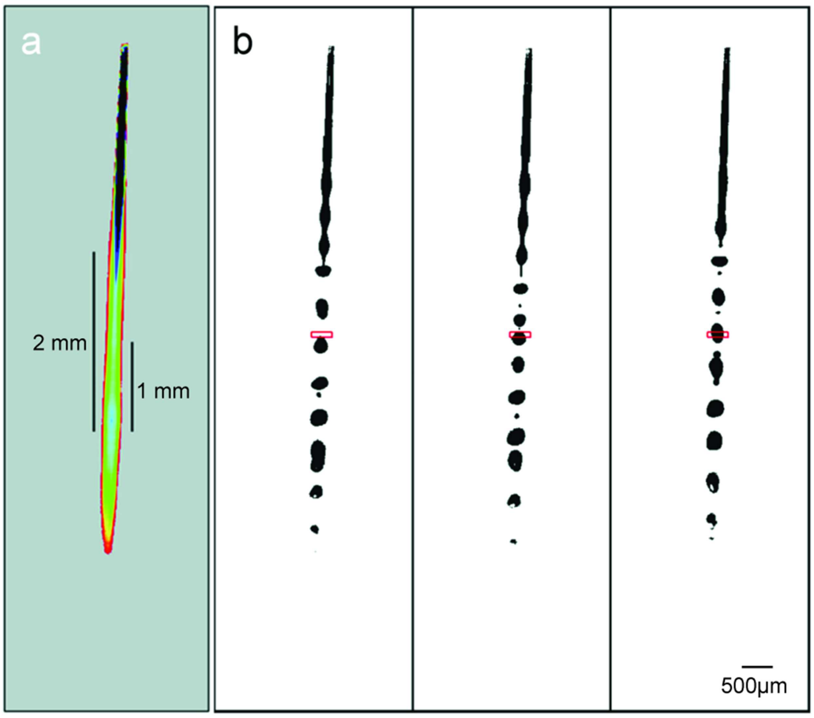

Figure S4. (a) Average jet (sum of all frames in the video). The background color has been set to grey in order to increase the color contrast. (b) Three frames of the same video acquisition. The time interval between the frames is approximately 5 ms between the first two, and 2 ms between the last two. The rectangle in the center of the image represents the approximate X-ray beam footprint in the SAXS experiments.

4. Breakup length for water mixtures of ethanol and IPA

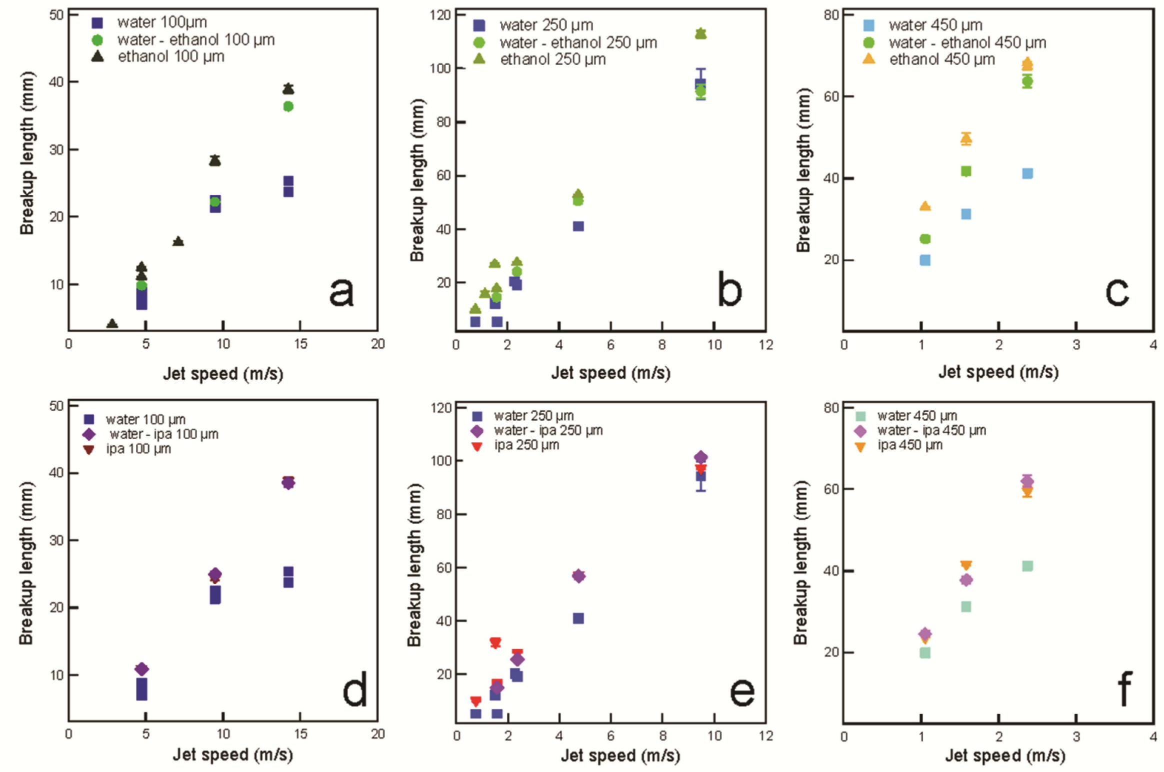

Figure S5. (a) Breakup length as function of the jet speed at the nozzle exit for water, ethanol and water-ethanol mixtures at 50% volume: diameter of the nozzle 100 µm, (b) diameter 250 µm, (c) diameter 450 µm. (d) Breakup length as function of the jet speed at the nozzle exit for water, IPA and water-IPA mixtures at 50% volume: diameter of the nozzle 100 µm, (e) diameter 250 µm, (f) diameter 450 µm. It can be noted that for both mixtures the behaviour is more similar respectively to the one of ethanol and IPA than to water.