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# Oscillation, coalescence and shape deformation of confined bubbles in an oscillating depth microchannel: application to the fragmentation of electrocrystallized ramified branches for an alternative synthesis of colloidal metallic nanoparticles

Fabien Chauvet<sup>1</sup>

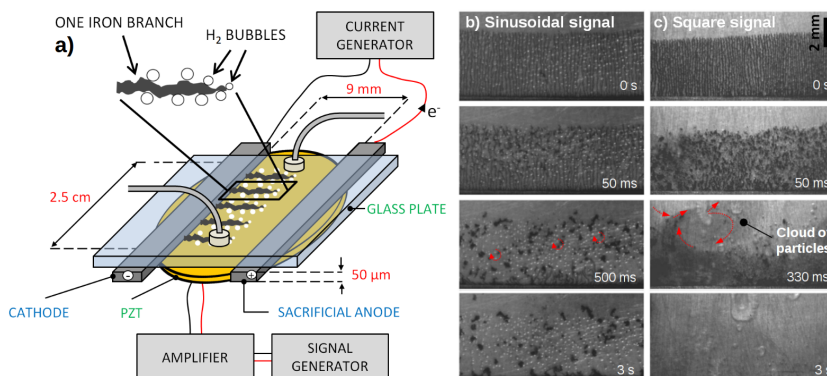
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## Introduction

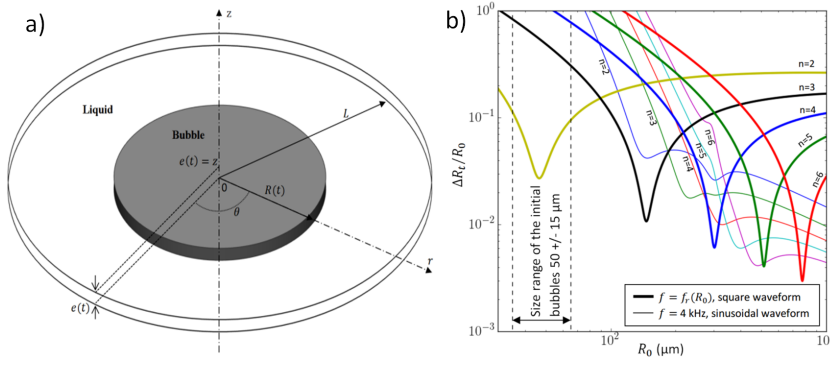
This work focuses on the behavior of bubbles confined in an oscillating depth microchannel. This situation is encountered in a recently developed device, an “electrochemical and vibrating Hele-Shaw cell”, designed to synthesize colloidal metallic nanoparticles (cMnP) [1]. The principle is to make grow metallic ramified branches by galvanostatic electrolysis of a metal salt aqueous solution inside the cell, Fig. 1a. These fragile branches are composed of nanocrystals (the desired cMnP), whose the dissociation is achieved by the mechanical action resulting from the activation of a piezoelectric diaphragm (PZT) integrated into the cell as one of its largest side, Fig. 1a; the variations of the channel depth are induced by the bending of the PZT surface. In the case of the production of iron cMnP, the branches growth is accompanied by the formation of H<sub>2</sub> bubbles (co-reduction of H<sup>+</sup>), Fig. 1a. High speed visualizations of the fragmentation process highlight the key role of these bubbles whose the oscillations induce both microstreaming and branches fragmentation in close vicinity of their surface, Fig. 1b-c. An efficient fragmentation is obtained only when the PZT is driven with a square signal, Fig. 1c. The observed dependence of the bubbles behavior, to the waveform used, is analyzed with the support of specifically derived theoretical models for both breath oscillations (Rayleigh-Plesset like equation) and shape oscillations (stability analysis) of a single bubble in an oscillating depth microchannel. The specificity of the oscillating depth (the driving force), acting simultaneously on both the bubbles size and the liquid flow, is taken into account using a modified Darcy’s law adapted to small depth variations.

## Experimental results

The images sequence of the fragmentation scene, using a sinusoidal signal ( $f = 4$  kHz and  $V_{pp} = 250$  V), Fig. 1b, shows that the branches are broken and the fragments are grouped into several blocks, set in a stationary rotational motion in the plane of the channel (red arrows in Fig. 1b), between the bubbles. These rotational motions are the signature of microstreaming induced by the bubbles oscillation. Nevertheless, these flows are not sufficiently fast to fragment the branches into small particles. It has to be noted that the bubbles network is unaffected by the PZT vibrations (no motions and no surface deformations). By performing the same experiment, but using a square signal, Fig. 1c, a more complex scene is visualized. Some of the initial bubbles coalesce to form larger bubbles which exhibit surface deformations, splitting/coalescence events and motions inside the channel. Microstreaming is also observed (red arrows in Fig. 1c) and the resulting flows break the branches and set them in motion. The fragments end up being “attracted” to the surface of unstable bubbles, where they are fragmented and ejected in the form of a cloud of particles, as it is shown in



**Figure 1:** a) Sketch of the electrochemical and vibrating Hele-Shaw cell. b-c) Images sequences of the fragmentation process for sinusoidal b) and square c) signals, the arrows indicate the observed particles motions ( $f = 4$  kHz,  $V_{pp} = 250$  V, acquisition frequency = 1500 FPS; FeCl<sub>2</sub> 0.1 M, 80 mA/cm<sup>2</sup>, growth duration = 300 s).



**Figure 2:** a) The model situation considered. b) Pulsation amplitude threshold for the onset of shape deformations as a function of the bubble size for several modes  $n$  and for both a sinusoidal waveform at 4 kHz (thin lines) and a square waveform at  $f = f_r(R_0)$  (thick lines).

Fig. 1c. To sum-up, the fragmentation of the branches is observed only when large bubbles, formed by coalescence, oscillate with surface deformations when the PZT is driven with a square signal. To explain the appearance of the first coalescence events, and so the setting in motion of the initial bubbles, the assumption is made that using a square signal, bubbles pulsation amplitude is sufficiently high to induce the onset of shape deformations which lead to erratic bubbles motion, when different modes of shape oscillation interact; this phenomenon is known as “dancing bubbles” for spherical and unconfined bubbles [2].

### Derivation of the pulsation amplitude threshold for the onset of shape deformations

The bubbles oscillation is considered in a simplified and model situation: a single confined bubble, of rest radius  $R_0$ , located at the center of an oscillating thickness circular film as sketched in Fig. 2a. The oscillation of the bubble is driven by an imposed oscillation of the film thickness  $e(t) = e_0 + \epsilon(t)$  which the fluctuation  $\epsilon(t)$  is synchronized with the driving voltage. The modeling is based on the classical theory for the oscillations of unconfined bubbles aiming to derive a Rayleigh-Plesset equation, to describe the temporal evolution of the bubble radius  $R(t)$ , and an equation for the amplitude of surface fluctuations. The liquid flow is modeled by a modified Darcy’s law (taken into account inertia) adapted to small depth variations. This law is derived assuming the liquid velocity profile across the thickness is parabolic (this is valid for small Reynolds number [3] and so small  $|\epsilon|$ ). First, breath oscillations (no shape deformations) are considered and after integration and linearization of the Darcy’s law, the equation of an equivalent forced and damped harmonic oscillator, providing the resonance frequency of the bubbles  $f_r$  (among other parameters), is obtained.

Next, the stability of the circular shape of the bubble is considered by authorizing small distortions of the surface  $r_s(\theta, t) = R(t) + a(t)\Psi_n(\theta)$  (with  $|a| \ll 1$  and  $\Psi_n$  is a circular harmonic of degree  $n$ ) and assuming that the corresponding flow perturbation is the same as in the case of a long 2D bubble ( $e \gg R$ ). The previous integrations are re-performed, to obtain a second order equation on  $a$  which is converted into the Mathieu’s equation, as classically done. Using the theory on the stability of the corresponding solutions, an expression for the threshold,  $\Delta R_t / R_0$ , is obtained. This result is easily applied to the case of a sinusoidal waveform but it cannot be directly applied to a square waveform. Nevertheless, since the resonance frequency of the initial bubbles  $f_r = 30$  kHz ( $R_0 = 50 \mu\text{m}$ ) is well higher than the applied frequency of 4 kHz, the bubbles should oscillate at their resonance frequency after a step (rise or fall) of the square signal. By considering these oscillations as stationary (low damping parameter), the threshold for the square waveform used can be estimated. The predicted thresholds are plotted as a function  $R_0$  for several  $n$  in Fig. 2b. The developed theory shows that no shape deformations should occur for the initial bubbles when using a sinusoidal signal at 4 kHz whereas they should appear if pulsation amplitude exceeds  $\sim 3\%$  when using a square signal. The pulsation amplitude has not been measured, but it has been verified that below a threshold voltage amplitude ( $\sim 120$  V), no coalescence events are observed. Additionally, keeping the same voltage amplitude of 250 V, but varying the frequency (0.1 - 4 kHz), coalescence and fragmentation events are always observed. These results are in agreement with the developed theory.

### Conclusion

The observed coalescence of bubbles confined in an oscillating depth microchannel, occurring when using a square waveform, is required to dissociate the cMnP. This particular behavior is explained by the appearance of shape deformations leading to a dancing bubbles effect. The developed theoretical models can be applied to other acoustofluidics devices using a low-frequency piezoelectric diaphragm in direct contact of confined bubbles.

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