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ABSTRACT

The dynamics of liquid-liquid mixing is a difficult problem, encountered in many scientific and engineering branches. Experiments in this field are mandatory to help building sound mathematical models, finding out the best fit parameters, evaluating the degree of confidence of these models, or detecting traces of unwanted dangerous substances. The investigations reported here are driven by water pollution concerns. For analyzing the water-pollutant blending behavior, dynamic speckle interferometry has been preferred to more standard optical full field methods, like deflectometry, or classical and holographic interferometry. The choice of this technique is vindicated. The opto-fluidic system is described. A first series of results is presented, demonstrating the effectiveness of the technique and showing qualitatively how two liquids blend in controlled conditions. In the last part of the paper, recently appeared processing schemes, including empirical mode decomposition, Hilbert transform and piecewise treatment, give access to the numerical values of the phase maps computed for each frame of the recorded sequence. These phase maps represent the refractive index distributions integrated along the line of sight. They provide a better visualization of the dynamics of the blending behavior and therefore an improved understanding of the phenomena. These encouraging preliminary results should open the door to a full characterization of the method and to further flow investigations and diagnostics.

Keywords: liquid blending, water pollution, blending flow visualization, dynamic speckle interferometry, phase extraction, refractive index map.

1. INTRODUCTION

One of the most serious problematics in environmental protection is the release of pollutants in natural water. Industrial activities have greatly increased the transport and use of harmful products, multiplying the risks of accidental leakages. Such accidents can lead to considerable eco-toxicological effects on river, lake and maritime fauna and flora and sometimes on the population. The maritime route is the main way of transportation and is constantly increasing. For example, an increase of 270% of the transport of chemicals has been observed from 1975 to 1995. As a consequence the probability of chemicals accidental releases is increasing as well. For example, the English Channel, which is one of the busiest seas over the world, concentrates 20% of the world marine transport and during the last decade many accidents occurred.

This fact induces the necessity to better understand the behaviour of chemicals releases at the sea surface and also from a shipwreck. This paper concerns this latest aspect. Indeed, in the case of a floating product released from depth, the physico-chemical mechanisms appearing during the products way up to the surface, such as the rate of rise and of solubilization, are still poorly understood, although these parameters influence the extent of pollution at the surface.

As examples, we can mention the *Ievoli Sun* (2000) and the *ECE* (2006) sinking. They both sunk with their noxious cargos: the *Ievoli Sun* carried 4000 tonnes of styrene and 1000 tonnes of Methyl Ethyl Ketone (MEK) and the *ECE* sunk with 10,000 tonnes of phosphoric acid on board. The lacks of knowledge concerning the behaviour of chemicals at sea can be dangerous for people. During the *ECE* accident, French Navy divers were exposed to the acid because it did not dilute as the literature predicted it. During the *Ievoli Sun* accident, small leaks of styrene were observed underwater which rose to the surface and exposed people in charge of the intervention to styrene emanations. Moreover, the MEK presence in the cargos would have lead to disastrous consequences. Indeed the MEK has a lower density than water and is not completely soluble in water. So in case of release from depth it would rise quickly to the surface and form a slick at the sea surface. The evaporation rate of this product is very high and the cloud is an explosive one. In this respect, this behavioural should be well understood for responders on scene.

At large, fight against water pollution includes the refinements of both computer models and experimental investigations, aiming at a better understanding of the dynamics of liquid-liquid mixing [1]. Blending flows and rates need to be better known within a large range of representative volume elements. On the experimental side, however, much of the sought-after pollutants, e.g. Methyl Ethyl Ketone, being as limpid as clear water, cannot be seen by standard imaging systems that record only the object intensity. Phase imaging techniques are required and fluid mechanics usually makes use of phase-contrast, spatial filtering, classical or holographic interferometric methods [2-5]. Here, we report on a laboratory experiment aiming at controlling the pollutant release in a clear water cell, using speckle interferometry (SI). The cell is optically projected onto a ground glass diffuser and the coherent light emanating from the diffuser forms one arm of a Mach-Zehnder type speckle interferometer; the reference arm is directly superimposed in-line to the speckle arm in front of the digital camera. This disposition offers the great advantages of being much easily adjusted than the corresponding classical interferometer, to require cheap off-the-shelf optical components and to facilitate the observation of the samples. Specklegrams are recorded at frame rates of 200fr/s by a high speed imaging system.

The differences of refractive index between water and pollutant create phase changes exhibiting temporally a large dynamical range and spatially rather complicated patterns. Extracting the phase of the specklegrams is not a trivial task. Movies showing the live correlation fringes by subtracting consecutives specklegrams, in fixed or floating reference modes, serve to control the speed of the pollutant injection. Jointly with tests of the modulation depth of the temporal pixel signal, they allow selecting the best intervals for computing the phase. Among an open choice of possible approaches [6], the empirical mode decomposition (EMD) is firstly used to adaptively remove the background intensity from the temporal pixel signal. The 1D Hilbert transform (HT) is then used to compute the phase from the so-centered signals and a filtering technique based on the Delaunay triangulation (DT) is conducted to eventually recover a smooth phase map at each instant of the liquid blending experiment [7, 8]. Morlet wavelet analysis is also considered as a complementary and competitive processing technique for performing the task [9].

These results are thought to provide a set of boundary conditions for blending liquids simulation purposes and to help fixing the ad hoc parameters of the numerical models. Further developments will include the comparison of the results with other techniques like interferometric schlieren.

2. CHOICE OF SPECKLE INTERFEROMETRY

Speckle interferometry (SI) is now widely used to measure 3D displacement maps of rigid bodies. Most of the time these objects are optically rough and then, generate speckle while illuminated with coherent light. If objects under study are specular some hints can be used to make them generate speckle. So SI can also be applied to optically polished objects [10]. This is the particular case of this study concerning fluid mixing. For optically polished objects, to pass from classical to speckle interferometry is basically achieved by letting the direct light object beam illuminate a ground glass diffuser.

Dynamic study of speckle changes is of great interest and is now discussed in several international conferences with special topics session [11].

Compared to other techniques widely used in fluid dynamics (shadowscopy, schlieren, …) SI exhibits high sensitivity to refractive index changes and can lead to quantitative measurements. Fundamentally, this sensitivity is the same as for classical and holographic interferometry, i.e. $2\pi/\lambda$ per unit of optical path change where λ is the wavelength of the operated source. This high sensitivity can also be considered as a drawback when the phase change is very fast. In opposition to classical interferometry, and in the setup presented in the next section, the fine adjustment of the parallelism of the two interfering beams does not need to be done. Due to the differential modus operandi of SI, the optical components can be of a standard quality – as opposed to the expensive interferometric class. Superimposing the object- and reference-beam on the photo-detector with the appropriate field of view is much easier in diffuse than in collimated light because an afocal system is no more necessary to adapt the beam size to the photosensitive area.

To ensure successful experiments involving liquid mixing, high speed imaging has been used to cope with the high rate of refractive index changes. Some authors have already included phase shifting for high speed imaging [12]. This can require, e.g., corner cube mirror travelling along an optical bench in the reference arm [13]. To keep acquisition speed while preserving simplicity, phase-shifting is not attempted in our experiment, the phase extraction being performed by means of more recent temporal analysis methods, as reported in section 5.

The final goal of this experiment is to determine the phase change of the mixing fluid. Therefore we have to make the selection between in-plane and out-of-plane sensitivity vectors. In plane sensitivity is not compliant with fluid mixing in our configuration, so we use pure out-of-plane sensitivity with speckle reference beam.

3. EXPERIMENTAL SETUP

The experimental setup is presented Fig. 1. The light source is a frequency-doubled solid state YAG 2W continuous wave at 532nm of wavelength. Optically rough ground glasses have been placed symmetrically from the mixing plate to create the speckles in the Mach-Zehnder interferometer. Different roughnesses of the ground glass can be used. The optical tank is placed in one arm while the second arm is acting as the reference arm. The optical tank is made of plastic and is a COTS sample from visible spectrometry (1x1x3 cm^3 parallelepiped). This will be replaced in the future by a clear optical quality glass tank as MEK is corrosive for plastics. Moreover it will be possible to test the position of the optical cell in front or behind the diffuser.

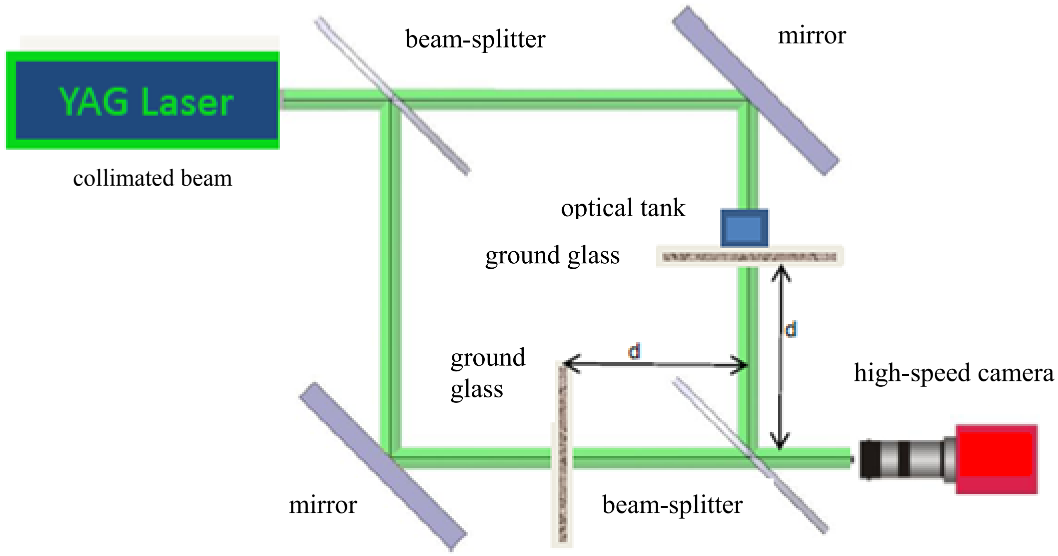

Figure 1: Experimental setup

The specklegrams resulting from the interferences of the two arms are then grabbed with a high speed imaging system. The imaging camera is able to grab 200 frames/second with maximal resolution of 640×480 pixels. It is coupled with a 105 mm macro objective to image the ground glass surfaces onto the sensor and to ensure the full field of view of the optical sample. The size of the speckles is chosen to get the best fringe contrast after subtraction of a recorded state. During the mixing process, the water cell is filled with MEK from a syringe pin placed in the bottom of the tank. The rate of mixing is done manually and this will also be improved in the next future. Nevertheless we got bright fringes by subtracting a reference state. This reference state can be static or dynamic. For the static case, a reference image is subtracted from each image of the movie while for dynamic case the live displayed *n*-image is subtracted from another running *n-x* image, where *x* denotes the number of images in between. This leads to fringes that will be further interpreted by real time correlation.

4. QUALITATIVE RESULTS: FLOW VISUALIZATION BY MEANS OF REAL TIME CORRELATION FRINGES

Correlation fringes created by the subtraction of two specklegrams recorded at two different instants of the blending process provide a visualization of the phenomenon. We present in Fig. 2 such a visualization for the instants corresponding to frames # 10, 50, 100, 150 and 200, 50 frames being separated by ¼ of a second. A movie made of the same type of differences, but now by incrementing one by one the frame number of the frame subtracted to the initial reference specklegram, show still better than the frozen fringes of Fig. 2 the formation and the evolution of zones where laminar or turbulent flows dominate. Correlation fringes, which can be displayed afterwards at standard video frequency, are used to determine both the adequate rate of injection of the pollutant and the corresponding frame acquisition rate ensuring a correct temporal sampling of the phase change. They serve also to select the regions of interest exhibiting the best contrast, where a meaningful phase extraction can be attempted.

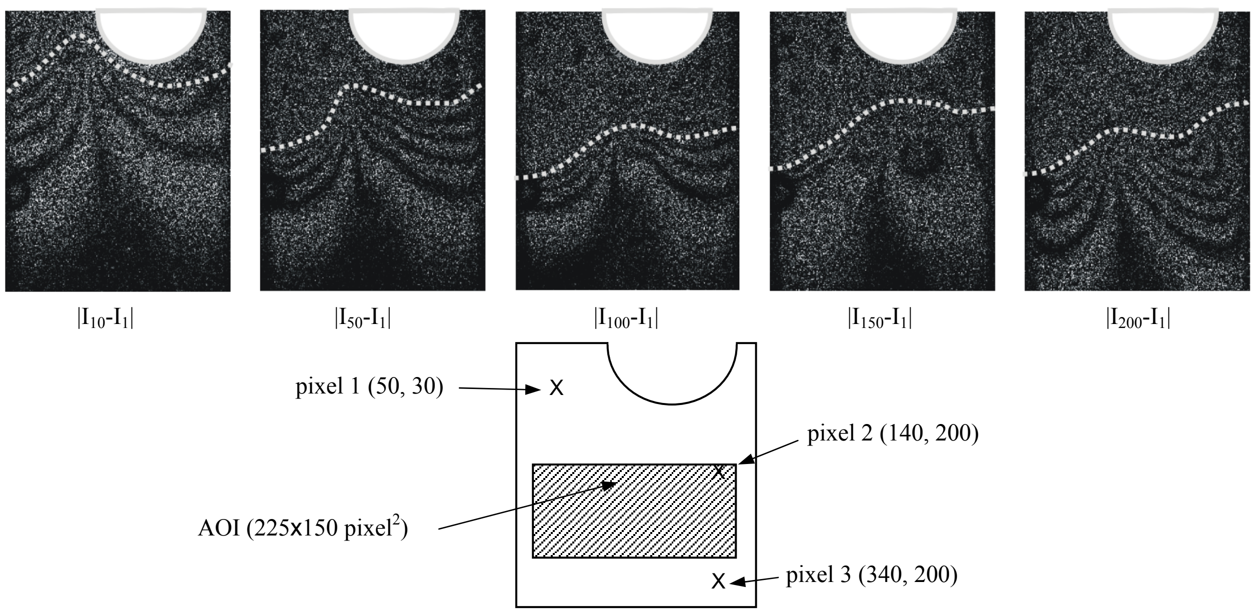

Figure 2: Correlation fringe patterns obtained by subtracting the specklegram recorded at frame # 1 from the specklegrams recorded at frame # 10, 50, 100, 150 and 200 showing qualitatively how the two liquids in the cell progressively blend with the appearance of laminar and turbulent flow regions. The half circle is a dead zone corresponding to the position of the liquid injector. The lower drawing sketches the pixels (x) and the AOI (area of interest) used for subsequent quantitative determination of the phase.

5. QUANTITATIVE PHASE EXTRACTION: INTEGRATED REFRACTIVE INDEX DISTRIBUTIONS

5.1 Phase extraction methods

Among the diverse well known possibilities to extract the phase in a dynamic SI experiment [6], we preferred to treat the set of the concerned 1D temporal pixel signals rather than the corresponding sequence of 2D correlation fringe patterns. Two broad categories of methods are retained: the Morlet wavelet analysis [9] and a combination of empirical mode decomposition, Hilbert transform and piecewise processing, named in short EMD+HT+3DPP [7-8, 14-15].

As a quick reminder, the wavelet analysis is the construction of a two-dimensional distribution or scalogram, which represents the signal energy distribution per time and frequency unit, by convolving the original signal with a bench of functions which are all the scaled and time-shifted versions of a mother wavelet. In the case of the Morlet wavelet which remarkably fits with our application[9], the mother wavelet is a complex sine wave whose support is limited by a smooth function, usually a Gaussian window. The sought-after instantaneous frequency (IF) can then be computed by looking for the maximum of the so-built distribution modulus (ridge) at each instant, but a much neater and more robust method, though less sensitive to brutal changes of IF, consists in tracking its ridge, exploiting the fact that the signal and the wavelet phases coincide in there.

The EMD[16-17], fed with any nonstationary signal, yields a sparse set of functions which are its intrinsic oscillations modes. The basic idea is to extract the features of the original signal from the highest frequencies to the lowest, by iteratively subtracting the mean envelope. This is achieved by averaging the top and the bottom envelopes, obtained by spline fitting respectively the maxima and the minima of the signal. It has been shown [6-7] that EMD can be profitably applied to the context of SI by efficiently removing the randomly fluctuating background intensity and thus put the temporal pixel signal in the ad-hoc shape for subsequent phase computation. For that matter, the analytic method is probably the most wide-spread technique for computing the phase from a real-valued centered signal. It consists merely in computing a quadrature signal with the HT to eventually construct a complex-valued signal, from which the phase can be straight-forwardly obtained. Finally, the 3DPP [6-7] technique is a filtering technique which consists in discarding the less reliable pixels, following a given criterion on their modulation, and spline fitting the remaining points non-uniformly spread across the area of interest, with the crucial requirement to best-preserve the spatial resolution. This is done by interpolating the areas where the information is missing with the closest valid data, and this is where the Delaunay triangulation (DT) appears. The DT is a triangulation where each facet is the less stretched possible, or in other words, the facets of a DT are the closest possible to the equilateral triangle, indeed best-preserving this way the spatial resolution.

5.2 Experimental results

5.2.1 Phase extraction for a selection of three temporal pixel signals

We report here the temporal pixel signals during 200 frames with their centered versions, obtained through the EMD, and the corresponding extracted phase with the Morlet wavelet transform and the EMD+HT method.

a)

b)

c)

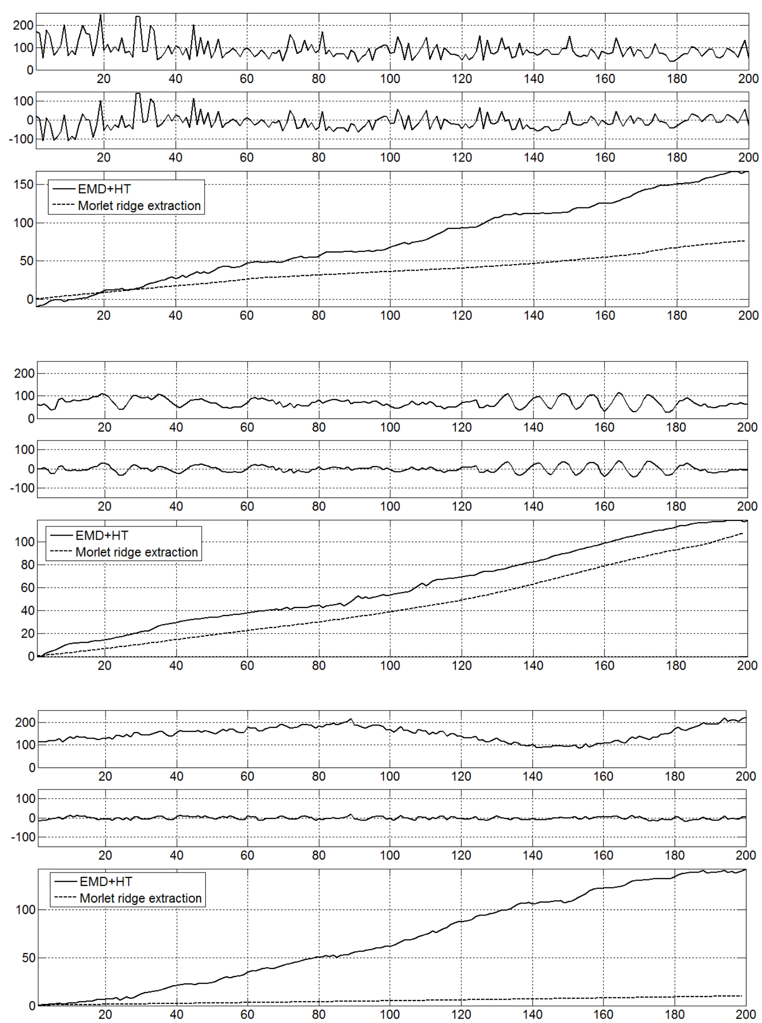

Figure 3: Temporal signal, its centered version and the extracted phase respectively at pixel 1 (a), 2 (b) and 3(c).

As predicted before, based on the correlation fringes, the strong under-sampling of the phenomenon forbids any kind of reliable phase extraction from the temporal signal recorded at *pixel 1*. The temporal signal recorded at *pixel 2* features a perfectly exploitable fringe modulation. The "EMD+HT" performs well in that case – especially from time instant 120. The Morlet wavelet transform and its implementation with the ridge extraction algorithm yields of course a smoother result than "EMD+HT" only but the 3DPP technique has proved to greatly improve the result [6-7]. For the last case presented here, the *pixel 3* manifestly experienced little activity (one fringe and half) and the strong oversampling does not ease the task of removing the background intensity. EMD extracted only the noise and the subsequent phase extraction is soiled with salt noise, which is efficiently removed with 3DPP. The Morlet wavelet yields a relevant result as soon as the ridge tracking algorithm is correctly initialized.

5.2.2 Examples of full field phase maps obtained for definite AOIs and time intervals

We give below the final phase maps obtained within the selected AOI (see Fig. 2), with the three considered methods, i.e. the "EMD+HT", the Morlet wavelet transform and the "EMD+HT+3DPP" methods. The considered time interval is from frame 120 to frame 190.

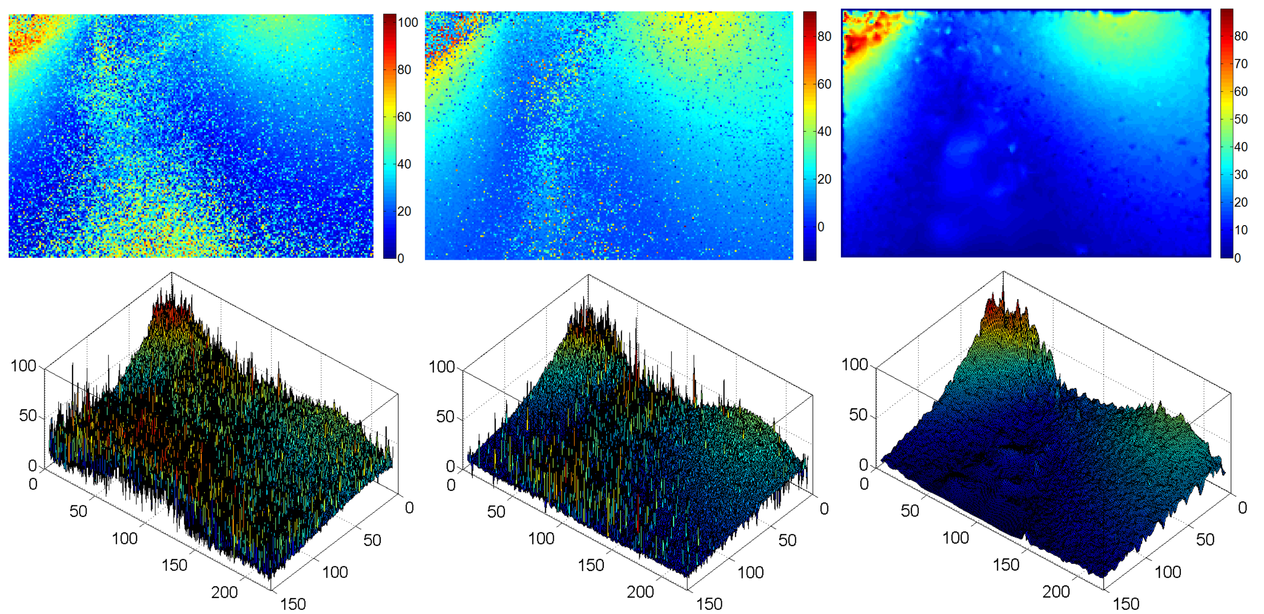

Figure 4: final phase map for the selected AOI for respectively the EMD+HT method (left), the Morlet wavelet method (center) and the EMD+HT+3DPP method. The colorbar in the top row and the z-coordinate in the bottom one are given in radians.

As expected from the analysis of a temporal signal recorded at a pixel which experienced few activity (*pixel 3* in Fig. 2), the central bottom part of the AOI is highly polluted by salt noise, which is greatly reduced by 3DPP. As also expected, the ridge tracking algorithm performs better than EMD+HT in low SNR regions. Salt noise is also noticeable in the top left part where the blending phenomenon is slightly under sampled. For the 3DPP application, we used a threshold so that only 15 % of the pixels have been kept. The choice of the threshold is up to the operator and represents a trade-off between computation load, noise rejection and spatial resolution loss. In the low activity area, few pixels have been classified as relevant and the DT produced large facets: the 3DPP technique discards just as well pixels with low modulation and pixels with low activity. Consequently, the cubic spline interpolation step produces artificial smooth oscillations.

5.3 Some brief comments on phase extraction methods

The chosen isolated pixels and full field examples of phase extraction show that it is important to select the most appropriate method, and to run the corresponding program with the most adequate user-defined parameters. Starting from the same set of recorded rough data, several processing approaches and techniques could even be used conjointly.

A very important point is to be able to assess everywhere the quality of the interferometric signals. In this respect, EMD gives an easy access to the modulation depth of the signal and responds to this point by providing the lengths and positions of the intervals where this modulation is in excess of a predefined threshold. Finally, an obvious, although worthy remark: one is forced to recognize that the best processing tools, powerful though they are, are indeed inefficient when dealing with signals of too poor quality. This is what happens in some AOIs in the presented experiments. In these cases, any endeavor to first improve the rough signals should be done.

6. OUTLOOK AND CONCLUSION

Those first results show the behaviour of two fluids (liquids) while mixing. As argued in the body text, it is a preliminary study, mainly concentrated on the optical aspects of the experimental part of the problem. Hopefully, further results will be obtained with better contrast expected from different means such as:

- Clear optical glass tank and well-controlled injection system to avoid caustics: some areas of the blending region act as microlenses and disturb the specklegrams acquisition, by essentially saturating the array sensor.
- Adjunction of a fringe carrier for sign ambiguity removal
- More appropriate sampling by the use of a faster camera (2000fr/s, 1024×1024 pixels), which should help to better fulfil the correlation conditions between any two consecutive specklegrams.

As a consequence of these improvements, signals of higher modulations should be obtained, giving rise to an easier and more accurate phase computation, and making possible in addition the simplification and the standardization of one or the other of the variety of above investigated phase extraction methods. High quality dynamic phase maps should then lead in turn to the ultimate goal, namely to the characterization of biphasic flows in terms of fluid mechanics parameters, including the feasible computation of their spatiotemporal distributions.

These results will also be compared with other optical techniques for fluid mixing investigation[18].

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