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# Validation of X-ray radiography for characterization of gas bubbles in liquid metals

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**Abstract.** X-ray radiography has proved to be an efficient and powerful tool for the visualization of two-phase flows in non-transparent fluids, in particular in liquid metals. This paper presents a validation of the X-ray radiography by comparing measurements in water with corresponding results obtained by optical methods. For that purpose Ar bubbles were injected through a single orifice. The measurements results are compared in terms of bubble size, bubble shape and velocity. Furthermore, visualization experiments were performed in the eutectic alloy GaInSn where the image contrast between the liquid phase and the gas bubble is much stronger. Some obvious differences of the bubble dynamics in water and GaInSn are discussed.

## 1. Introduction

Liquid metal two-phase flows are an important part of many technical applications in metallurgy and continuous casting. Argon gas is injected during continuous casting in order to prevent clogging of the submerged entry nozzle and to separate undesired inclusions from the melt. Gas stirring is used in ladle metallurgy to homogenize the melt and to improve the cleanliness of the steel. On the other hand, injection of gas implicates many side effects as for example the generation of highly turbulent complex two-phase flows and creation of additional defects in final casted products. Effective control and optimization of this process requires a comprehensive understanding of the behavior of liquid metal two-phase flows. Many numerical and experimental studies of gas bubbles rising in water and transparent viscous liquids exist so far covering a broad range of Reynolds and Eötvös numbers (see for instance [1–9]). Many studies follow the approach to extrapolate the bubble behavior in liquid metals from experiments in water. However, strong differences in material properties such as density, viscosity and surface tension lead to discrepancies in essential non-dimensional parameters as the bubble Reynolds number, the Weber number or the Morton number. Therefore, direct experimental investigations in liquid metal two-phase flows become important and desirable. However, the value of experiments carried out in real liquid metal flows depends on the availability of trustworthy and efficient measurement techniques. Previous measurements in liquid metal bubbly flows were performed by means of ultrasound Doppler velocimetry (UDV) [10], by conductivity probes [11], by Local Lorentz force velocimetry (LLFV) [12] or by neutron radiography [13,14]. Most of these techniques have limitations. For example, ultrasonic methods meet problems with increasing gas content when many bubbles enter the measuring volume. Multiple echoes at the bubble interfaces may generate signal artifacts. Conductivity probes are intrusive and provide only local information. Recent studies have demonstrated the capability of X-ray radiography to be an efficient tool for the

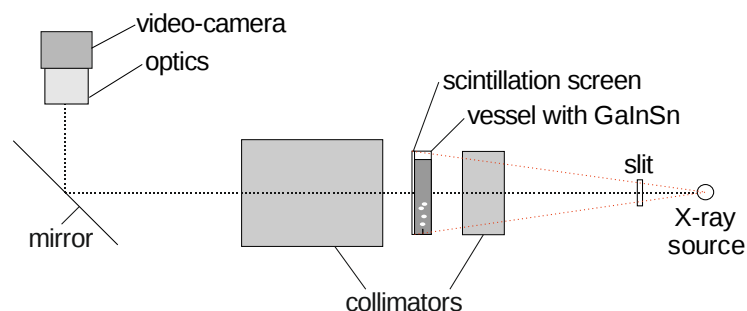


visualization of liquid metal two-phase flows [15–20]. X-ray radiography is a fully contactless method based on the absorption contrast between the liquid and gas phase. The weak point of this technique is the limitation of the sample thickness due to the high attenuation coefficients in liquid metals. Previous publications did not discuss the accuracy of the X-ray radiography for determining parameters like bubble size, bubble shape and bubble velocity in detail. The aim of this work is to demonstrate that the analysis of the X-ray radiography images provides accurate results for bubble size and shape. For this purpose we compare the results of the X-ray radiography with optical measurements performed in water. Further experiments were carried out in the eutectic GaInSn alloy.

## 2. Setup and methods

### 2.1. Experimental setup

Bubble visualization experiments were carried out at the X-ray laboratory at HZDR. The scheme of the setup is demonstrated in figure 1. A high power X-ray source (ISOVOLT 450M1/25-55 from GE Sensing & Inspection Technologies GmbH) operating with a maximum voltage of 320 kV and a current of 14 mA generates a divergent polychromatic X-ray beam. A scintillation screen (SecureX HB from Applied Scintillation Technologies) is attached to the surface of the container as shown in Figure 1. The non-absorbed part of the X-ray beam comes upon to this scintillation screen where its intensity is converted into visible light. The further imaging is completed with a lens system (Thalheim – Spezial - Optik) and a high-speed video camera (Pco.edge from PCO) equipped with a sCMOS-sensor. The images were captured with 100 frames per second (fps) and an exposure time of 3 milliseconds. The exposure time was optimized to achieve a good signal-to-noise ratio without causing bubble blurring due to their high rising velocities. The container is made of acrylic glass because the walls do not cause significant attenuation of the X-ray beam intensity. The container represents a rectangular tank with 12 mm gap and 144 mm width which was filled with water or with liquid GaInSn alloy up to a height of 144 mm. The eutectic GaInSn alloy is liquid at room temperature. Thermophysical properties of the alloy are reported in [21]. In our experiments the field of view was approximately  $60 \times 110 \text{ mm}^2$ . The inert Ar gas was injected through a long bevel stainless steel orifice of 1.1 mm outer diameter (from Sterican®) positioned in the middle of the bottom part of the container. The Ar gas flow rate was  $50 \text{ cm}^3/\text{min}$ . A diffused light source was used for the optical measurements to minimize the light reflection at the bubble interface.

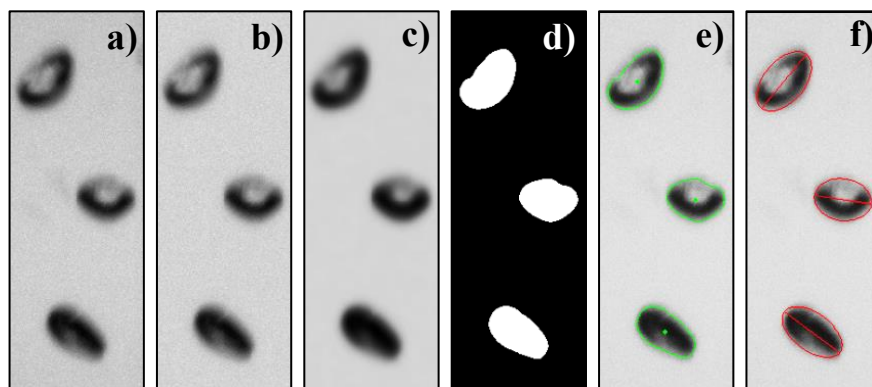


**Figure 1.** Scheme of the X-ray diagnostic setup

### 2.2. Image data processing

The quantitative analysis of the bubble dimensions, positions and velocities are performed by off-line data processing using Matlab scripts. The procedure of image processing is illustrated in figure 2 where an exemplary raw image is shown in figure 2a. Prior to the image analysis a shading correction is done by subtracting a mean reference image measured at zero gas flow rate (figure 2b). As a next step a Gaussian filter is applied to the images to reduce the noise signal (figure 2c). Further, a

thresholding algorithm is applied to separate the individual bubbles and bubble clusters from the background. As a result, the images are converted to binary images where all pixels that belong to the bubbles are marked as 1, while the pixels marked as 0 correspond to the background (figure 2d). The corresponding parameters for the Gaussian filter applied to the optical measurements was chosen in such a way that the dimensions of the bright reflecting regions were reduced but no blurring of the bubble borders was caused (i.e. the Gaussian filter was applied using small values of the variance  $\sigma^2$ ). An appropriate threshold value was chosen which guarantees that all dark pixels in Figure 2c were counted as part of the bubbles. In turn, the parameters for the Gaussian filter and for the thresholding for GaInSn were obtained from a calibration measurement of two glass balls with 5 and 10 mm diameters surrounded by GaInSn at a zero gas flow rate. As a next step the binary images were analyzed using the function ‘*regionprops*’ integrated into Matlab which allows to directly extract parameters like perimeter, area, center of masses, etc. Figure 2e presents the raw image with the determined bubble perimeters and their center of mass and Figure 2f shows the raw image with fitted ellipses and their main axis. The obtained parameters were converted from pixel to metric values using the image scaling. The equivalent bubble diameters are then calculated from the bubble projection area assuming a spherical bubble shape. Additionally a ‘*Simple tracker*’ algorithm was implemented into the Matlab script. It allows to track the bubbles and, hence, to calculate the bubble velocities along their trajectories. All bubbles which move in close vicinity to each other or even overlap were excluded from further evaluation. The parameters derived from the X-ray radiography were compared to the parameters obtained from the optical measurements. Self-evidently, both measurements were performed in water applying the same process conditions.

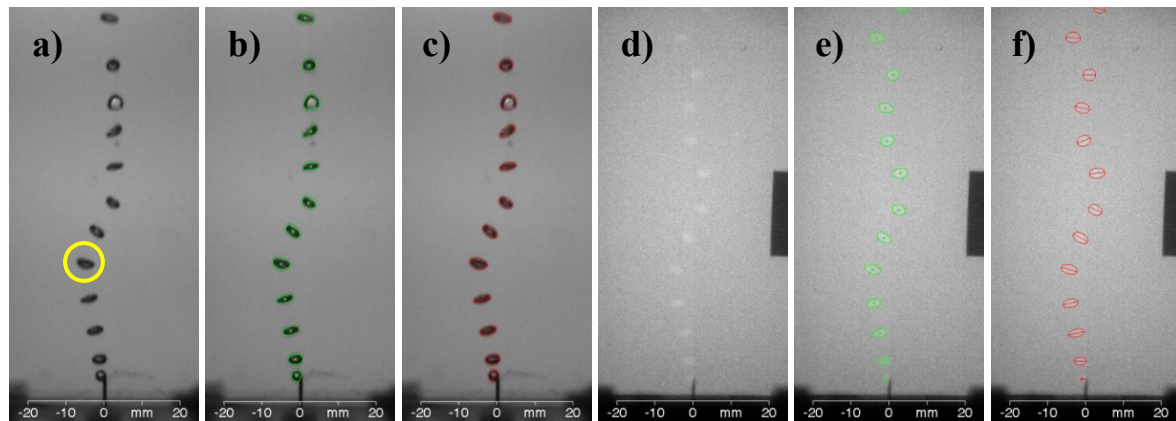


**Figure 2.** Image processing steps: a) raw image (cut from the full field of view), b) image with reduced reference image, c) gauss filtered image, d) binary image, e) raw image with delineated bubble interface and bubble centers, f) raw image with fitted ellipses and their main axis.

### 3. Results and discussion

#### 3.1. Validation of X-ray radiography in water

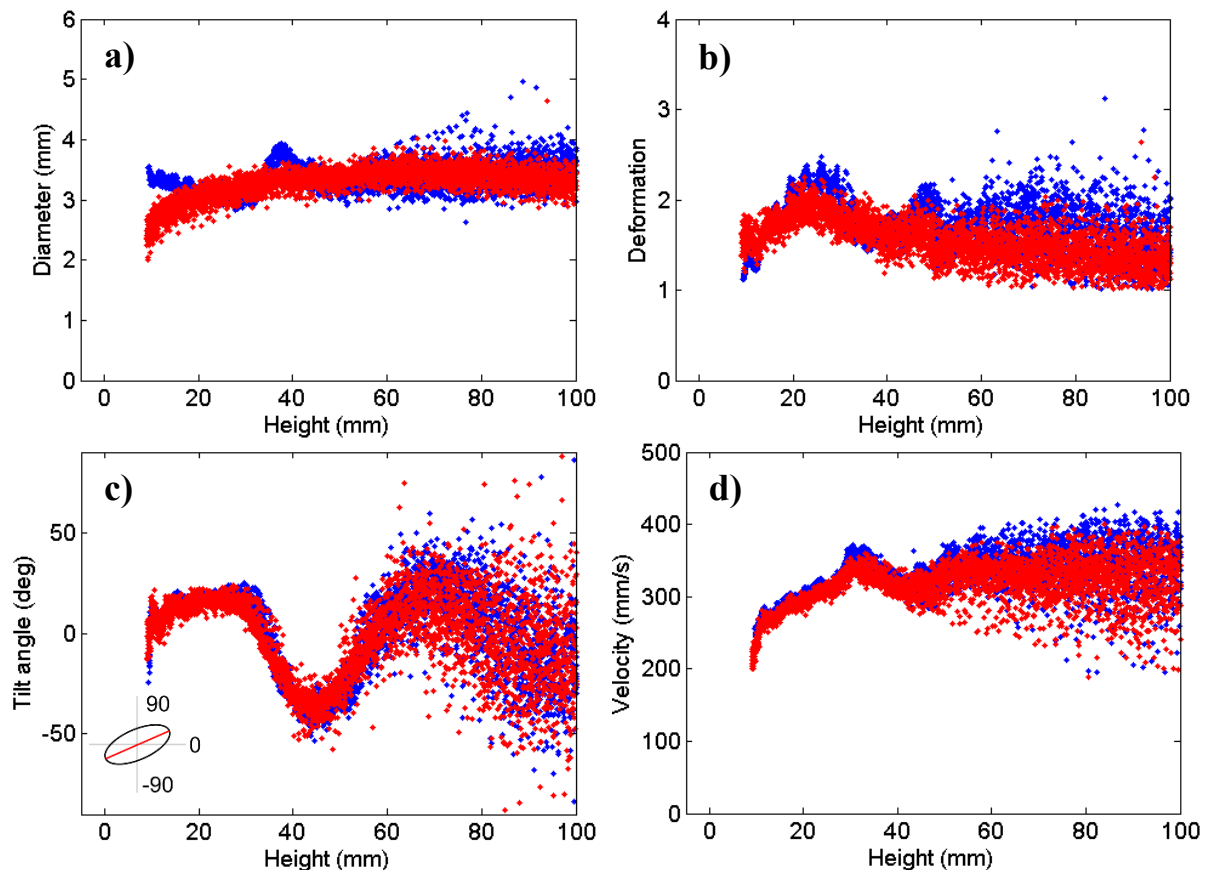
Figure 3 displays snapshots of bubble chains rising in water. The optically captured image is shown in figure 3a while X-ray image can be seen in figure 3d. The gas bubbles can be clearly identified in Figure 3d even though the X-ray image demonstrates a rather weak contrast between the bubbles and the background. The subsequent bubble analysis is performed according to the algorithm described in the previous section. The parameters for the Gaussian filter applied at the X-ray measurements were taken over from the optical measurements. The only difference was the threshold value, which was chosen so that all the bright pixels in Figure 3d were assigned to the bubbles. Figures 3b and 3e illustrate the raw images with the bubble interface and the bubble centers. Figures 3c and 3f present the raw images with fitted ellipses and their main axis.



**Figure 3.** Snapshots of the bubble experiment in water measured at an Ar gas flow rate of  $50 \text{ cm}^3/\text{min}$  by the optical method: (a) raw image, (b) raw image with delineated gas-liquid interface and bubble centers, (c) raw image with fitted ellipses and their main axis; and by means of X-ray radiography: (d) raw image, (e) raw image with delineated gas-liquid interface and bubble centers, (f) raw image with fitted ellipses and their main axis.

The corresponding sauter mean bubble diameters were calculated from the bubble projections according to the formula  $d = 2 \sqrt{S/\pi}$ , where  $S$  is the area covered by the bubble in the image. The results for bubble diameter, bubble velocity, etc. are shown in figure 4 where the blue and red data points correspond to the optical measurements and the X-ray radiography, respectively. The average bubble diameter derived from the optical measurements is  $\sim 3.4 \pm 0.3 \text{ mm}$  while a value of  $3.25 \pm 0.4 \text{ mm}$  was found for the X-ray measurements. These results are in a very good agreement and correspond very well to the value calculated directly from the bubble detachment frequency taking into account a gas flow rate ( $50 \text{ cm}^3/\text{min}$ ) and a number of 37 bubbles being ejected from the orifice per second ( $d \approx 3.5 \text{ mm}$ ). Figure 4a displays the evolution of the bubble size along the height. Remarkable differences can be observed near the nozzle and at a height of approximately 37.5 mm. Moreover, the bubble sizes obtained from the optical measurements in the upper part of the container are slightly larger than the ones obtained from the X-ray measurements. These differences could be explained by two reasons. First, the X-ray beam has a Gaussian shape showing a maximum at a height of 60 mm height. Therefore, the whole container is not illuminated homogeneously. Since the image thresholding is performed using a fixed value for the entire image the less illuminated bubbles provide a weaker signal and the algorithm might determine a too small bubble size. This problem explains the differences in the bubble size near the nozzle and in the upper part of the container near the free surface. Second, the interface between bubble and liquid is directly detected by the optical method whereas the X-ray radiography relies on the absorption contrast which is mainly determined by the local bubble cross-section along the X-ray beam direction. Therefore, it can happen that two bubbles of the same volume but different shapes (see the two examples on the left-hand side of figure 5a) will provide different X-ray signals. The corresponding X-ray intensity can be estimated using the Beer-Lambert law  $I = I_0 e^{-\mu x}$ , where  $I_0$  is the primary beam intensity,  $\mu$  is the X-ray absorption coefficient and  $x$  is the thickness of the liquid. Calculations of the X-ray intensity were carried out taking into account the values  $I_0 = 100$  and  $\mu = 0.1$  the latter is the water absorption coefficient for the X-ray energy of 320 KeV. As expected, the results show that bubbles with the larger thickness parallel to the X-ray beam (case A in figure 5) deliver a stronger X-ray signal compared to those with the smaller thickness (case B). A thresholding of both bubble signals using the same threshold value leads to a distinctly larger error in case B in comparison to case A. The consequence is that the total bubble size for case B will be underestimated. This effect might explain the differences in the bubble size at a height of approximately 37.5 mm. At that position the bubbles show significant increase of the size in the optical image (see the bubble highlighted in figure 3a by a circle). It is known from ultrasonic

measurements that significant deformations of the bubble occur after detachment of the bubbles from the injector [22]. These fluctuations of the bubble shape lead to a flattening of the bubble in the vertical plane. The increased cross-section detected by the camera simulates a supposed but not real increase in the bubble size

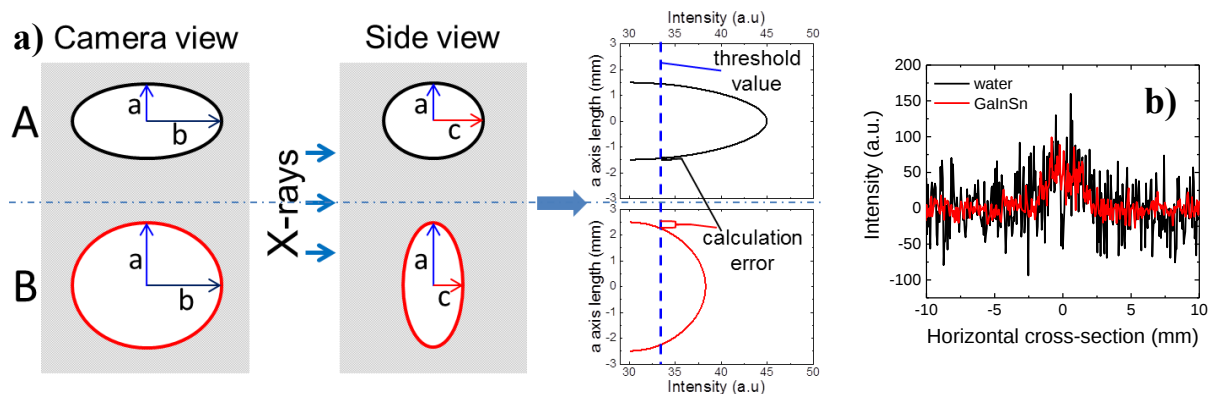


**Figure 4.** Bubble parameters obtained from optical measurements (blue) and X-ray radiography (red) in water: a) Bubble diameters, b) Bubble deformation, c) Bubble tilt angle, and d) Bubble velocity.

The bubble deformation is defined as the ratio between the length of the major and the minor axis of the ellipses. Corresponding results are shown in figure 4b. The bubble shape undergoes strong deformations along the whole bubble trajectory and reaches a maximum at the height of  $25 \pm 5$  mm. Another deformation peak is observed at a height of  $48 \pm 2$  mm. According to Bhaga [3] the bubble shape can be considered as an oblate ellipsoid with a wobbling surface. The bubble tilt angles (inclination angle of the major axis) and velocities are shown in figure 4c and 4d, respectively. The tilt angle reveals that the bubbles are detached from the nozzle with the main axis inclined positively to the horizontal line which is governed by the bevel shape of the injection nozzle. Then, the zig-zag motion of the bubble is starting fairly quickly. At a height of 29 mm the bubble orientation changes and the bubble tilt angle starts to decrease drastically till a turnaround point is reached at a height of 45 mm. Beyond the height of 68 mm the bubbles tilt angle data are strongly scattered but the zig-zag motion can be clearly identified. The data for the bubble velocity in figure 4d indicate a short acceleration phase just after detachment from the injection nozzle. The final velocity reaches values up to 400 mm/s with a pronounced peak at a height of 31 mm before the zig-zag motion appears. The scatter of the velocity data increases with increasing distance from the gas injection point.

Figure 5b illustrates the experimental X-ray signals from a bubble horizontal cross-section (black) at a height of  $\sim 25$  mm above the bottom of the container. It becomes obvious that the X-ray signal in

water is strongly affected by the noise. The corresponding signal-to-noise ratio calculated as  $SNR = (A_{signal}/A_{noise})^2$ , where  $A_{signal}$  is the signal amplitude and  $A_{noise}$  is the average noise amplitude, amounts to 1.3. Such a low signal-to-noise ratio is attributed to strong scattering of the X-rays in water. However, despite of the weak contrast between the bubble and the background and the related low signal-to-noise ratio the algorithm applied for the analysis of the X-ray images provides fairly accurate results with only slight discrepancy in the bubble size and shape (see figure 4). In conclusion, it can be successfully applied for the analysis of bubble rising in liquid metals where the higher X-ray attenuation of the metallic melt provides a better contrast and a better signal-to-noise ratio (see red line in figure 5b).

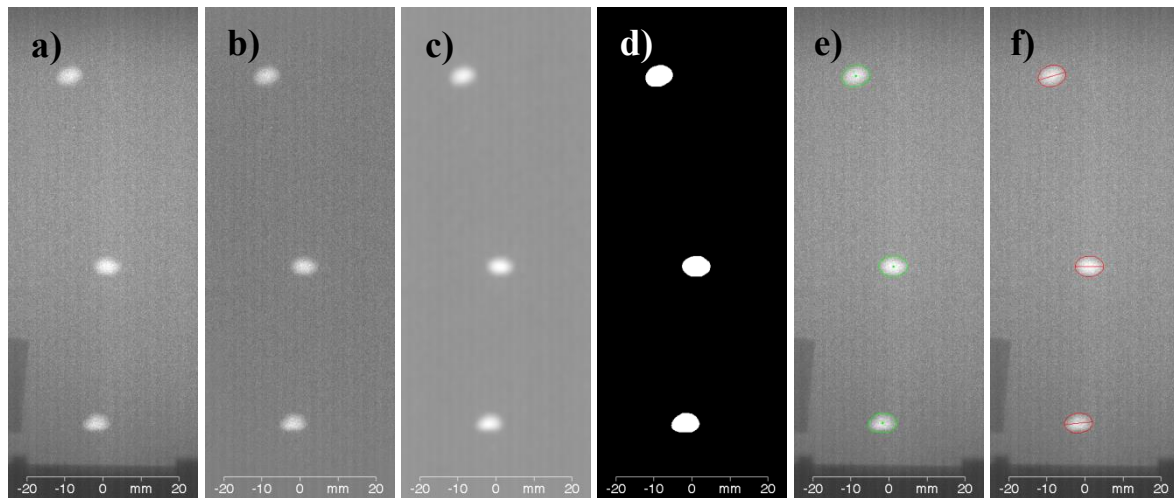


**Figure 5.** a) Camera view and side view of two bubbles of equivalent volume. Simulated X-ray intensity obtained from the case A (black) and case B (red) for the largest bubble cross-section parallel to the X-ray beam. b) X-ray signal from a horizontal cross-section of a bubble having a diameter of 3 mm diameter in water (black) and in GaInSn (red).

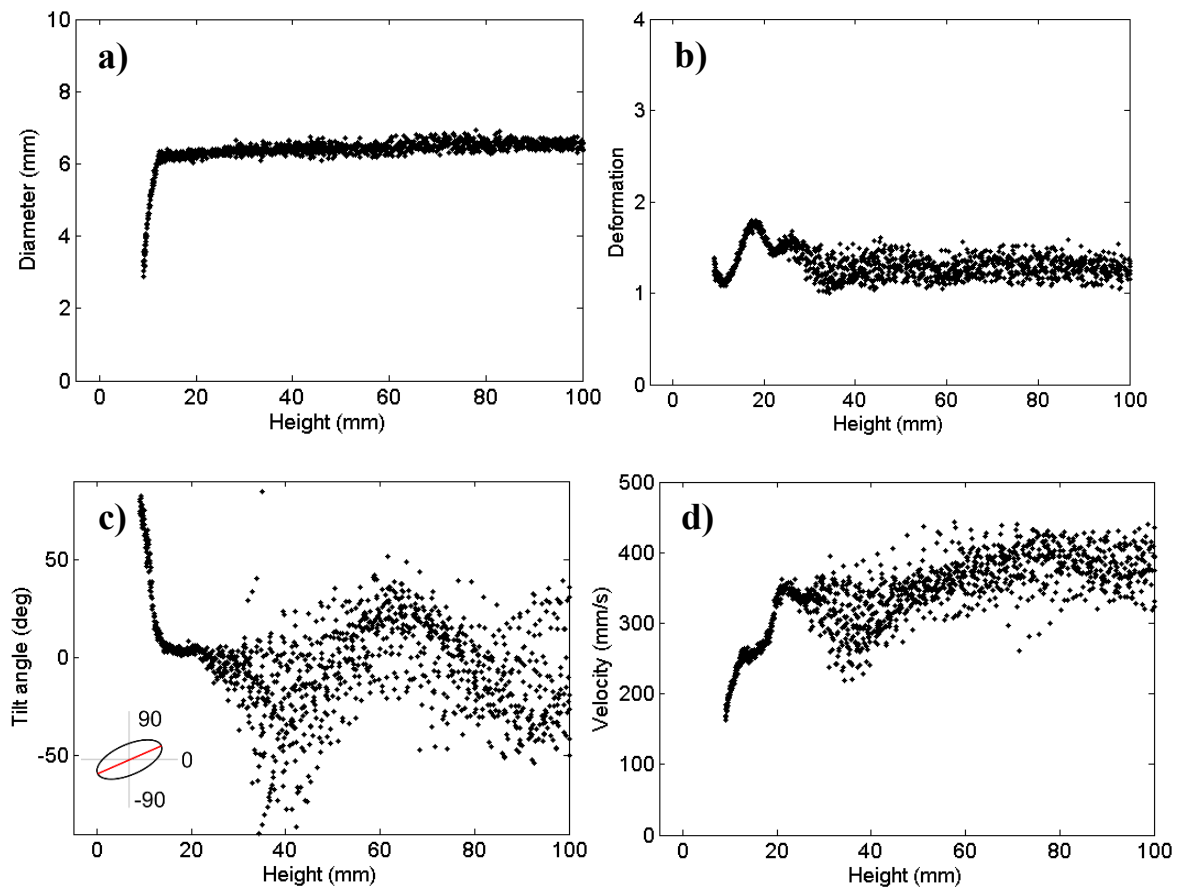
### 3.2. Comparison GaInSn versus water

This section aims to demonstrate the suitability of the X-ray radiography in the eutectic GaInSn alloy and to show some differences with respect to the bubble rising dynamics in water and GaInSn for a given Ar gas flow rate of 50 cm<sup>3</sup>/min. Figure 6 presents the X-ray radiography image analysis for bubbles in GaInSn according to the image processing steps described in section 2.2: a) raw image, b) image with reduced reference image, c) gauss filtered image, d) binary image, e) raw image with bubble boundaries and bubble centers, f) raw image with fitted ellipses and their main axis. In comparison to the situation in water the horizontal cross-section of a 3 mm bubble in GaInSn shows a much better signal-to-noise ratio of 14.7 (see figure 5b). Such a large value is rather convenient for further data processing. As a result the analysis to be done for the experiments in GaInSn should provide much more accurate values for bubble size and shape as in water where the validation of the method was carried out.

The experiments in GaInSn were performed using the same gas injector and the same gas flow rates as in water. The most striking difference is that only a number of 6 bubbles are injected per second for the same gas flow rate from the orifice in GaInSn while a bubble detachment rate of 37 bubbles/s was observed in water. The average bubble diameter is  $6.35 \pm 0.4$  mm which is twice as large as in water. The measured value is in a very good agreement with the value calculated from the gas flow rate and bubble detachment frequency ( $d \approx 6.42$  mm). The significant deviations with respect to the bubble number and size can be attributed to the strong differences in the surface tension and the wetting behavior at the injection nozzle [14].



**Figure 6.** Image processing steps demonstrated for an exemplary image in GaInSn: a) raw image, b) image with reduced reference image, c) gauss filtered image, d) binary image, e) raw image with bubble boundaries and bubble centers, f) raw image with fitted ellipses and their main axis.



**Figure 7.** Bubble parameters determined by X-ray radiography in the eutectic GaInSn: a) Bubble diameter, b) bubble deformation, c) bubble tilt angle, d) bubble velocity.

Bubbles undergo the maximum deformation just after the detachment from the nozzle. The less scattering of the data points for the bubble deformation (see figures 4b and 7b) is also a direct result of the larger surface tension for GaInSn which prevents significant surface wobbling. During the initial stage of their rise the bubbles show a distinct deformation and can be described as ellipsoids but on their further trajectory the bubbles approach almost spherical shape: the deformation tends to become close to 1 at container heights above 30 mm. When leaving the nozzle the bubble main axis is almost parallel to the bottom of the container. Strong variations start beyond a height of 25 mm. At the positions below 10 mm the bubbles are still attached to the nozzle. This elongation of the bubble along the vertical direction explains the large tilt angles in figure 7c. The strong scattering of the tilt angle data at the upper part of the container is caused by the almost spherical bubble shape where all diagonals are almost equivalent. The almost spherical bubble shape impedes the accurate determination of the main axis by the Matlab algorithm. The bubble velocity in the GaInSn melt reaches values up to 400 mm/s with a pronounced peak at a height of  $21 \pm 1$  mm.

As a summary, it can be concluded that the applicability of X-ray radiography for quantitative measurement of bubble parameters rising in a stagnant liquid was successfully validated by parallel optical measurements in water. Investigations in liquid metals benefit from a better signal-to-noise ratio due to better X-ray contrast. The experiments showed that for a chosen Ar gas flow rate the total number of injected bubbles is much larger in water than in GaInSn. Likewise, the bubble deformation is larger in water than in GaInSn. The bubbles in GaInSn tend to have a more spherical-like shape while the bubbles in water can be described as oblate ellipsoids with a wobbling surface. The main reason for these effects is the higher surface tension in GaInSn in comparison to water. The rising velocities of the gas bubble are found to be rather similar both water and GaInSn. The higher buoyancy in the liquid metal is obviously compensated by the increased drag force owing to the larger bubble size [10].

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