Generalisation of the Critical Angle Refractometry for the characterisation of clouds of bubbles

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Abstract The principle of the Critical Angle Refractometry is extended to characterize simultaneously the size distribution and the relative refractive index (i.e. composition) of a cloud of particles with relative refractive index below unity (i.e. bubbles). For the description of the near-critical-scattering pattern, the advantages and limits of various models and theories are compared and discussed: geometrical optics, physical optics approximation, the Lorenz-Mie and the complex angular momentum theories. Numerical investigations show that the statistical properties of a cloud of bubbles: mean refractive index, mean and standard deviation of the size distribution, have a strong influence on the global angular position, the angular spreading and the amplitude contrast of the near-critical-scattering pattern respectively. This behaviour allows developing a simple method to inverse the scattering pattern. Preliminary experimental results confirm the great potentialities of the proposed technique.

1. Introduction

Bubbly flows occur in many industrial and natural processes, such as boiling heat transfer, cloud cavitation in hydraulic systems, aeration and stirring of reactors, aeration in water purification, bubble columns and centrifuges in the petrochemical industry, scavenging of dissolved gases, the exchange of gases and heat between the oceans and the atmosphere, and explosive volcanic eruptions. Understanding the evolution and properties of bubbly flows is, therefore, of major technological as well as scientific interest (Bunner and Tryggvason, 1999). For this reason, experimental data and diagnosis tools of bubbly flows are still required.

From the optical point of view a bubble is a particle, with a refractive index \( m(\lambda_0) \), surrounded by a medium with a larger refractive index \( m'(\lambda_0) \). Its relative refractive index is \( m_r(\lambda_0) = m / m' < 1 \) and a bubble can be either a gas particle in a liquid (i.e. air/water, \( m_r \approx 0.75 \)), a liquid particle in a liquid (i.e. water/oil, \( m_r \approx 0.88 \)) or a solid particle in a liquid (i.e. PPMA/heavy oil, \( m_r \approx 0.98 \))… Various laser techniques (Xu, 2001) can be used or have been specially developed to characterise the size of a single bubble (Phase Doppler anemometry, critical scattering, optical probes, DDPIV…) or the size distribution of a cloud of bubbles (laser diffractometry, light extinction…). Some of them, like the Phase Doppler anemometry (Gréhan et al., 1996) or more recently the DDPIV (David et al., 2002) allow to measure the bubble’s velocity and then, the dynamic of the bubbly flow. Nevertheless, except the critical scattering, the aforementioned techniques give no information about the composition of the bubbles or the surrounding medium, which may be an important parameter when considering mixing or coalescence phenomena for instance.

The Critical Angle Refractometry was developed originally for the characterisation of the size of a single bubble of known composition (Langley and Marston, 1984; Marston P. L. and Kingsbury D. L., 1981; Marston, 1979) and afterwards, to determine simultaneously the bubble size and its relative refractive index, i.e. composition (Onofri F., 1999). Theses information are obtained from
the analysis of the angular spacing of the fringes (§ 2.2) observed around the critical angle, when a bubble is passing through a small optical probe volume (whose dimensions are comparable to the mean size of the bubbles under study). To obtain the size distribution and the statistical moments, a temporal integration is required, which may be a limiting point for some applications. In addition, the critical scattering diagrams are noisy and difficult to analyse, like the rainbow scattering patterns (Massoli et al., 1993; Roth et al., 1991; Sankar et al., 1993; van Beeck, 1997; van Beeck and Riethmuller, 1994).

In the present paper, we introduce a generalisation of the Critical Angle Refractometry for the simultaneous characterisation of the size distribution and the composition of a cloud of bubbles. For this purpose, the incoming laser beam is expended to generate a probe volume of few cubic centimetres, so that multiple bubbles are illuminated at the same time. Section 2 describes the scattering models developed to predict the scattering pattern observed under a single scattering regime assumption. Section 3 presents some numerical results showing the influence on the critical scattering pattern of the bubble clouds mean size, dispersion and refractive index. Section 4 presents some preliminary experimental results. Our conclusions are given in Section 5.

![Diagram](image.png)

(a) Sketch of the rays scattered in the near-critical-angle scattering region and b) corresponding scattering diagram according to the Lorenz-Mie Theory (LMT), the Physical Optics Approximation (POA) and Geometric Optics (GO).

### 2. Scattering of light by a single bubble, near the critical angle

From Descartes’ refraction optics laws we know that in the case of light rays passing from a medium of higher refractive index \( m' \) to a medium with a lower refractive index \( m = m / m' < 1 \), there exist an angle of incidence \( \phi = \sin^{-1}(m) \), leading to an abrupt transition to total reflection for \( \phi > \phi_c \), Fig. 1 a). In the case of light rays incident on a spherical bubble, with radius \( a \), the same phenomenon occurs but for a curved surface (Fiedler-Ferrari et al., 1991), giving rise, near the so-called “critical angle”, to a complex scattering pattern around the scattering angle \( \theta_c = \pi - 2\phi_c \). The scattering pattern of an air bubble in water (\( m_r = 1.0/1.332 \)) with diameter \( D = 100\mu m \), a plane wave with parallel polarization and wavelength \( \lambda = 0.488\mu m \), is shown in Fig. 1b). Different approaches are used to predict this pattern: a) the Lorenz-Mie Theory (LMT), which is perfectly rigorous (Bohren and Huffman, 1998; Gouesbet et al., 1988; Onofri et al., 1995); Geometrical Optics (GO) when only reflected (\( p=0 \)) and refracted (\( p=1 \)) processes are considered (Davis G.E., 1955; van de Hulst, 1957), the Physical Optics Approximation (POA, §2.2) with the reflected light (\( p=0 \)) only or
the reflected and the refracted light ($p=0$ and $p=1$). LMT predicts a scattering diagram which is characterized by strong oscillations: a coarse structure (large ‘bright’ and ‘dark’ fringes) superimposed on a fine structure (small amplitude and high frequency fringes). Note that the critical scattering is highly polarization sensitive. The parallel polarization is the one which gives the most contrasted scattering pattern so that, in the following, all results are given for this polarization state.

### 2.2 Physical optics Approximation (POA)

Marston et al. (Langley and Marston, 1984; Marston P. L. and Kingsbury D. L., 1981; Marston, 1979) have developed a so-called « Physical Optics Approximation » (POA) where the contribution from surface reflection is treated by a procedure similar to Airy’s theory of the rainbow (Airy, 1838): a Kirchhoff-type approximation is applied to the amplitude distribution along a virtual reflected wavefront. The reflection contribution ($p=0$) is approximated as a step function. This « edge reflectivity » (Fiedler-Ferrari et al., 1991) gives rise to an angular distribution of scattered intensity similar to a Fresnel straight-edge pattern, which accounts for the diffraction fringes of low angular frequency. Similarly to the rainbow phenomenon, the fine structure is shown to be unrelated to the critical scattering. It has to do with an interference phenomenon occurring between near side and far side refracted rays ($p=2, 2'$, Fig.1 a).

In the POA, only the reflection ($p=0$) and the refraction ($p=1$) are taken into account for the calculation of the amplitude functions:

$$ S = S_0 + S_1 + S_2 + S_3 + ... $$

(1)

The amplitude functions read as:

$$ S_p = \frac{-ika}{2} F_p \exp\left[i\gamma_p\right] $$

(2)

where $k$ is the wave number, $k = 2\pi / \lambda$, with $\lambda = m_r\lambda_0$. The total scattered intensity is $I = |S|^2 (2/ka)^2$.

**Reflected contribution ($p=0$)**

As mentioned above, in the POA analysis, the reflection process is treated as a diffraction process, with Fresnel integrals (far-field intensity of the virtual wave front near the critical angle). This analysis leads, for the amplitude of the scattering function of the reflected/diffracted light, to:

$$ F_0 = |F(\omega) - F(-\infty)\exp[-i\pi/4]|/\sqrt{2} $$

(3)

where $\omega = \sin(\eta)\sqrt{(a/\lambda)\cos\phi}$ expresses the dependence of the critical scattering pattern with the particle’s radius, its relative refractive index and the incident wavelength. The angle $\eta = \theta - \theta_c$ is a deviation from the position of the critical angle predicted by geometrical optics, with $\theta = \sin^{-1}\left(\omega/\sqrt{(a/\lambda)\cos\phi}\right) + \theta_c$. The oscillatory function $F(\omega)$ is defined with the Fresnel’s cosine and sine integrals (Abramowitz and Stegun, 1964; Chang et al., 1996):

$$ F(\omega) = \int_0^\omega \exp\left(i\frac{\pi}{2}z^2\right)dz = C(\omega) + iS(\omega) = \int_0^\omega \cos\left(i\frac{\pi}{2}z^2\right)dz + i\int_0^\omega \sin\left(i\frac{\pi}{2}z^2\right)dz $$

(4)

The phase term in Eq. (2) can be deduced from geometrical and physical considerations:

$$ \gamma_0 = -2ka\cos(\tau_o) - \Lambda[\theta_0 - \theta_c]\delta_0 $$

(5)

$\Lambda$ is the Heaviside function with, $\Lambda[\theta_0 - \theta_c]=0$ for $\theta > \theta_c$ and $\Lambda[\theta_0 - \theta_c]=1$ for $\theta \leq \theta_c$. In Eq. (5) the first term comes from the classical phase delay associated to the path of geometrical rays, with $\tau_o = (\pi - \theta_0)/2$ and $\rho_0 = \tau_o$ (van de Hulst, 1957). The second term $\delta_0$ is a phase delay associated to the tunnelling effect (Langley and Marston, 1984; Löttsch, 1971):
\[ \tan(\delta_0/2) = m^2 \sqrt{\sin^2 \tau_0 - m^2 \cos \tau_0} \] (6)

If only the reflected/diffracted process is taken into account in Eq. (1), the near-angle-critical scattering intensity is of the following form:

\[ I_0(\theta, m_r, D) \propto \left[ C(\omega) + 1/2 \right]^2 + \left[ S(\omega) + 1/2 \right]^2 = H(\omega) \] (7)

Looking for the angular position of the bright and dark fringes of the critical scattering pattern is equivalent to look for the zeros \( \omega_n \) of the first derivative of \( H(\omega) \). They can be found numerically, see table 1. If the relative refractive index \( m_r \) is prior known (i.e. bubble and surrounding medium composition), we have shown in a previous work (Onofri F., 1999) that one can deduce the bubble radius from the measurement of the angular position of the \( n^{th} \) fringes

\[ a_n = \frac{\lambda_r m_r \omega_n^2}{\sin^2(\theta_c - \theta_n) \cos \phi_c} \] (8)

When both the bubble relative index and radius are not prior known, they can be deduced from the measurement of the angular position of two fringes \( n = p, q, q>p \) of the critical scattering pattern (Onofri F., 1999) with:

\[ m_{r,pq} = \sin^3 \left[ \frac{1}{2}(\pi - \theta_p - \Omega_{pq}) \right] \] (9)

\[ a_{pq} = \frac{\lambda_r m_{r,pq} \omega_q^3 + \omega_q^3 - 2 \omega_p \omega_q \cos \Delta_{pq} \sin^2(\Delta_{pq}) \sin \left( (\theta_p - \Omega_{pq})/2 \right)}{\sin^2(\Delta_{pq})} \] (10)

with \( \Omega_{pq} = \tan^{-1}\left[ \sin(\theta_q - \theta_p) / \left( \cos(\theta_q - \theta_p) - \omega_q / \omega_q \right) \right] \) and \( \Delta_{pq} = \theta_q - \theta_p \).

Compared with GO predictions, the first term of the POA provides a significant improvement in the description of the near-critical-scattering pattern, Fig. 1 b). The angular position of the first two extremes (i.e. \( \theta_1, \theta_2 \)) are in rather good agreement with the ones predicted by LMT. This is confirmed by Fig. 2 which presents POA and LMT’s predictions for \( \theta_1 \), various bubbles sizes and relative refractive indices. Nevertheless, the first term of the POA gives a very poor estimation of the angular position of the other fringes (\( \theta_n \geq \theta_2 \)) and of their relative amplitude, Fig. 1b).
Table 1. Tabulated zeros of $H(\omega)$

<table>
<thead>
<tr>
<th>$j$</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\omega$</td>
<td>1.2171983</td>
<td>1.8725191</td>
<td>2.3448538</td>
<td>2.7390081</td>
<td>3.0881958</td>
<td>3.3913356</td>
<td>1.2171983</td>
</tr>
</tbody>
</table>

Refracted contribution ($p=1$)

Following, van de Hulst (van de Hulst, 1957), the phase and amplitude of the contribution of the refracted process ($p=1$) read as:

$$\gamma_{ij} = 2ka \left( m \cos \rho_i - \cos \theta \right)$$  \hspace{1cm} (11)

$$F_1 = 2 \left( 1 - r_1^2 \right) \sqrt{D_i A [\theta_i - \theta_1]}$$  \hspace{1cm} (12)

$D_i$ is a divergence function which takes into account the effect of the particle surface curvature on the Fresnel coefficients, $r_i$, with $D_i = \sin \tau_i \cos \tau_i / \left( 2 \sin \theta \left| 1 - m_r^{-1} \cos \tau_i / \cos \rho_i \right| \right)$. For the parallel polarisation we have $r = \tan \left( \tau_i - \rho_i \right) / \tan \left( \tau_i + \rho_i \right)$. $\tau_i$ and $\rho_i$ are the angles between the incident ray and the bubble surface, the bubble surface and the internally refracted ray (Langley and Marston, 1984; van de Hulst, 1957). These two angles can be expressed as functions of the scattering angle of the refracted rays ($p=1$):

$$\tau_i = -\frac{\theta}{2} + \tan^{-1} \left( \frac{m_r^{-1} \sin \left( \theta_i / 2 \right)}{m_r^{-1} \cos \left( \theta_i / 2 \right) - 1} \right)$$

$$\rho_i = \sin^{-1} \left( m_r^{-1} \sin \tau_i \right)$$  \hspace{1cm} (13)

Fig. 1 b) and Fig. 3 show the near-angle-scattering pattern obtained with POA when the reflected/diffracted ($p=0$) and the refracted ($p=1$) terms are both included or not. Clearly, adding the refracted term significantly improved the prediction of the relative amplitude of the fringes (at least up to $\theta_0$) and it reduces the shift observed with LMT predictions for the first fringes. Nevertheless, there is no improvement of the asymptotic behaviour of the scattered intensity for $\theta > \theta_1$, see Fig. 3.

2.3 Complex Angular Moment theory (CAM)

Fiedler-Ferrari, Nussenzweig & Wiscombe (Fiedler-Ferrari et al., 1991) have developed a zero order approximation of the near-critical-angle scattering from a curved interface. It is based on the Complex Angular Moment theory (CAM). The description of the CAM theory is clearly beyond the scope of this paper. Our objective, here, is to evaluate the potentialities of this theory in order to...
predict, and inverse, the critical scattering pattern. Nevertheless, in few words, let say that the CAM
theory is based on i) the scattering particle is described as an effective potential: a Debye
electromagnetic potential (Fiedler-Ferrari et al., 1991; Nussenzveig and Wiscombe, 1980;
Nussenzweig, 1992); ii) the concept of the localization principle (van de Hulst, 1957) according to
which, the electromagnetic field expansions term, $n$, in the LMT, can be associated with incidents
rays having an impact parameter $n + 1/2 = x = \pi D/\lambda$, see Fig. 1a). Fig. 3, compares the predictions
of the CAM approximation (only for the rays $p \leq 2$), LMT, POA ($p=0 \& p=1$) and GO ($p=0$ only).
The numerical results of the CAM theory were obtained with a Fortran code available on the
Warren J. Wiscombe’s ftp site (Wiscombe, 2006). Nevertheless, to run this code, the present
authors were obliged to modify the subroutine that calculates the Airy function with large complex
arguments. The CAM approximation provides a good description of the coarse structure of the
critical scattering pattern of large bubbles (i.e. $D=1000 \mu m$, size parameter $\beta = \pi D/\lambda \approx 8581$),
Fig. 3. But, for smaller bubbles size (i.e. $D=50 \mu m$, size parameter $\beta \approx 429$), strong discrepancies
appear between the CAM approximation and LMT. This last results, is not totally surprising and
was already noticed by (Fiedler-Ferrari et al., 1991).

To conclude on these results, it is clear that only the results obtained with the LMT can be
considered as reliable, but LMT calculations are time consuming. The POA provides analytical
expressions and allows fast calculations but it gives only a rough estimation of the main
characteristics of the near-critical-angle scattering pattern. From a metrological point of view,
the interest of the CAM approximation is found to be quite limited as large bubbles are usually not
spherical.

3. Scattering of light by a cloud of bubbles, near the critical angle

3.1 Model

Classically, in the limit of a single light scattering regime and for particles randomly positioned
in space, the scattering of a cloud of particles can be approximated, in the far field region, by the
incoherent summation of the contributions of all particles that are illuminated. In this study, the
cloud of bubbles is only characterized by a mean relative refractive index $m_r$ and a log-normal
probability density function $f(D)$ for the size distribution, with mean size $\bar{D}$ and a standard
deviation $\sigma_D$. Using the Lorenz-Mie theory, the POA or the CAM… to calculate the light intensity
scattered by a single bubble $I(\theta, D, m_r, \lambda_0)$, the intensity scattered by the cloud of bubbles is

$$\langle I(\theta, \bar{D}, \sigma_D, m_r, \lambda_0) \rangle = \frac{1}{D_{\text{max}}} \int_{D_{\text{min}}}^{D_{\text{max}}} I(\theta, D, m_r, \lambda_0) f(D) dD$$

(14)

with for the Log-Normal distribution of parameters $(m_D, s_D)$:

$$f(D) = \frac{1}{Ds_D \sqrt{2\pi}} \exp \left[ -\frac{1}{2} \left( \ln D - m_D \right)^2 \right]$$

(15)

with the relations between $(m_D, s_D)$ and $(\bar{D}, \sigma_D)$:

$$s_D = \sqrt{\ln \left( \sigma_D^2 / \bar{D}^2 \right)} + 1 \quad m_D = \ln \bar{D} - s_D^2 / 2 \quad D_{\text{max}} = \exp \left( m - s_D^2 \right)$$

(16)

$$\bar{D} = \exp \left[ m_D + s_D^2 / 2 \right] \quad \sigma_D = \exp \left[ 2m_D + s_D^2 \right] \left( \exp \left( s_D^2 \right) - 1 \right)$$

(17)

In the following, the integral limits $D_{\text{min}}$ and $D_{\text{max}}$ of Eq. (14) are set equal to the solutions
of $f(D)/\max\{f(D)\} = 1/1000$. 

- 6 -
Fig. 4. Influence of the bubbles mean size $D$ of a cloud of bubbles, on the near-critical-angle scattering: a) scattering diagrams and b) evolution of the basic scattering diagrams properties.

Fig. 5. Influence of the width of the size distribution of a cloud of bubbles, on the near-critical-angle scattering: a) scattering diagrams and b) evolution of the basic scattering diagrams properties.

Fig. 6. Influence of the relative refractive index of a cloud of bubbles, on the near-critical-angle scattering: a) scattering diagrams and b) evolution of the basic scattering diagrams properties.
3.2 Numerical results

For a given size distribution, the near-critical-angle scattering diagrams can be calculated with LMT, for various mean sizes \((D=25-1000\mu m)\), size distribution widths \((\sigma_D/D=0-100\%)\) and refractive indexes \((n/n'=1.32-1.52)\), see Figs. 5-6.

**Influence of the mean size**

Fig. 4 a) shows the critical scattering diagrams produced by clouds of bubbles with a Log-Normal size distribution. The relative refractive index and the size standard deviation are fixed with \(m_r=1.334\) and \(\sigma_D=0.05D\) respectively. The mean diameter increases from \(D=12.5\mu m\) up to \(D=800\mu m\). Clearly, the main influence of the mean diameter is in the angular spreading/compression of the whole critical scattering diagram. Fig. 4 b) shows the evolution of the angular spacing of fringes \(\theta_1\) and \(\theta_2\), \(\Delta\theta_{12}=\theta_1-\theta_2\), which decreases rapidly with an increasing mean size. This parameters, as well as the asymptotic parameter \(\chi_1\), can be used to deduce the size distribution mean diameter.

**Influence of the width of the distribution**

Fig. 5 a) shows the influence of the bubbles size standard deviation on the critical scattering pattern, when the mean diameter and the refractive index are fixed: \(\bar{D}=100\mu m\) and \(m_r=1.334\). The main influence of \(\sigma_D\) is in the fringes contrast, or “visibility” (relative intensity of the first bright and dark fringes, \(V=(I_1-I_2)/(I_1+I_2)\)). This visibility decreases rapidly with an increasing size distribution width, Fig. 5 b), so that it can be used to infer the size distribution width.

**Influence of the refractive index**

Fig. 6 a) shows the influence of the bubbles relative refractive index when the size distribution is fixed, \(\bar{D}=100\mu m\) and \(\sigma_D=0.25\bar{D}\). Here also, this parameters has a well defined influence of the critical scattering pattern: it controls the angular position of the whole pattern. The scattering diagrams appear to be just “translated” by a change in refractive index.

4. Experiments

4.2 Experimental setup

The setup described in this section provides absolute-angle relative intensity measurements of the far field scattering from multiple bubbles in the critical angle region, Fig. 7. The cell in which the scattering takes place is, basically, a rectangular aquarium (300x200x200mm) with glass walls, filled with bi-distilled water or other fluids. The incident light, from a 2W Argon laser, is expanded before entering the cell, to get a collimated beam with a diameter of 12 mm, a parallel polarization and a wavelength \(\lambda_0=0.488\mu m\). The bubbles are produced by a porous medium, supplied with an air pump, which can generate single columns of bubbles (almost monodisperse) or a dense bubbly flow. The scattering of the bubbles located in the ‘probe volume’ is collected with a Fourier lens \((f/\phi=1.5)\) whose optical axis is perpendicular to the cell’s wall. An optical diffuser is placed in the collection lens Fourier plan, to get an image of the far-field scattering pattern. This pattern is recorded with a 1 Mpix 12 bits B/W digital camera and analysed with specially developed software. Since the camera is in air, refraction at the viewing wall has to be taken into account in determining the absolute scattering angles. An angular/pixel calibration of the camera is also necessary. For this
purpose, we have used a motorized microstep goniometer (resolution $\delta \theta \approx 0.01^\circ$) and taken advantage of the laser beams reflections on the glass walls. For a water solution, the angular range of the system is of $\theta = 62.55^\circ - 86.83^\circ = 24.28^\circ$, and the angular resolution of $\approx 0.02^\circ / \text{pix}$. It is important to remark that only the measurable angular range is fluid refractive index dependent, not the pixel-angular calibration of the system. Therefore, there is no need to calibrate afterwards the system when, for instance, the fluid’s composition or the temperature are changing.

Fig. 7. Experimental setup

4.2 Preliminary experimental results

Fig. 8 presents experimental scattering patterns recorded for a a) low and a b) high air supply flow rate, i.e. a small and high bubbles number density. Fig. 9 presents shadow images of the corresponding bubbly flows. The fluid is composed of bi-distilled water at ambient temperature.

Case a) corresponds to the scattering pattern produced by a single column of bubbles. For this reason, one can expect a quite narrow size distribution for theses bubbles. This is confirmed by Fig. 8 a) as well as Fig. 9a). In fact, from the previous numerical simulations we know that a large numbers of critical fringes (i.e. high visibility) is a signature of a size distribution with a small standard deviation. Fig. 10a) compares the experimental intensity profile of Fig. 8 a) and numerical simulations (LMT and POA). For LMT, the best agreement is found for $\bar{D} = 675\mu m$, $\sigma_D = 7\mu m$ and $m_r = 1.334$. Theses values are very close to the expected ones: $\bar{D} \approx 650\mu m$ (photographic diameter) and refractive index $m_r = 1.335$ (Abbe refractometer). Note that the values found with the first term of the POA, Eqs (9)-(10), are also correct ($\bar{D} = 656\mu m$, $m_r = 1.338$).

Case b) corresponds to a much higher bubbles number density (we were not able to measure this parameter). The bubbles are also larger and they can differ significantly from the assumed spherical shape, see Fig. 9 b). Nevertheless, few critical scattering fringes can be observed, see Fig. 8 b). In Fig. 10b) and for LMT, the best agreement is found for $\bar{D} = 1180\mu m$, $\sigma_D = 225\mu m$ and $m_r = 1.334$. Theses values are rather close to the expected ones: $\bar{D} \approx 1050\mu m$ (photographic diameter) and refractive index $m_r = 1.335$ (Abbe refractometer). For this last case, the agreement found between the theory and these preliminary experimental results is a little bit surprising when considering the non sphericity of the measured bubbles, Fig. 9 b). Nevertheless, thinking about the first term of the POA, it is clear that the critical scattering pattern is a local phenomenon occurring in the scattering plan. So that, the size measurement should be considered as a local measurement of the bubble’s surface curvature in the scattering plane, see Fig. 9 b). It is also the reason why, the critical scattering pattern was recorder in a horizontal plan.
Fig. 8. Experimental near-critical-angle scattering patterns obtained for a) low and b) high bubbles number density.

Fig. 9. Shadow images of some bubbles corresponding to the cases considered in Fig. 8.

Fig. 10. Comparison of experimental and numerical critical scattering intensity profiles.
5. Conclusion

The principle of the Critical Angle Refractometry has been extended to allow the simultaneous and instantaneous characterization of the size distribution (mean and standard deviation) and the relative refractive index of a cloud of bubbles. The Lorenz-Mie theory appears to be still the most reliable tool to predict the critical scattering pattern, whatever the Physical Optics Approximation can be used to give a first estimation of the bubble clouds properties. This technique is thought to have great potentialities to characterize on-line the size distribution or the change in composition of bubbly flows, provided that the surrounding fluid remains homogeneous. Future work will concern the modelling of the influence of the bubbles non sphericity and in additional experiments with statistical results.

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References


