Critical angle refractometry and sizing technique for bubbly flows characterization: particular effects

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Abstract  The Critical Angle Refractometry and Sizing technique (CARS) allows characterizing simultaneously and “instantaneously” bubble size distribution and composition. In the present study, we report particular effects that were previously considered as negligible: Gaussian beam and spatial filtering effects, interferences effects between dilute and dense aggregates of bubbles, bubbles non sphericity. Experimental results clearly demonstrate that the CARS technique is a robust technique that can already be used to characterize real bubbly flows.

1. Introduction

Under a single scattering assumption and for bubbles randomly distributed in space, the collective Critical Scattering Pattern (CSP) produced by a cloud of bubbles can be modeled by an inhomogeneous Fredholm equation of the first kind:

$$\mathcal{T} = N \int_0^\infty I(\theta, D, m_\lambda) f(D) dD \quad (1)$$

where $f(D)$ and $N$ are the bubble size distribution (BSD) and number concentration respectively, $I(\theta, D, m_\lambda)$ represents the intensity scattered by a single bubble with diameter $D$ and relative refractive index $m = m_p / m_s$, $m_p$ and $m_s$ are the bubble material and the surrounding medium refractive indices for the laser wavelength $\lambda_0$. For spherical bubbles, $I(\theta, D, m_\lambda)$ can be determined using the Lorenz-Mie (LMT, e.g. Bohren and Huffman 1998) or the Complex Angular Momentum (CAM, Nussenzveig and Wiscombe 1980) theories, a physical optics approximation (POA, Marston 1979, Onofri 1999).

The CARS technique (Onofri 1999; Onofri et al. 2007) aims to find $f(D)$ and an average relative refractive index $m$ from the measurement of $\mathcal{T}$ and the knowledge of the continuous kernel function $I$. This problem is predetermined, ill-posed and requires a specific inversion procedure (e.g. Lawson and Hanson 1974). In previous works, different inversion methods based on dependent (referenced as Log-Norm, Onofri et al. 1997) and independent (referenced here as NNLSQ, Onofri et al. 2009, Krzysiek 2009) models have been developed and evaluated. Indeed, Eq (1) is a classical integral for optical particle characterization (OPC) techniques based on the analysis of collective ensemble scattering. It is interesting to notice that Eq (1) differs only from the laser diffraction or the rainbow techniques integral equation, by the shape of the kernel function shape,

![Fig. 1 Intensity of the electromagnetic near-field inside and around an air bubble in water ($D=100\mu m$, $m_p=1.0$, $m_s=1.333$) calculated with LMT for a parallel polarized plane wave ($\lambda=0.532\mu m$).](image-url)
i.e. the underlying optical phenomenon. For the CARS technique the basic phenomenon is a semi-
discontinuity of the amplitude coefficient of the rays reflected onto optical bubbles \((m < 1)\). This
process generates, in the far-field, a critical-scattering pattern characterized by circular fringes on
the bubbles side-ways (Krzysiek, 2009). As an example, Fig. 1 shows the intensity of the
electromagnetic near-field inside and around an air bubble in water, for a parallel polarized plane
wave. The far-field critical-scattering pattern (CSP) is generated by the rays that are reflected with
impact parameters above \(x = mD/2\).

Whatever the CARS technique capabilities and resolution have been validated with success on
various bubbly flows (Onofri et al. 2009, Krzysiek 2009), in Eq. (1) there are still many underlying
hypotheses, that are commonly accepted for OPC techniques, but rarely evaluated and tested. So,
this paper aims to bring physical insights about these hypothesis and mainly: bubbles relative
refractive index, Gaussian beam and spatial filtering effects, interferences effects between spatially
close bubbles, bubbles non sphericity.

2. Experimental setup and procedure

The setup allows absolute angle measurements of the far-field CSP generated by all bubbles present
in a optical probe volume of about 1 \(cm^3\), see Fig. 2. The scattering takes place in a
300x100x100mm rectangular glass-walled aquarium (cell) in PVC with four glass walls. It is
alternatively filled with bi-distilled and dematerialized water; water-ethanol and water-glycerin
solutions, silicon oil. The bubble generators are usually placed at the bottom, sometimes at the top.
The incident beam from a (2) 50mW YAG laser is (3) coupled to a mono-mode optical fiber with
polarization conservation and finally (4) expanded (x10) to obtain a parallel polarized and collimated
beam, with diameter 12 mm and wavelength \(\lambda = 0.532\ \mu m\). Note that
in this setup, the beam polarization can be rotated continuously with a
(5) \(\lambda/2\) plate, whatever we report
here only results obtained with parallel polarization. For some
experiments, the beam expander magnification was reduced from a
factor x5, and even totally removed.
The rising bubbles (17) are
produced at the bottom of the tank
by different bubble generators:
piezo-jet, assisted air jet, porous
plate and electrolytic bubble
generator. Gaussian beam effects
can be neglected as the diameter of the bubbles is always much smaller than the diameter of the
incident laser beam (ratio \(\approx 1:70\) to \(\approx 1:10\)). To minimize the depth-of-fields effects, the scattering
of all bubbles located in the probe volume is collected by an optical system working in a Fourier
configuration (see also Fig. 5 a). The collection optics is first composed of a (7) high numerical
aperture camera lens. This lens collects the scattered light to form an image of the bubbly flow
within the optical probe volume onto a (8) circular iris diaphragm with adjustable size aperture. The
later component plays the role of a spatial filter and allows controlling the lateral dimension of the
optical probe volume. A pair of (9) achromatic doublets is used to obtain the Fourier Transform of
the bubbly flow image onto a (11) CCD chip. After a (10) interference filter centered on the laser
line $\lambda_0$, the far field CSP are recorded by a (11) 12 bits and $1024 \times 1024$ digital CCD camera. With this setup and a water solution, the typical angular range and resolution of this system are $\theta = 70.3 \pm 82.8^\circ$ and $\Delta \theta = 0.02^\circ$ respectively. This angular range can be adjusted by moving with respect to the collection lens, the Fourier lens and the camera. Whatever they are not part of the CARS setup, Fig. 2 shows additional systems used to compare and synchronize CARS measurements: (14) an interferometric Laser Imaging system (Maeda et al. 2000) using the CARS beam as the illumination source, a (12) flash lamp and a (13) long distance micro-video shadowgraphy imaging-system (with a usual CCD or ultra-fast CMOS camera); a (15) triggering system to detect ellipsoid bubbles with helical trajectories (Krzysiek 2009).

3. Refractive index

Refractive index of bubbles

To test the efficiency and resolution of the CARS technique for the measurement of bubbles refractive index, bubbly flows of water-glycerin droplets in silicon oil were produced. The silicon oil had a low refractive index ($m_0 = 1.4042$ at $\lambda = 532nm$) and a low viscosity (20 cSt) at ambient temperature. Water droplets in silicon are bubbles from the optical point of view, since their refractive index is lower than the one of the surrounding medium (i.e. $m \approx 0.953$), but they are droplets from the fluid mechanics one as they sink in the cell. To change bubbles refractive index, water-glycerin dilutions with different mass-fractions we prepared. The refractive index of the different solutions was controlled with an Abbe refractometer. Fig. 3 (a) shows the mean diameter measured with CARS and the micro-video imaging techniques for droplets containing different fractions of glycerin (0-12% in mass). Fig. 3 (b) shows a comparison between refractive indices measured with CARS (on-line, on bubbles) and the Abbe refractometry techniques (off-line, on sample solutions). Measurements obtained with the CARS technique, size and refractive index, fit really well with those of “reference” techniques.

Refractive index of the surrounding medium

The CARS was not initially thought as a technique that can measure the refractive index of the surrounding medium. But, at least, it was thought to be necessary to study the effect of this parameter on bubbles size and absolute refractive measurements. Indeed, each time the refractive index of the surrounding medium changes, it should be necessary to recalibrate the system. If it was the case, this would be a severe drawback of this new optical sizing method. Indeed, the recalibration is not strictly necessary. To demonstrate this, the piezo-jet is used to produce air bubbles in different water-ethanol solutions. Figures 4 (a) and (b) show statistics obtained for mass
fractions in ethanol increasing from 0 up to 40%, giving a relative refractive index ranging from $m^{-1} = 1.3345$ to $m^{-1} = 1.3555$. Figure 4 (a) compares the mean diameters measured with two inverse methods, while Fig. 4 (b) shows the relative refractive index measurements obtained with the NNLSQ-LMT method. CARS and Abbe refractometry measurements are here also in really good agreement. The size measurements seems almost non-sensitive to changes in the liquid composition. Refractive index measurements with CARS, on rising bubbles, are better than $\pm 0.002$. In fact, the overall standard deviation on refractive index between CARS and Abbe refractometry measurements is as small as $\pm 0.0002$. So that CARS can estimate the water/ethanol concentration at better 4% (i.e. 0.4% when considering the global standard deviation). From these results, we can conclude that for most applications, requiring bubbles size characterization, the recalibration of the CARS system would be not necessary (Krzysiek 2009).

Fig. 4 CARS response without recalibration, when the composition of the surrounding fluid is changing: (a) mean and standard deviation, (b) refractive index.

4. Spatial filter and Gaussian beam and effects

Spatial filter

The nominal probe volume of the CARS system is limited by the laser beam diameter but also by the collection optics aperture (spatial filter). To test the influence of the latter component, a piezo-jet was positioned with micro-displacements at different locations within the nominal probe volume (5x5 positions, results averaged over 100 bubbles, see Fig. 5 b). For streams of monodisperse air bubbles in water, three spatial filter apertures (a) 23mm (fully open); (b) 13mm and (c) 6 mm were considered. Figures 6 and 7 shows the corresponding results. To simplify the understanding of Fig. 7, let’s point out that, as a first approximation, results in the same “column” (i.e. $x \approx x$) are assumed to be mainly sensitive to the laser beam intensity profile whereas, results in the same “row” (i.e. $z \approx z$) are expected to be more sensitive to spatial filter effects. Looking at colons of Fig. 7 it appears that CSP intensity profiles change but without significant modification in shape, whereas for rows, changes are significant on the sideways. Indeed, when the bubbles are on the probe volume side $z > 0$ a part of the light they scatter at $\theta < \theta$ is blocked by the collection optics, and vice versa. All these effects are more pronounced for case (b), and even more for case (c). For cases (a), (b) and (c) CSP are severely disturbed for bubble all positions corresponding to $|\Delta z| \geq 6mm$, $|\Delta z| \geq 4mm$ and $|\Delta z| \geq 2mm$ respectively.
Looking at the previous results one could be afraid about the influence of the spatial filter on CARS measurements. However, for bubbly flows where bubbles have random trajectories within the probe volume, the latter effects are negligible. Figure 6, compares with the CSP of bubbles passing through the center of the probe volume, the mean CSP profiles obtained by averaging the CSP corresponding to all bubbles positions considered in Fig. 7. The conclusion is clear, in contrary to what may be concluded from a quick view of Fig. 7: the smaller the spatial aperture is, the smaller are optical distortion effects. It is the reason why, for BSD and refractive index measurements, a good comprise (between distortion and probe volume size) is to use a spatial filter aperture of about 10 mm in diameter.

**Gaussian beam effects**

To introduce the principle of the CARS technique, as well as for all numerical results presented earlier, the intensity gradients within the probe volume were not taken into account. The reason for that is that we restrict ourselves to bubble sizes much smaller than the probe volume dimensions. Indeed, as a technique allowing instantaneous spatial statistics, there is no need to consider probe volumes smaller than the bubble size. However, for some applications, it can be necessary to study the bubbles one by one (low bubble concentrations), i.e. to use a small probe volume (huge bubble concentrations, correlation with other technique, time resolution, etc.). In fact it was the original operating mode of the CARS technique (see Onofri 1999a). As an numerical example of Gaussian beam effects, Fig. 8 shows the evolution of the scattering diagram of an air bubble in water lighted by a laser beam with beam-waist diameter $2\omega_0=75\mu m$, parallel polarization and $\lambda_0=0.532\,nm$, and for three positions of the bubble along the x-axis, with $y=z=0$ and $D=465\mu m$. Note that the CARS optical axis is along the z-axis.
The calculations were performed with the generalized Lorenz-Mie theory (Gouesbet et al. 1988). Fig. 8 shows that the maximum amplification of the critical scattering is reached for $x \approx -200 \mu m$; the CSP is only composed of low frequency fringes and there is no more signature of contributions from rays $p \geq 2$. Eliminating the case $x \approx -100 \mu m$ (whose behavior is rather similar to the previous case), the second CSP with maximum intensity appears for $x \approx +100 \mu m$. The corresponding CSP is rather flat in the critical scattering region and correspond to rays $p = 2$ (Onofri et al. 2009). Extended numerical and experimental investigations (Krzysiek 2009) show that, Gaussian beam effects induce a severe distortion of CSP only for bubbles whose diameter is like $D/2\omega_0 \leq 3$ and for some particular trajectories. However, like for spatial filter effects, these effects compensate more or less each other for bubbly flows provided that bubbles have random trajectories within the probe volume.

5. Interaction between bubbles spatially close bubbles

Twins bubbles and dense aggregates

In Eq. (1), it is assumed that multiple and coherent scattering effects are negligible. This is a commonly accepted hypothesis provided that we are facing to dilute flows as well as bubbles that are spatially (and statistically) not organized. To bring some physical insights about coherent scattering effects, dedicated experiments were performed with water droplets sinking in silicone oil ($m^{-1} \approx 1.0522$), on twins bubbles with particular orientations (see Fig. 9 (a) and Fig. 10) as well as dense aggregates of bubbles (see Figs. 11 and 12). As an example, Fig. 10 presents two micro-video images and the CSP produced by two twins bubbles that are aligned parallel and perpendicular to the CARS collection optics axis. Interference effects that are clearly visible in the former case, are negligible in the latter one. These interference effects have statistically a rather weak influence on average CSP, the inversion procedure, the estimation of the bubble size distribution and
composition (Krzysiek 2009). Note that for the case where the twins bubbles are oriented like $(\phi_1, \phi_2, 0^\circ)$ the rear bubble was “invisible” to the micro-video imaging system.

Using the same setup but with a higher water flow rate, it is possible to streams of rather dense aggregates of water droplets in silicon oil. To illustrate the results obtained (for more details see Krzysiek 2009), Figs. 11 and 12 shows typical micro-video images and the corresponding CSP. In Fig. 11, the stream of bubbles is somewhat destabilized but the effective bubble aggregate is still dilute and the CARS technique is able to infer the bubble size and refractive index with a good accuracy (Krzysiek 2009). In that case, depending on the inversion method used (LMT-LogNorm.) or (LMT-NNLSQ) the CARS succeed plus or less to reconstruct the CSP profile and then, the BSD. In Fig. 12, the bubble aggregate is much more dense and it is clearly multimodal (see micro-video results). Looking at Fig. 12, and more generally, it appears that CARS fails to detect the smallest modes of the BSD. This is emphasizing with Fig. 13 which compares BSD parameters (mean size and standard deviation) obtained for various dense aggregates.

**Bubbles outside the probe volume**

For all OPC techniques, the influence of particles surrounding the nominal probe volume is always a difficult question as, for instance, they can decrease the input laser beam intensity and coherence as well as depolarized the scattered light. To investigate the influence of surrounding bubbles onto CARS response, a curtain of bubbles that was generated to block alternatively the input beam or output beam (left and right sides in Fig. 9 b) or to block the light scattered by the bubbles within the probe volume: towards or in opposite CARS collection optics direction (front and back sides in Fig. 9b). The plan of the the curtain of rising bubbles was kept parallel to the glass windows at a distance of about $\Lambda \cong 30 - 40mm$ from the probe volume center. Bubbles within the probe volume were produced by the piezo-jet in order to ensure a highly monodisperse and continuous stream of bubbles. Fig. 13 shows typical images recorded with the micro-video systems when the curtain of bubbles was on the left side and, on the right side (most unfavorable cases). Bubbles generated within the probe volume are pointed out with a circle and an arrow. Note that since the micro-video system was focused onto bubble within the probe volume, the images of the surrounding bubbles are blurred (out-of-focus) but can hide some of the bubbles within the probe volume (which should be normally detected). Fig. 13 shows also typical CSP for the four curtain’s positions. The influence of the surrounding bubbles is really weak on CSP and then, on CARS measurements (only few percent deviation on the mean size, see Krzysiek 2009 for more details).
Fig. 9 (a) Coordinate system and setup for (a) the twins bubbles and (b) the curtain of rising bubbles.

Fig. 10 From left to right: twins water bubbles in silicon oil oriented along the CARS collection axis (φx = φz = 0°) and close to the vertical axis (φx = 5°, φz = 0° and $L_{tw} \approx 1.1D$), $L_{tw} \approx 1.1D$.

Fig. 11 Left: Stream of water droplets in silicon oil and CSP profiles (measured and reconstructed); Right: CARS results for dilute to dense aggregates of bubbles.

6. Bubbles non sphericity
CARS is devoted to characterize spherical bubbles (typically: rising air bubbles in water with diameter below 800-900μm). However, in some bubbly flows bubble shapes can deviate notably from the spherical one. According to the bubble-shape diagram of Grace et al. (1976), the ellipsoidal shape appears to be the most common when the bubbles begin to deviate from the sphericity. For this shape the interesting thing is that the response of the CARS technique could be interpreted, at least to a certain extent. Indeed, according to the POA, the CSP is mainly sensitive to
the radius of curvature of the bubble in the scattering plane. For a bubble aspect ratio \( \xi = a / b \leq 1 \), where \( a \equiv a^\alpha \) is the small axis (gravity direction) and \( b \equiv b^\beta \) is the major axis (horizontal/scattering plane) of the bubble; the diameter measured is expected to be \( D \parallel b \) (see Fig. 14 a).

Fig. 12 Stream of dense aggregates of water droplets in silicon oil, CSP profiles (measured and reconstructed) and Bubble size distributions (micro-video and CARS).

Fig. 13 Left: Typical micro-video images for a curtain of bubbles localized (a) on the right side or (b) left side of the cell. Right: comparison of recorded CSP for different positions of the curtain of bubbles.

To investigate experimentally the response of CARS system to non spherical bubbles, streams of bubbles with different aspect-ratios were generated. Figure 14 (b) shows the range covered versus the Tadaki number (measured with the micro-video system and a particle tracking method, see Krzysiek 2009). As a first step and to not mix several complex effects, we have only considered non spherical bubbles that are symmetric with respect to the gravity direction (large axis within the scattering plane). They are referenced here as bubbles with a tilt angle equal to zero \( \delta = 0^\circ \). For this purpose, the CARS system was only trigged on the corresponding bubbles. As illustrative results, Fig. 15 shows two limit cases: a bubble close to sphericity \( \xi \approx 0.90 \) and a clearly ellipsoid one \( \xi \approx 0.54 \). For both cases, the experimental and the reconstructed CSP profiles match almost perfectly. This is an important result, indicating that in the scattering plane the CSP of elliptical bubbles are equivalent to the ones of spheres: CSP of ellipsoid bubbles are “understandable”,

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which is not necessarily the case for the rainbow scattering for instance. Figure 18 summarizes and compares the results obtained for different aspect-ratios. Note that CARS measurements are in good agreement with micro-video measurements for the large ellipsoid axis $b$. Similar good results were obtained for refractive index measurements (see Krzywiek 2009). The problem here is that, for the moment, CARS do not provide any information about the smallest ellipsoid axis. So that it overestimates the void fraction of highly ellipsoid bubbles (see in Fig. 18, the equivalent volume estimation).

![Diagram of ellipsoid bubble model](image1)

**Fig. 14 (a) Coordinate system of ellipsoid bubble model and (b) Measured bubble aspect-ratio.**

![Micro-video images and CSP for elliptical air bubble in water](image2)

**Fig. 15 Micro-video images and CSP for elliptical air bubble in water with larger axis in the scattering plane.**

There is many publications in the literature investigating tilt angle of free falling droplets. One main conclusion of these works is that, statistically, droplets tilt angle is null $\bar{\delta} = 0^\circ$. For rain droplets, the standard deviation of the tilt angle is $\sigma_\delta \approx 3^\circ$. We did not find such studies for bubbles. But, by analogy and from symmetry considerations, we can infer that the mean tilt angle of bubbles should also be null. The underling idea is that, if positive and negative tilt angles produce CSP that
statistically compensate each other (like for spatial filter effects), we could interpret these with those of spherical bubbles. To investigate the effect of ellipsoidal bubbles tilt angle, we basically did the same than for Fig. 10, but by triggering the CARS systems onto tilted bubbles. Figure 16 shows a typical micro-video image recorded as well as the corresponding CSP, and the inverse BSD. Clearly, the measured and the reconstructed CSP do not fit like for ellipsoid bubbles with $\delta = 0^\circ$. The BSD measured with the CRAS technique shows that it is necessary to use a broadband range of spherical bubbles (i.e. circular diameters) to describe the measured CSP. In other words, the spherical bubble model is not really appropriate to describe the scattering properties of tilted ellipsoid bubbles. Indeed, Fig. 17, which summarizes and compares CARS and micro-video size measurements, shows that for largely tilted bubbles the estimation of the bubbles larger axis is also biased.

![Fig. 16 Recorded micro-video and CARS results for a tilted bubble.](image)

![Fig. 17 Comparison of CARS response for ellipsoid bubbles with (a) large axis $b$ in the scattering plane (tilt angle, $\delta = 0^\circ$) or (b) bubbles tilted from the scattering plane ($|\delta| > 0^\circ$).](image)
7. Conclusion

The Critical Angle Refractometry and Sizing technique (CARS) allows accurate and fast characterization of bubble size distribution and refractive index (Onofri et al. 2009). In the present study, to further improve the CARS technique capabilities, we have investigated various effects: bubble relative refractive index, bubbles non sphericity, Gaussian beam intensity profile and spatial filter effects. For the two latter cases, it is shown that for bubbles with random trajectories in the probe volume these effects are naturally compensated. However, for some particular flows, composed of a regular stream of bubbles, caution must be paid to centre the bubbles within the probe volume. For bubbles non sphericity, it is emphasize that, with a spherical bubble model, CSP of ellipsoid bubbles with larger axis within the scattering plane can be inverse with high accuracy, whatever CARS do not yet provide any information on ellipsoid bubbles smaller axis. For tilted ellipsoid bubbles, the inversion procedure seems to be not so straightforward. For reasonably spherical bubbles (aspect ratio greater that 0.9), refractive index measurements are shown to be accurate at better that ±0.002 or ±0.0002 (over all the considered refractive index range). It is the reason why, CARS is thought to be promising for bubbles composition recognitions or coalescences studies. Perspectives for this work concern the scattering properties and inversion of largely ellipsoidal bubbles.

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9. References


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