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LIGHT SCATTERING MEASUREMENT OF THE GAS BUBBLE DYNAMICS

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Abstract. In order to optimize the LIBS signal from bulk waters, laser pulse energies were varied through changing of the QS trigger delays. It was noted that at mid pulse energies the LIBS signal had a tenfold enhancement. In order to explain such a phenomenon the dynamics of the gas bubble generated after the first laser pulse was studied through measurements of the HeNe laser light scattered on the bubble.

1. INTRODUCTION

One of the factors to take into consideration for double pulse LIBS applied on bulk liquids or immersed samples is the timing between the pulses. The maximum LIBS signal can be obtained if the second pulse hits the sample when the gas bubble produced by the first pulse reaches its expansion maximum, (Casavola et al. 2005).

Dynamic of the laser generated bubbles in liquids was mainly studied for laser medical applications (see Vogel et al. 1999 and therein cited papers) with the aim to avoid an excessive tissue damage, and also in the attempt to obtain efficient tissue ablation or high efficiency of the shock-wave generation (laser lithotripsy). The time evolution and maximum radius of the laser-induced bubble in a certain liquid are strongly dependent on the experimental conditions (see Vogel et al. 1999 and therein cited papers), such as laser wavelength, pulse duration and numerical aperture after the focusing lens.

One of the methods for studying the gas bubble dynamic is laser scattering, (Holt, Crum, 1990 and Lentz et al. 1995). The laser-induced bubbles have diameter in order of $0.010 - 1$ mm (Kennedy et al. 1970) and the scattering of light in the visible can be described by Mie's theory (Holt, Crum, 1990 and Kerker 1969).

The final scope of here described research was to improve the sensitivity of LIBS technique applied on bulk liquids, also intended to employ for sub-glacial lake exploration. To this aim, the influence of the laser energy, divided in two or more nanosecond pulses, on underwater plasma emission was studied. Dynamics of the lateral gas bubble expansion after the first laser pulse was measured by light scattering techniques and for different laser pulse energies.

2. EXPERIMENT

The underwater plasma was produced by a Q-Switched (QS) Nd:YAG laser operated at 1064 nm, with maximum pulse energy of about 300 mJ and with repetition rate of 10 Hz. The QS trigger was externally controlled, thus to have a possibility to extract also two laser pulses during single lamp flashing. In such case, the separation between two laser bursts was typically varied between $50 - 100$ μs, which together with the first trigger delay, determines the energy partition between the two pulses. The triggering scheme and examples of pulse energy partitions are reported in our recent papers (Lazic et al. 2005).

2. 1. SCATTERING MEASUREMENTS

The gas bubble produced by the first laser pulse was illuminated by a 35 mW HeNe laser. The scattered signal was measured at 90^o using the same collection optics as for LIBS measurements or at 20^o in forward direction where the light was collected with a plano-convex lens, (Fig. 1). In both cases the full angle aperture for the scattered light collection was larger than 15^o in order to minimize angle dependent signal ripples characteristic for Mie scattering (Holt, Crum, 1990, Lentz et al 1995, and Kerker 1969).

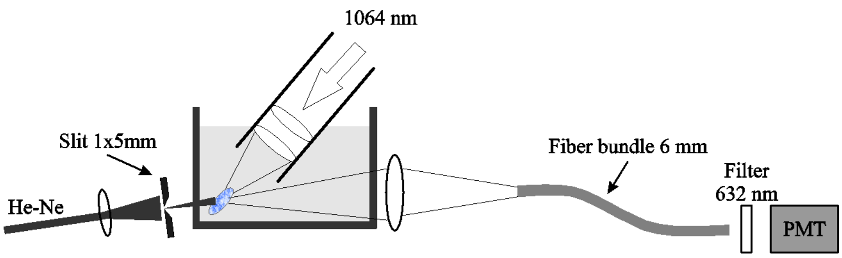

Figure 1: Experimental lay-out for forward scattering measurements.

The signal was brought to a photomultiplier (PMT) diameter fiber bundle. Between the fiber bundle exit and the PMT, an interferential filter centered at the HeNe laser emission was placed. The PMT used for the present experiment is a Hamamatsu R928. The high voltage power supply feeds a divider designed for high linearity; and the output anode is pre amplified and AC coupled with a Tektronix 2430A oscilloscope to record light transient. An additional gate circuit to electronically switch off the PMT gain during the time elapsing between consecutive laser shots was also used. Gated operation allowed us to use a CW laser, minimizing problems related with the high mean photon flux incident on the PMT cathode due to light scattering by hydrosols and particles suspended in water. However, this background scattering was always present and the PMT was operated at relatively low voltage (425 V) in order to avoid nonlinearities in its response.

Considering that the laser produced gas bubble might be elongated, in order to measure only its lateral expansion, a 1 mm high slit, with precisely adjustable vertical position, was placed in proximity to the optical window of the beaker wall. Position of laser generated plasma and consequently the bubble center might depend on the laser pulse energy, (Kennedy et al. 1997). In order to keep the illumination constant across the bubble, a negative lens ($f = -50$ mm) was placed before the slit, which horizontal width was restricted to 5 mm. The power distribution through the slit, scanned vertically in 3 mm range, was checked by a power-meter and resulted uniform within 5%. This range of the slit positions encloses both the bubble produced by low laser energy pulse (8 mJ) and the one produced at the maximum laser energy (300 mJ, single pulse).

The measurements of the bubble dynamics by the laser scattering technique were performed on tap water, whose standard impurities content was previously determined, (De Giacomo et al. 2005). In order to avoid disturbances from eventual nanoparticle formation, water in the beaker was exchanged each 30 minutes of the measurements. During water exchange, the focusing lens was always cleaned.

3. RESULTS AND DISCUSSION

In order to monitor only the lateral bubble expansion, which we consider more important for the LIBS signal after the arrival of the second laser pulse, the HeNe beam was sent through 1 mm high slit, moved with 1 mm step, and scattered signal was measured at different slit positions. At maximum laser energy here used (280 mJ) the signal could be observed for three slit positions (over 3 mm), while up to 170 mJ the signal is existent for only two slit positions. In all the cases, the same slit position gave the signal maximum and corresponding scattered light intensity as a function of time is depicted in Fig.2a.

The first, narrow peak, correspond to the laser pulse arrival, which produce the plasma continuum emission also in the spectral range transmitted by the interferential filter. In the same figure (2b), the scattering signal obtained without slit is also reported. Here, we could observe that the first bubble collapse occurs between 240 μs and 320 μs, followed by the first rebound. At the maximum energy, also the second and weak third rebounds were detected (Fig. 2b).

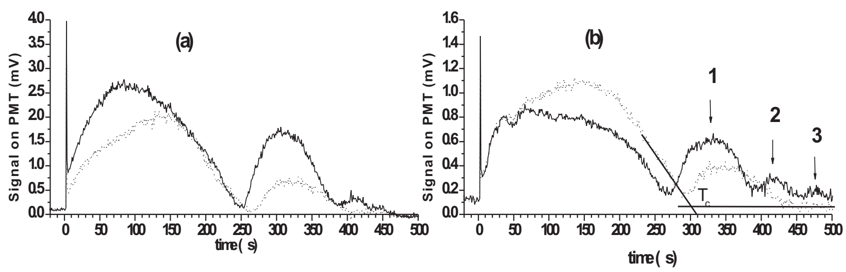

Figure 2: Signal from the light scattered at 90^o on the gas bubble produced by the laser with pulse energy 65 mJ (dots) and 280 mJ (solid): a) without slit; b) with slit; T_c is measured first collapse period at lower energy; 1-3 indicate the bubble rebounds at the higher laser energy.

The signal shape in presence of the slit is quite different from the one in the absence of the aperture. With the slit, the signal is more symmetrical, and the highest peak, as well as the longest first collapse period, is observed at a relatively low energy, thus indicating largest lateral bubble expansion.The intensity of the light scattered by the spherical bubble of radius R is proportional to R^2 for all scattering angles (Kerker 1969 and Barber, Putterman 1992). Consequently, the gas bubble diameter is proportional to the square root of the PMT voltage V (Barber, Putterman 1992). The maximum bubble radius achieved R_{max} is proportional to the first collapse period T_c through Rayleigh relation (Vogel et al. 1999). The first collapse period T_c was determined from the intersection of linearly fitted decaying signal on PMT and the final voltage level (see Fig. 2b) (Hubert et al. 1998). The maximum lateral bubble expansion corresponds to the laser energy of 65 mJ. Up to this energy, V_{max} value increases approximately linearly. After achieving the maximum, the gas bubble starts to elongate or to form in multiple sites as the breakdown threshold is reached also out of the focal volume. Therefore, a smaller part of the available optical energy is coupled to the mechanical energy (bubble expansion) in the focal point. The measured values of V_{max} and T_c are correlated with factor 0.91. The slightly different behaviour between these two parameters could be observed for the laser pulse energies between 65 mJ and 125 mJ, where probably an elongated bubble or multiple bubbles are formed inside the illuminated section, which corresponds to about 1.5 mm length along the laser axis.

From the results reported in this section, we might conclude that for the present focusing conditions, the optimal energy of the first laser pulse, used to prepare LIBS analyses by the second pulse, is about 65 mJ. Further increase of the first pulse energy leads to the LIBS signal deterioration due to the plasma elongation and reduced lateral bubble expansion.

References

Barber, B. P., Putterman, S. J.: 1992, *Phys. Rev. Letters*, **69**, 3839.
Casavola, A. et al.: 2005, *Spectrochim. Acta Part B*, **60**, 975.
De Giacomo, A. et al.: 2005, *Appl. Surf. Sci.*, **247**, 157.
Holt, R. G., Crum, L. A.: 1990, *Appl. Optics*, **29**, 4182.
Hubert, P. et al.: 1998, *Phys. Med. Biol.*, **43**, 3113.
Kennedy, P. K. et al.: 1997, *Prog. Quant. Electr.*, **21**, 155.
Kerker, M.: 1969, Academic Press, New York.
Lazic, V. et al.: 2005,*Spectrochim. Acta B*, **60**, 1002.
Lentz, W. J. et al.: 1995, *Appl. Opt.*, **34**, 2648.
Vogel, A. et al.: 1999, *Appl. Phys. B*, **68**, 271.