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Memory-Effect on Acoustic Cavitation

Oğuz Yavaş, Paul Leiderer,
Department of Physics, University of Konstanz, D-78434 Konstanz, Germany

Hee K. Park, Costas P. Grigoropoulos,
Department of Mechanical Engineering, University of California Berkeley, CA 94720

Chie C. Poon, and Andrew C. Tam
IBM Almaden Research Center, 650 Harry Road, San Jose, CA 95120-6099

ABSTRACT

The formation of bubbles at a liquid-solid interface due to acoustic cavitation depends particularly on the preconditions of the interface. Here, it will be shown that following laser-induced bubble formation at the interface the acoustic cavitation efficiency is strongly enhanced. Optical reflectance measurements reveal that this observed enhancement of acoustic cavitation due to preceding laser-induced bubble formation, which could be termed as a memory effect, decays in a few hundred microseconds. By performing a double-pulse experiment using two excimer lasers the influence of process parameters, such as liquid temperature and salt concentration, on the temporal decay of the memory effect has been studied. An analysis of the experimental results by a diffusion model is presented.

1. INTRODUCTION

Acoustic cavitation, i.e., the formation of bubbles or cavities in a liquid induced by the tensile pressure of an acoustic field, is of importance in many technical applications, such as sonochemistry, ultrasonic cleaning, laser surgery in medicine, and lithotripsy.[1–4] Theoretically, it has been predicted that a tensile pressure of about 500 MPa is needed to generate cavitation in water. However, cavitation can experimentally be achieved at acoustic amplitudes of the order 0.1 MPa. In order to account for this discrepancy it has been assumed that long-lived cavitation nuclei, such as ultramicroscopic bubbles or other inhomogeneities, are usually present in liquids.[5] However, small bubbles are thermodynamically unstable and may either dissolve and vanish quickly or grow and rise to the liquid surface.[6] Therefore, several models have been suggested to account for the generation and stabilization of microbubbles, including cosmic radiation,[5,7,8] clusters of organic or ionic molecules,[9–11] van der Waals stabilization,[12] and gas inclusion in crevices on container wall or solid impurities in the liquid.[13,14] In addition, for highly purified and degassed water a significant decrease of the cavitation threshold due to neutron irradiation has been observed. This decrease is reversible, i.e., the threshold returns to the initial level in about half an hour when the source of radiation is removed,[5] and can be termed as a memory effect.

The physical mechanism underlying this effect has not been understood yet, and, to our knowledge, there has not been any study on a possible memory effect for acoustic cavitation at a liquid-solid interface. Here, we demonstrate the generation of acoustic cavitation by nanosecond pulses and show for the first time the existence of a bubble memory effect at a liquid-solid interface. The temporal decay of this memory effect has been determined and can be explained by a diffusion process.

2. EXPERIMENTAL PROCEDURE AND RESULTS

A detailed description of the experimental approach can be found in Refs. 15 and 16, and shall be discussed in the following only briefly: A solid film ($0.2 - 0.3$ μm thick chromium films on a sapphire substrate) is immersed in a test liquid contained in a quartz cuvette. A short pulsed laser irradiation heats the surface of the solid film above the boiling temperature of the liquid in about 20 ns. As a result, bubbles are formed at the liquid-solid interface, which can be monitored by the transient change of an optical reflectance signal. The laser irradiation also leads to the generation of an acoustic pulse of high intensity via the thermoelastic effect[17] and bubble collapse.[16] Previously, it has been observed that this high intensity acoustic pulse causes cavitation at the liquid-solid interface after being reflected at the quartz window of the cuvette, giving rise to repetitive "echoes" in the optical reflectance signal when the surfaces of the solid sample and the quartz window are parallel.[16,18]

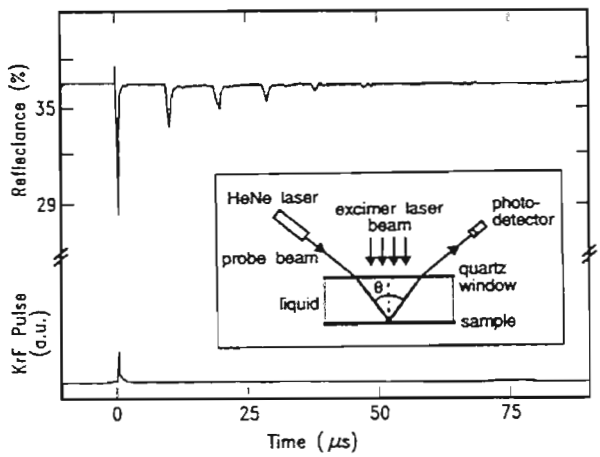

Fig. 1. Optical reflectance signal due to bubble formation at a methanol-chromium interface. $\lambda_{probe} = 632.8$ nm, unpolarized, $\Theta = 15°$. The solid sample and the quartz window are parallel as shown in the inset. The initial reflectance drop is due to thermally induced bubbles and the following echoes are due to acoustic cavitation caused by the travelling acoustic pulse. (The distance between the solid sample and the quartz window is ~ 7 mm.)

An example for a methanol-chromium interface is shown in Fig. 1. While the initial reflectance drop is due to bubble formation induced by the excimer laser heating, the succeeding echoes are solely due to acoustic cavitation induced by the travelling pressure pulse. The decrease of the amplitude of these successive echo signals had been ascribed to the attenuation of the acoustic pulse.[16] However, as shall be seen, this decrease is also due to the slow decay of the cavitation efficiency, i.e., the memory effect.

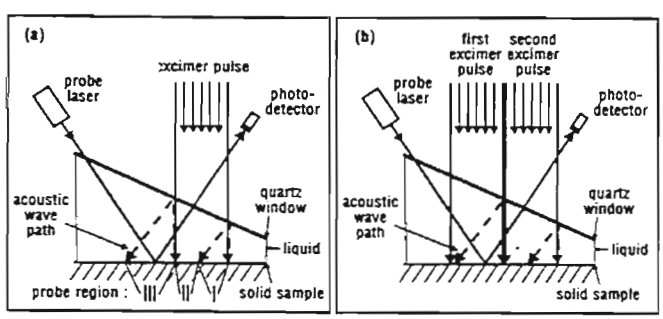

Fig. 2. (a) Experimental setup used to proof the existence of the memory effect. Solid line represents the path of the laser beam, dashed line the path of the acoustic pulse in the liquid. (b) Experimental setup for the measurement of the temporal decay of the memory effect. The first laser pulse initiates the memory effect and the second generates the acoustic pulse.

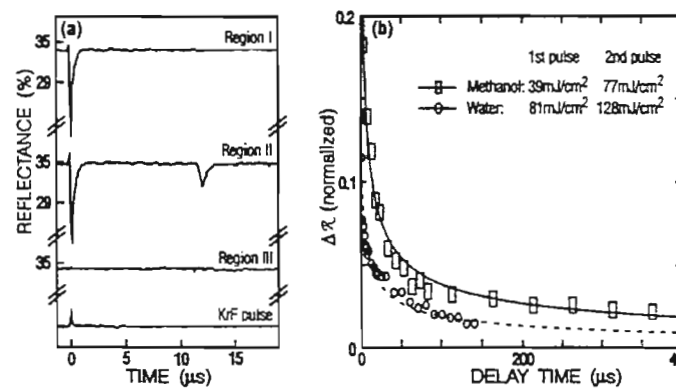

Fig. 3. (a) The optical reflectance signal for the three regions shown Fig 2(a) confirming the existence of a memory effect at the laser-irradiated area. (b) The echo signal peak amplitude as a function of delay time between the laser pulse and the acoustic pulse (temporal decay of the memory effect) for liquid-chromium interface. The lines represent the fit of the experimental data with the diffusion model.

The memory effect is demonstrated using the experimental configuration shown in Fig. 2(a). The quartz window is tilted with respect to the solid sample so that after a round-trip travel in the cuvette the acoustic pulse is displaced and partly overlaps with the laser-irradiated area. Consequently, there are three distinct regions on the solid surface. Region I is irradiated by the excimer laser pulse only. Region II is irradiated by the excimer laser and additionally subjected to the acoustic pulse with a delay time corresponding to its round-trip period in the cuvette. Region III is free from laser irradiation and is subjected to the acoustic pulse only. The HeNe laser beam is positioned to probe each of the three different regions. Representative results for a water-chromium interface are shown in Fig. 3(a). For Region I, only one reflectance signal is observed, which is due to initial bubble formation induced by the excimer laser heating. For Region II, two separate reflectance signals are observed. The first is due to the initial bubble formation and the second due to acoustic cavitation. For Region III, no reflectance signal is observed, i.e., the acoustic pulse cannot cause any cavitation at a previously unirradiated surface. Thus, the laser-induced bubble

formation at the solid surface results in a long-term enhancement of acoustic cavitation. Any thermal effect can be excluded as a possible explanation for this effect since our heat diffusion computations reveal that the temperature at the water-chromium interface decreases to room temperature in about 1 μs following the laser pulse.[16] Therefore, it is hypothesized that the laser-induced bubbles collapse into metastable ultramicroscopic bubbles, i.e., long-lived cavitation nuclei. These nuclei, which are deemed absent on the unirradiated surface, subsequently enhance acoustic cavitation.

In order to determine the temporal decay of the memory effect, we have performed a series of double laser-pulse experiments. The setup of this experiment is shown in Fig. 2(b). The beams from two separate 248-nm KrF excimer laser units are directed to the solid sample without any spatial overlap. The HeNe laser beam probes the location which corresponds to Region I for the first pulse and to Region III for the second pulse. The first laser pulse initiates the memory effect and the second pulse generates the high-intensity acoustic pulse of short duration. While the time delay between the first and the second laser pulses is varied, the fluences are kept constant to assure that at each time both the amount of microbubbles formed and the intensity of the acoustic pulse are about the same. When the two lasers are fired simultaneously, the same signal as in Fig. 3(a) is observed (signal for Region II). With increasing delay time the amplitude of the second reflectance signal decreases, i.e., the memory effect decays. Eventually, the acoustic cavitation returns to normal efficiency. Representative results for water-chromium and methanol-chromium interfaces are shown in Fig. 3(b). The temporal decay in the reflectance signal can be well fitted with the following formula:

$$\Delta R(t) = C/\sqrt{t + t_0},$$

where $\Delta R(t)$ is the peak reflectance drop normalized with respect to the initial reflectance value, C the proportionality constant, t the delay time between the laser pulse and the acoustic pulse $t \geqslant 0$, and t_0 a constant to account for the time required for the laser-induced stabilized nuclei formation. Typical values for C are ranged as $0.1 - 0.2\ \mu s^{1/2}$ for water and $0.2 - 0.4\ \mu s^{1/2}$ for methanol. The values for t_0 are found to be $0.3 - 3.2\ \mu$s for water and $0.9 - 9.7\ \mu$s for methanol. These constants are dependent upon the first and the second laser fluences. The values of t_0 are consistent with the expected higher bubble collapse speed for water because of its higher surface tension compared to methanol. The fitting equation resembles the well-known equation for the surface history for a one-dimensional diffusion process.[19] The change on the reflectance signal due to scattering losses caused by small non-absorbing particles in a host medium, such as micron-sized bubbles in a liquid, can be approximated by:[20,21]

$$\Delta R(t) \approx \sum_r N(t, r)\, r^2(t),$$

where $N(t, r)$ is the bubble number density of radius r. Thus, the reflectance signal is only a function of the distributed bubble radius and number density. Since the intensity of the acoustic pulse is kept constant, the changes on these properties of grown-up bubbles (post-cavitation bubbles) are directly related only to the changes on the properties of laser-induced metastable ultramicroscopic bubbles (cavitation nuclei). Therefore, the observed temporal decay of the peak reflectance drop, i.e., the diffusive decay of the quantity $\sum_r N(t, r)\, r^2(t)$, is attributed to a net diffusive change on the properties of laser-induced ultramicroscopic bubbles.

The observed diffusive decay of the memory effect could be ascribed to three different mechanisms. First, the bubble radius can decrease by diffusion of entrapped gases into the liquid. Second, stable microbubbles may diffuse away from the sample surface to yield a smaller bubble number density. However, this effect is expected to be small since the diffusion velocity for bubbles is small[6,11] and the time scale considered is less than hundreds of microseconds. The third mechanism is the coalescence of microbubbles to form bigger bubbles. From the present data it is not possible to rank the first and the third mechanisms because the bubble size and the number density have not been independently determined.

The interpretation of the temporal decay in the reflectance signal by a diffusion process of microbubble properties could be verified by varying the laser fluences in the double-pulse experiments. When the fluence of the first laser is increased more microbubbles will be formed on the solid surface. Similarly, when the fluence of the second laser is increased an acoustic pulse of higher intensity will be generated. Both mechanisms will lead to an increase of cavitation yield. This has indeed been observed as shown in Fig. 4(a) for water-chromium interface, where the decay of the memory effect is compared for two different fluence values of the first laser pulse. The amplitude of the reflectance drop increases as the laser fluence is increased. The same trend is observed when the fluence for the second laser is varied. An interesting result is obtained when the ion concentration of water is increased by introducing NaCl. Figure

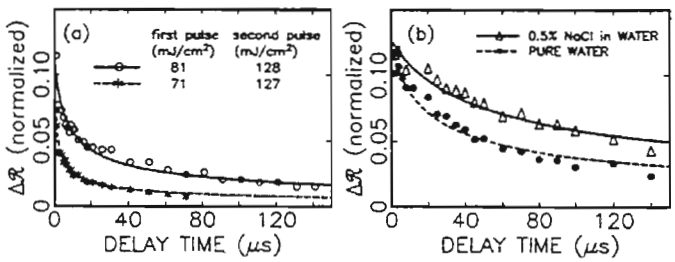

Fig. 4. (a) Effect of fluence variation for the first laser pulse on the memory effect. (b) Stabilizing effect of ion concentration on the temporal decay of the memory effect for water-chromium interface. $F_1 = 82$ mJ/cm^2 (first pulse) and $F_2 = 120$ mJ/cm^2 (second pulse). The lines represent the fit of the experimental data with the diffusion model.

4(b) shows the results for pure (spectroscopic grade) water and for water with NaCl (0.5%). As can be seen, the introduction of ions has a stabilizing effect on the microbubbles and consequently the memory effect is prolonged. This result could be explained by the hypothesis of the ionic cluster formation[10,11] as a possible stabilization mechanism for ultramicroscopic bubbles.

3. CONCLUSION

We have demonstrated the existence of a long-term memory effect, i.e., the enhancement of acoustic cavitation at a liquid-solid interface following laser-induced bubble formation. The temporal decay of the memory effect for two different liquids on a chromium surface has been determined and could be explained by a diffusion process.

4. ACKNOWLEDGMENTS

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