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GHz ultrasound wave packets in water generated by an Er laser

U Störkel†, K L Vodopyanov‡ and W Grill§

† Hahn-Meitner-Institut, Glienicker Strasse 100, D-14109 Berlin, Germany
‡ Blackett Laboratory, Physics Department, Imperial College, London SW7 2BZ, UK
§ Institut für Experimentelle Physik II, Universität Leipzig, Linnéstrasse 5, D-04103 Leipzig, Germany

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Abstract. A variety of schemes are presented, suitable for the temporally and spatially controlled generation of ultrasound pulses with a centre frequency near 1 GHz. Direct excitation of acoustic waves in water, serving as the coupling and transport medium, relies on the resonant absorption of short λ = 2.8 μm erbium laser pulses in water: at the interface with an adjoining solid material or at a free water surface. Ultrasonic amplitudes, leading to nonlinear acoustic effects, have been demonstrated. The photoacoustic excitation methods presented here are suitable for scanning acoustic microscopy, for example for the real-time study of biological cells *in vivo*.

1. Introduction

Scanning acoustic microscopy (SAM) as introduced by Lemons and Quate [1] is based on focusing transducers. Excitation and detection of acoustic wave packets is achieved by the reverse and direct piezoelectric effect converting the signals in a coherent way.

With water as the coupling medium, suboptical spatial resolution can be achieved in SAM, using a sound frequency of >3 GHz. For instance, spatial resolution better than 2000 Å has already been reported at 4.4 GHz [2]. However, the upper frequency limit is set by the restricted bandwidth of acoustic transducers and high hypersound absorption in water which is 2200 dB cm^{-1} at 1 GHz and increases with the square of the frequency. In addition, there is a large acoustic impedance mismatch between the acoustic lens material (for example sapphire) and the transport fluid (water), so that a large part of the acoustic power is reflected from the lens.

Thus, to compensate for elevated losses in the coupling media at high frequencies and reduction of detection efficiency, one needs to increase ultrasonic power. Yet for applications involving high acoustic power levels at GHz frequencies, the piezoelectric excitation scheme sets narrow limits on the achievable power levels. This is mainly caused by the damage threshold of the piezoelectric transducers (which are rather thin, $\sim$1 μm, films, deposited on a planar surface) with respect to the applied electric fields.

In photoacoustic microscopy, introduced by von Gutfeld [3], generation is based on the absorption of pulsed laser radiation, leading to a thermally induced expansion and the subsequent excitation of acoustic waves (thermoelastic mechanism). Since thermodynamic power conversion is involved, conversion efficiency depends on the actual temperature variation in the photoacoustic excitation process. For practical applications at room temperature with water as a conversion medium, the observed amplitude of the excited ultrasonic wave packet is to a first-order approximation proportional to the absorbed laser power and thermal expansion coefficient of the coupling medium.

Subsequently it has been shown by Vodopyanov *et al* [4] that by using Er laser ($\lambda \approx 3$ μm) excitation with its very high ($> 10^4$ cm^{-1}) resonant absorption in water (and other OH-containing liquids, for example ethanol, glycerin etc) one can achieve efficient generation of GHz sound pulses directly in the liquid, thus avoiding the problems arising from the acoustic impedance mismatch. Sound pressure amplitudes as high as 20 kbar have been achieved in water with about 6% light to acoustic power conversion efficiency.

The aim of the present paper is to demonstrate the feasibility of photoacoustic excitation of ultrasound pulses with high power levels at frequencies of typically 1 GHz and above, representing the frequency limit of commercially available SAMs (2 GHz). Higher acoustic power levels are not only needed to overcome the increasing absorption with rising frequency, but are also useful, if nonlinear acoustic schemes are to be exploited. Such techniques have already been used to enhance the spatial resolution of SAM [2]. The experimental work is based on the detection of the excited acoustic wave packets with commercially available focusing acoustic transducers (so-called 'acoustic lenses' from Leica). This allows a simple comparison with well established schemes. Signal acquisition for the converted acoustic signals

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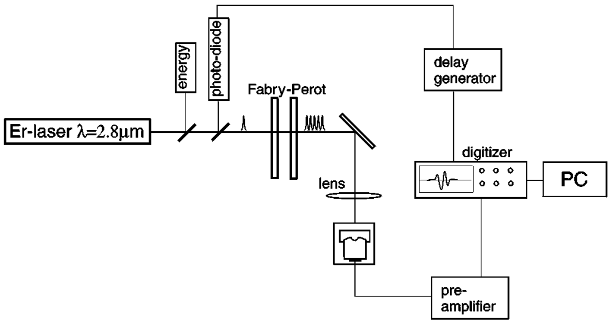
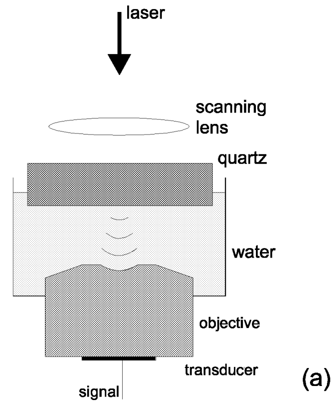


Figure 1. Experimental set-up for photoacoustic generation and piezoelectric detection of acoustic waves.



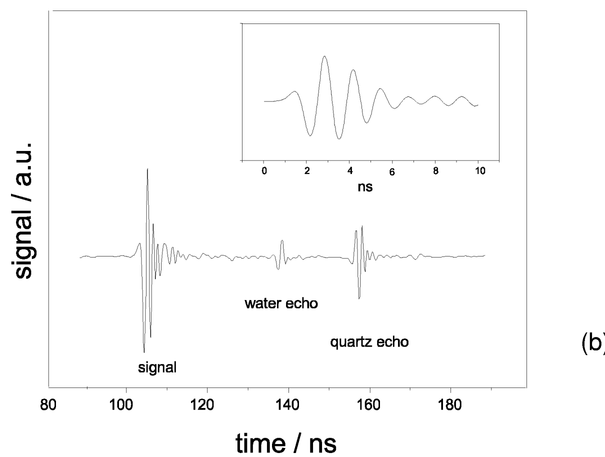


Figure 2. (a) Experimental set-up for the excitation of planar acoustic waves and subsequent spatially resolved detection with a focusing piezoelectric transducer.
(b) Typical acoustic signal for a single-pulse excitation (t = 0 corresponds to the arrival of the laser pulse). Inset: signal with an enhanced resolution and an arbitrary time offset.

can be achieved by transient digitizers, avoiding more elaborate optical detection schemes which have already been introduced for frequencies up to typically 200 GHz [5]. The restriction to the frequency range near 1 GHz also relaxes the interface surface quality requirements. Water has been selected as a coupling and transport fluid not only since it is widely used for this purpose but also because it is a dominant constituent and an appropriate contact medium for biological objects such as living cells, which is of interest for future applications of the developed schemes.

2. Experimental techniques

The experimental set-up sketched in figure 1. An actively mode-locked, Q-switched and cavity dumped Er:Cr:YSGG laser [6] with flashlamp pumping and a repetition rate of 3 Hz was used in our experiment as a primary source for the electromagnetic radiation. The laser emitted single 0.5 mJ pulses with 100 ps duration, TEM_{00} mode and beam diameter 1.5 mm. A Fabry–Perot cavity assembled from two flat mirrors with mechanical adjustment of the spacing and alignment is used to create pulse trains from the single pulses of the laser radiation. The repetition rate within the pulse trains synthesized by this device can be simply controlled by changing the spacing between the mirrors. The filter can be by-passed if single pulses are demanded. The laser radiation is directed to the area used for excitation of acoustic waves.

After transport in the water the excited acoustic wave packets are converted into electrical signals by a focusing transducer. The output signal is preamplified, if needed, and fed to the input of the transient digitizer used for recording. The digitizer is triggered by a split-off Er laser pulse, using a J12 series ultrafast (< 1 ns) InAs photodiode from EG&G. The appropriate time window with respect to the trigger signal is selected with a digital delay generator with 5 ps resolution and by adjustment of the time base of the transient digitizer. Depending on the actual digitizer employed, averaging is performed by the device itself or by a connected PC, which is also used for storage and processing of the data. A pyroelectric detector has been used to monitor the laser single pulse energy, which is controlled by attenuators.

The following schemes have been developed and tested.

2.1. Excitation of planar acoustic waves at an interface between quartz and water

For excitation of approximately planar acoustic waves with a weakly focused (beam size 1.5 mm) laser radiation, a fused quartz, transparent for the laser radiation, is brought into direct contact with water, which is in our case the coupling and transport medium (figure 2). The size and position of the laser beam were adjusted by a scanning lens. The excited plane waves are detected with a focusing transducer, selecting a diffraction limited subset of (approximately) spherical waves of the planar acoustic waves, according to Huygens principle [7]. This technique provides spatial resolution and does not need an angular adjustment of the detector, as compared with the case of planar transducers. The focus of the laser radiation has been adjusted to the quartz/water interface by observation of the echo pattern and appropriate mechanical alignment.

The typical time resolved response of the system following a single short-pulse laser excitation is shown in figure 2(b). The direct signal resulting from the acoustic excitation is observed at a time around 100–110 ns.

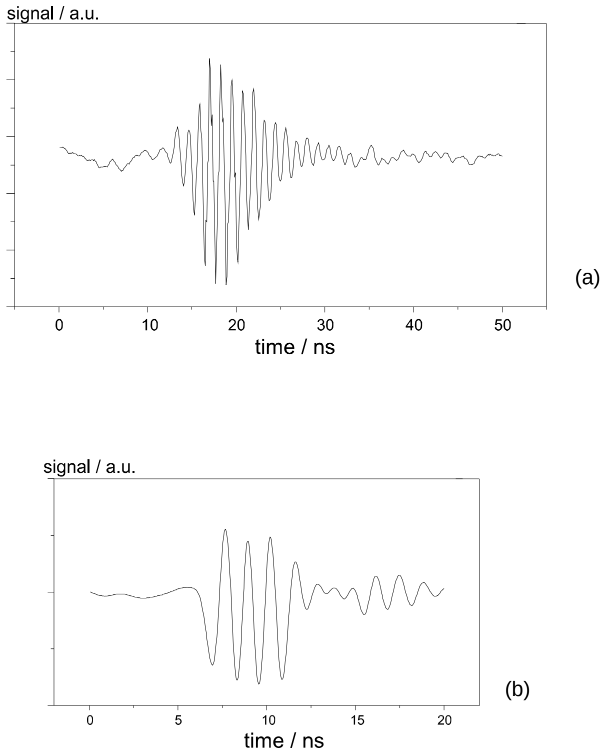


Figure 3. (a) Acoustic signal as in figure 2 but for excitation with a pulse train synthesized by an ideally aligned Fabry–Perot interferometer with a free spectral range of 1 GHz. (b) Acoustic signal with an almost abruptly switched sinusoidal response, achieved by axial detuning of the Fabry–Perot cavity.

The remaining part of the signal includes various echoes resulting from reflections at the interfaces between air, quartz, water and the focusing transducer. The oscillatory structure of the signal (Inset, figure 2(b)) is mainly because of bandwidth limitations present in the focusing transducer due to its acoustic and electromagnetic resonant circuitries.

With the aid of the adjustable Fabry–Perot interferometer different time structures representing pulse trains have been applied for excitation. In figure 3(a) the response of the system is demonstrated with the ideally aligned Fabry–Perot cavity with a free spectral range of $c/2L = 1$ GHz, where L is the spacing between the mirrors and c is the speed of light. This leads to a synthesized pulse train with a repetition rate of 1 GHz, coinciding with the nominal centre frequency of the focusing transducer used for detection. As expected, the build up of the oscillatory signal is mainly determined by the bandwidth of the detecting transducer and the trailing edge by the decay of the optical pulse train caused by the finite reflection (∼90%) of the Fabry–Perot mirrors. Figure 3(b) exhibits a different shape of acoustic pulse train (with a more abrupt envelope shape as compared with figure 3(a)), achieved by small axial detuning of the Fabry–Perot cavity. Such a response is especially suited for phase sensitive vector detection schemes as introduced by Grill *et al* [8]. An electronic reference signal (with respect to the laser pulse) can be obtained here from a fast InAs photodetector and an appropriate delay generator.

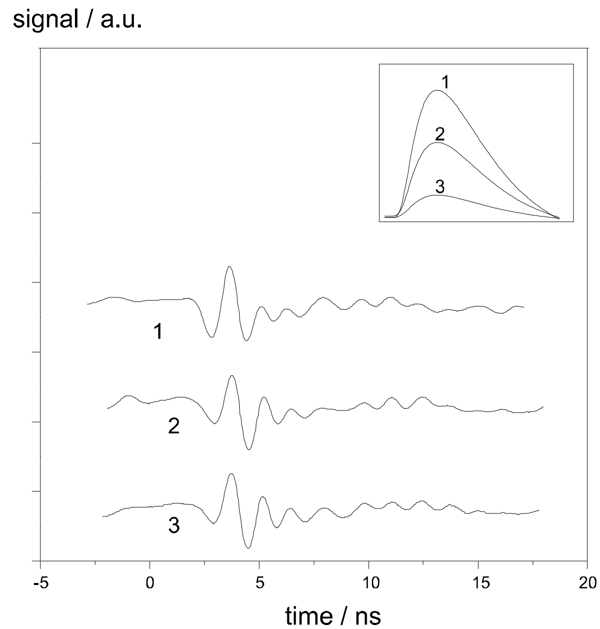


Figure 4. Acoustic signals as in figure 3 but for excitation with a 2 GHz laser pulse train with subsequently (from top to bottom) reduced Fabry–Perot quality factor. Inset: photodetector signal, proportional to the laser train energy, transmitted through the Fabry–Perot cavity.

In the next experiment, the centre frequency of the focusing transducer used for detection was still at 1 GHz, but the laser pulse repetition rate was tuned to 2 GHz. The temporal structure of the time response is shown in figure 4. Under these conditions the excited acoustic wave packets have a centre frequency beyond the bandwidth of the focusing transducer. The observable (at 1 GHz) relatively small signal at the onset of the pulse train results from the Fourier subharmonic and is temporally positioned at the sudden rise of the pulse train. The three signals in figure 4 represent the acoustic wave packets corresponding to the gradually increasing (from top to bottom) detuning of the Fabry–Perot cavity, leading to an overall reduction of the transmitted laser train energy, as demonstrated in the inset to figure 4. All three signals exhibit a comparable temporal structure and amplitude, since contributions to the detectable frequency range for excited acoustic waves result from the onset of the optical pulse train and remain unchanged when the Fabry–Perot cavity is detuned. This example also demonstrates the possibility of bandwidth limited detection at a frequency lower than the central frequency of the pulse train, which may be of great interest for nonlinear acoustic detection schemes. For example, optical excitation may be at 9 GHz and 10 GHz and detection at a difference frequency of 1 GHz.

2.2. Excitation of extended planar acoustic waves at an interface between an optical waveguide and water

Some applications, for example holography with planar waves, require an extended source for planar acoustic waves. In practical applications it is also helpful to avoid the necessity of laterally extended optical access to the

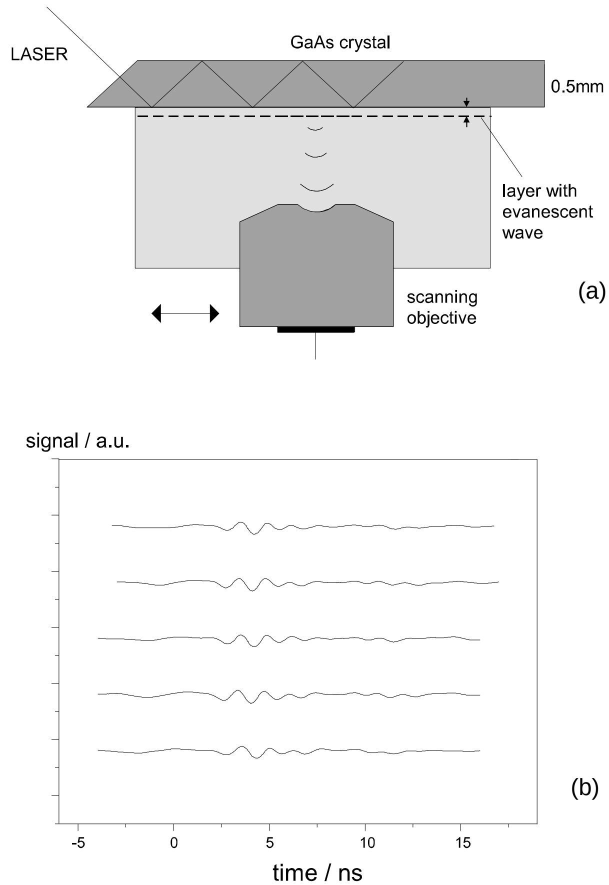


Figure 5. (a) Set-up for the excitation of extended planar acoustic wave packets based on a GaAs optical waveguide with a lateral extension of 5–10 mm in two orthogonal directions. (b) Acoustic signals obtained for different positions in the plane of incidence of the laser beam with a spacing between adjoining selected positions of ∼1 mm.

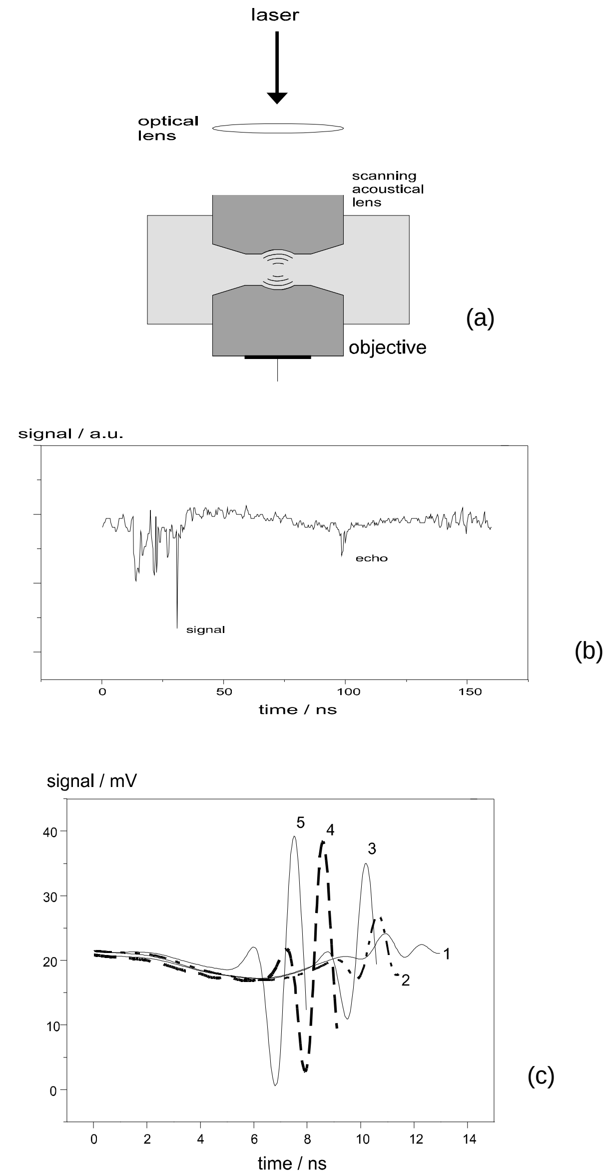


Figure 6. (a) Set-up for the excitation of acoustic waves in a confocal arrangement and (b) acoustic signal corresponding to a transit time of about 32 ns. (c) Signals observed for different amplitudes of the acoustic wave (arbitrary time offset) for increasing laser pulse energy (with an increment of ∼2 for curves 1–5). A substantial reduction of the arrival time, up to 5 ns, can be seen, together with a change of the shape of the signal.

excitation area. Furthermore, homogeneous illumination with laser radiation normally leads to a substantial loss in the available maximum photon flux. We have therefore exploited an excitation scheme based on a crystalline GaAs optical waveguide (0.5 mm thick) with direct contact (via evanescent wave absorption) to water on one of the extended planar surfaces as depicted in figure 5(a). The focus of the transducer is adjusted to the planar interface between the optical waveguide and the coupling medium. The transducer has been manually scanned in a plane parallel to the surface of the waveguide.

The homogeneity of the excitation of acoustic wave packets (with a lateral extension of 5–10 mm) under single laser pulse illumination is demonstrated with a set of signals obtained for different positions of the transducer (figure 5(b)).

2.3. Excitation of focused acoustic wave packets

To demonstrate confocal transmission, a focused beam of acoustic waves has been generated at the spherical interface of an acoustic lens and water. The lens has been taken from a commercially available focusing transducer similar to the one used for the detection of the acoustic wave packets. It consists of single-crystalline corundum (sapphire) with a spherical surface of a radius of about 20–30 μm, on which an antireflective coating for acoustic waves (probably manufactured from glass), optimized for a frequency of about 1 GHz, has been deposited. Confocal alignment is employed in the set-up depicted in figure 6(a). The alignment has been optimized by observation of the transit signal and the echo resulting from reflection at the spherical interfaces. In the time domain the detected acoustic wave packets (figure 6(b)) are similar to the signals resulting from planar acoustic waves. But due to the confocal arrangement, much higher signal levels are observed. The signals could therefore be detected without a preamplifier which had to be employed in all examples demonstrated so far in this survey.

To show the accessibility of nonlinear transport properties for the travelling acoustic wave packets, we have employed higher power levels for the laser radiation used for excitation. To avoid possible destruction of the rather valuable detecting transducer, we have limited the incident laser radiation fluence to a level of $< 10^{-2}$ J cm^{-2} leading to a signal of 2 V at the output of the detecting transducer, just safely below the recommended maximum voltage for excitation of acoustic waves (although we used the focusing transducer for detection only). The signals observed for increasing ultrasonic amplitudes (curves 1–5 in figure 6(c)) correspond to increasing excitation laser pulse energy (with an increment of 2, starting from curve 1). A substantial reduction of the arrival time, up to 5 ns (at the total transit time of 32 ns), can be seen, together with a change of the shape of the signal. The observed nonlinear effects demonstrate that the nonlinear acoustic (shock wave) regime is accessible with a confocal set-up and is suitable for scanning acoustic microscopy. Using the measured decrease of the total travel time by approximately 5 ns/32 ns = 16%, and using formulae (3)–(5) of [4], we estimated (taking into account the wave amplitude non-uniformity along the acoustic path in water) the shock wave amplitude in the focus to be > 10 kbar.

2.4. Excitation of acoustic wave packets at a free water surface

Finally, we demonstrate the feasibility of the excitation of GHz acoustic waves at a free water surface (in contact with air at normal conditions). For this purpose a water droplet has been deposited on the focusing acoustic transducer and adjusted in volume such that the focus of the transducer coincided with the free surface (figure 7(a)). The observed acoustic signal is shown in figure 7(b). The efficiency of the excitation of acoustic waves was similar to the case when the waves were excited at an interface between a solid medium and water. This method employs a self-organized surface of significantly minimized roughness, created by the surface tension, and restrictions for the minimum achievable wavelength are therefore relaxed. Interfaces between gas and fluids can be shaped with the aid of surface tension, gas pressure and suitable circular pinholes or other openings to fractions of a sphere with rather small radius. This offers a simple way to generate a focused acoustic beam at extremely short (a few μm) working distances. Such a self-organized device can easily be reconstructed after damage caused by excessive laser power, dirt or other conflicting conditions.

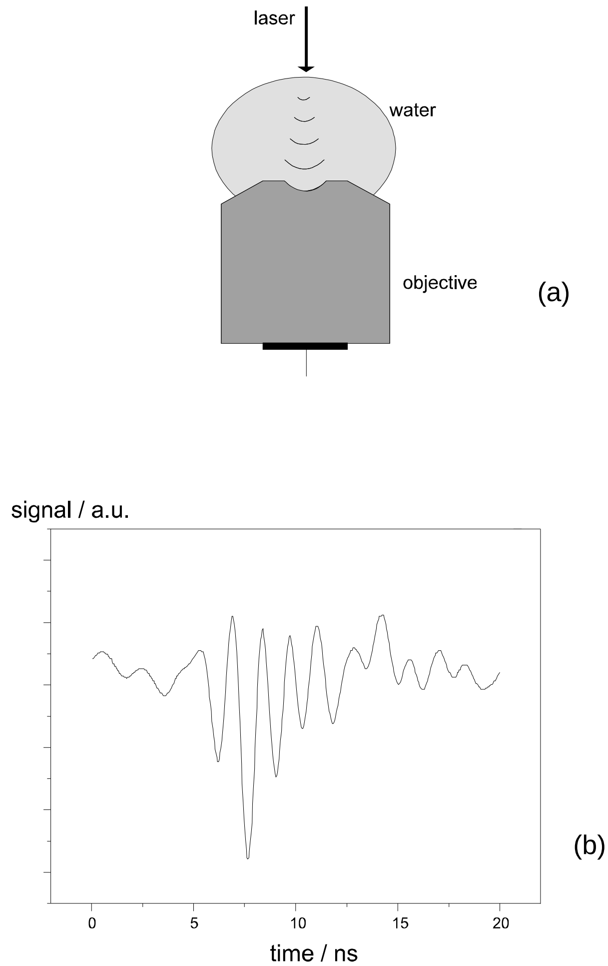


Figure 7. (a) Set-up for the excitation of acoustic wave packets at a free water surface and (b) corresponding acoustic signal.

3. Conclusion

We used picosecond mid-infrared laser pulses to generate, using various schemes, GHz acoustic wave packets, with controllable spatial and temporal characteristics. Generation of acoustic wave packets was based on the direct excitation of water, serving as the coupling and transport medium, which is especially suitable for biologically oriented applications. Amplitudes of the excited acoustic waves leading to a speed-up of the transport in water with velocities significantly above the sound velocity have been obtained in a controlled way. Free surfaces of water, self-organized by surface tension, have been demonstrated to be suitable for photoacoustic generation.

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