Laser Generation and Detection of Strongly Nonlinear Elastic Surface Pulses

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Very high amplitude nonlinear surface acoustic wave pulses with 20–100 ns duration and acoustic Mach numbers up to 0.003 were excited in fused silica by nanosecond laser pulses acting on a strongly absorbing overlayer. Absolute measurements were performed with a calibrated dual-probe-laser deflection setup. The formation of shock fronts during propagation and nonlinear broadening of the wave form were observed for the first time. Three nonlinear acoustic constants were evaluated and compared with values resulting from previously measured third-order elastic moduli. [S0031-9007(97)03749-6]

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The propagation velocity of Rayleigh-type surface acoustic waves (SAWs) with small amplitude does not depend on the frequency or the amplitude for a homogeneous solid [1]. Therefore SAWs with a plane front retain their shape and amplitude during propagation, provided the attenuation is negligibly small. For SAWs with large amplitudes the propagation velocity starts to depend on the particle velocity of the substance and, as a result, different parts of the SAW profile move with distinct velocities. This causes characteristic changes of the pulse shape. Nonlinear effects have been extensively studied for bulk waves, including transformation of the wave profile and formation of a shock front [2–5]. Nonlinear phenomena for SAWs have been studied to a much smaller extent, although they are of importance for strong seismic waves generated by earthquakes as well as in signal processing devices [6–9]. In previous experiments SAWs were excited using interdigital transducers, which allowed only the region of relatively weak nonlinear effects to be reached. The pulsed laser technique makes it possible to obtain very high amplitudes of elastic waves, as was demonstrated with laser-driven bulk shock waves [10]. For the excitation of short SAW pulses the laser pulses should be in sharp focus on the surface. As the laser fluence is increased the screening effect due to the onset of optical breakdown and plasma formation usually prevents an effective input of the laser energy into the material. In previous studies of linear elastic constants of thin films [11,12] and also anisotropy [13] with laser-generated broadband SAW pulses only relatively small amplitudes were excited.

In this Letter we report on the laser generation, propagation, and detection of SAWs with high amplitudes (up to the limit of cracking) in fused silica. The SAW pulses were generated with nanosecond laser pulses through a thin strongly absorbing layer as shown in Fig. 1 [14]. The nonlinear SAW pulses were registered with a cw laser dual-probe-beam deflection (PBD) setup which allowed the measurement at two different distances. The propagation of nonlinear SAWs was numerically simulated using a general theoretical model accurate to the elastic moduli of the third order for an isotropic solid without any *a priori* assumptions on the depth profile of the SAW.

The elastic moduli of the second order of fused silica, which is widely used as an optic and acoustic material in science and technology, are well known [15]. The measurements of the third-order nonlinear constants, mainly responsible for the nonlinear acoustic effects, were performed with different techniques [16,17], delivering data with significant deviations.

Nonlinear SAW pulses were excited in plates of fused silica (Herasil, 3 cm × 2 cm × 3 mm) with Nd:YAG laser pulses (wavelength 1.06 μm, duration 7 ns, energy up to 130 mJ). The laser pulse was focused into a strip with a length of 8 mm and a width of 10 μm (see Fig. 1). In the excitation region the surface of fused silica was covered with a strongly absorbing carbon layer in the form of an aqueous suspension or a solid film of about 100 μm thickness. The transient SAW pulses were registered with a PBD setup using a stabilized cw Nd:YAG laser

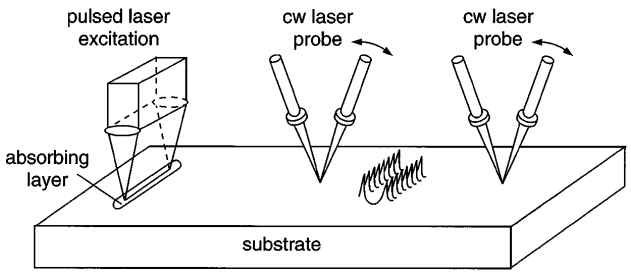

FIG. 1. Schematic of the experimental setup with line-source excitation and two-point probe of the propagating plane wave.

(wavelength 532 nm, power 120 mW). The distance between the two probe spots was about 15 mm. Each probe laser beam was focused by a gradient-index lens into a spot of approximately 4 μm diameter on the sample surface. The bandwidth of the whole setup was limited to about 500 MHz. Because of the low reflectivity of fused silica the power of the probe beams on each photodiode was only 0.2 mW and was far below the saturation limit. The signal, which is proportional to the surface slope of the SAW pulse and hence to the normal component of the surface velocity, was amplified by 48 dB and recorded by a Tektronix TDS 540 A oscilloscope.

To perform absolute measurements the probe setup was calibrated. The differential output of each pair of photodetectors was measured for a known angle shift. The calibration coefficients were found to be close to the value of 16 V/rad that was calculated using the approach given in [11]. The minimal detectable slope in a single-pulse measurement was about 0.5 mrad for fused silica and the signal was linear for a surface slope less than 10 mrad.

The propagation velocity of SAWs with a small amplitude was measured as $v_R = 3401$ m/s. The dimensionless diffraction parameter $D = (8x v_R/\pi f b^2)$, where b is the length of the excitation line, was small (about 0.2) even for the relatively low frequency of $f = 10$ MHz at the propagation distance of $x = 20$ mm. Thus diffraction effects could be neglected over the whole frequency range.

The shape of the SAW pulse measured at a small distance of $x_1 = 2.3$ mm [Fig. 2(a)] shows that the predominant mechanism of excitation corresponds to the transient normal force acting on the surface during the explosive evaporation of the strongly heated absorption layer [18]. In the vicinity of the excitation strip cracks were indeed observed. Outside this area the SAW pulse amplitude was just below the cracking threshold and depended only slightly on the laser pulse energy. In the strongly nonlinear regime the measured pulse profiles of the surface normal velocity acquired a quasisymmetrical shape with two sharp peaks separated by a long valley due to nonlinear transformation during propagation. This is demonstrated by the oscillogram of a SAW pulse measured at a large distance of $x_2 = 18.3$ mm [Fig. 2(b)].

For the in-plane component of the surface velocity, calculated with the Hilbert transform, the appearance of the peaks in the normal velocity distribution corresponds to the formation of two shock fronts, as shown in Figs. 3(a) and 3(b). The pulse in Fig. 3(a) corresponds to an acoustic Mach number of about 0.003, defined as the ratio of the amplitude of the in-plane oscillation velocity to the propagation velocity. At large propagation distances the SAW pulse acquires an inverse N shape with two sharp fronts. The leading front of negative polarity propagates with a higher velocity than the shock front of positive polarity in the trailing part. The difference in the propagation velocities of the shock fronts, which was 0.6% in the present case, caused a significant broadening

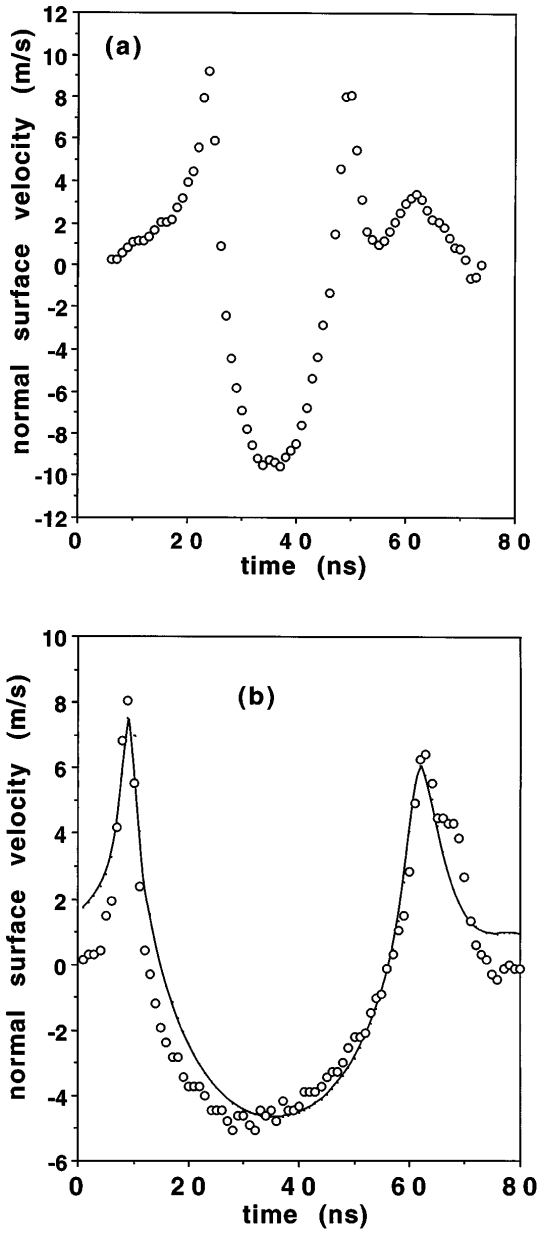

FIG. 2. SAW pulses registered at different distances from the excitation line: (a) $x_1 = 2.3$ mm, (b) $x_2 = 18.3$ mm (points) and calculated wave form (solid line).

of the SAW pulse and a frequency—down—conversion process.

An improved theoretical description of nonlinear SAWs was developed based on the assumption that the cumulative effect of nonlinear interactions causes a slow variation of the pulse shape during propagation in both time and space [19]. In the second-order approximation the relationship between the normal w and in-plane v components of the velocity $w = -1/\gamma H(v)$ remains valid, where $H(v)$ is the Hilbert transform. The evolution equation for the in-plane velocity component of the surface motion for the two-dimensional SAW wave propagating

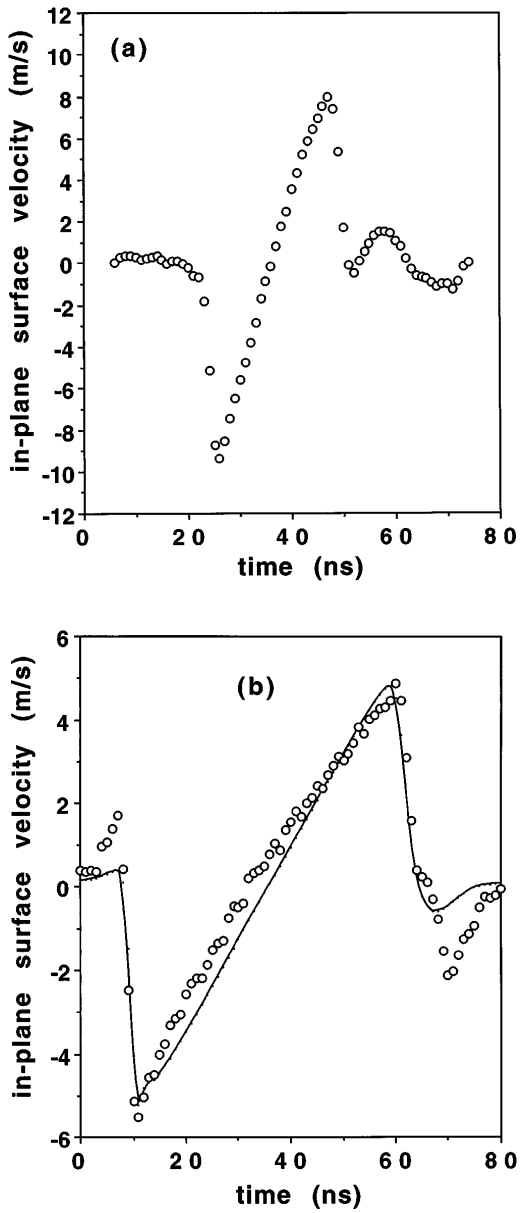

FIG. 3. Wave forms of the in-plane surface velocity corresponding to the pulses of (a) Fig. 2(a) and (b) Fig. 2(b) (points) and calculated wave form (solid line).

along the x-axis and decaying with depth into the solid half-space has the form [19]

$$v_R^2 \frac{\partial v}{\partial x} = \varepsilon_1 v \frac{\partial v}{\partial \tau} + \frac{\varepsilon_2}{2} \frac{\partial}{\partial \tau} \{v^2 + (H[v])^2\} + \varepsilon_3 \left\{ v \frac{\partial v}{\partial \tau} + H\left[v H\left[\frac{\partial v}{\partial \tau} \right] \right] \right\} + \beta \frac{\partial^2 v}{\partial \tau^2}, \quad (1)$$

where $\tau = t - x/v_R$ is the retarded time and v_R is the Rayleigh velocity of small amplitude SAWs. The nonlinear constants $\varepsilon_{1,2,3}$ represent complex combinations of the shear and bulk modulus as well as the third-order nonlinear moduli of the solid. These constants can be directly determined from a comparison of the experimental results with the theoretical description (1), and therefore are useful for the characterization of solids according to the evolution properties of nonlinear SAWs. Equation (1) differs from the corresponding equation derived in [19] by the last term on the right hand side which represents a "classical absorption" [20], proportional to the attenuation parameter β.

Equation (1) was solved numerically by using its spectral representation. The terms connected with the nonlinear constants $\varepsilon_{1,2,3}$ play different roles in the nonlinear evolution of the SAW profile. The constant ε_1 is responsible for the initial steepening of the pulse front, formation of the shock front, and broadening of the pulse. The corresponding term in Eq. (1) is known from the nonlinear acoustics of bulk waves [3]. The constant ε_2 also strongly affects the nonlinear extension of the pulse duration and both ε_2 and ε_3 influence the amplitude and the shape of the pulse peaks.

In order to compare the model with the experimental results the following fitting procedure was used. The spectrum of the in-plane velocity component for the first measured SAW pulse [Fig. 3(a)] was determined using the Hilbert transform and the fast Fourier transform. Then the spectrum of the SAW pulse at a given propagation distance was calculated according to the spectral form of the evolution equation (1). The physical attenuation for fused silica is rather small and an artificial attenuation with $\beta = 2.34 \times 10^{-8}$ cm was introduced. The value of this artificial attenuation was chosen to achieve the necessary stability of the calculation and not to affect the amplitude of the pulse. The mean-square deviation between the in-plane surface velocity wave form of the second pulse [Fig. 3(b)] and the calculated one was minimized by varying the three nonlinear constants $\varepsilon_{1,2,3}$. In this way a unique set could be determined for every measurement. In the numerical simulations the following values were used: moduli of the second order $C_{11} =$ 78.5 GPa, $C_{44} = 31.2$ GPa, density $\rho = 2.2$ g/cm^3, and the constant $\gamma = 1.39$.

The solid curve in Fig. 2(b) presents the best fit calculated with $\varepsilon_1 = -1.2$, $\varepsilon_2 = -0.3$, and $\varepsilon_3 = -1.7$. The fitting procedure was performed for five measured pairs of pulses and the average values of the nonlinear acoustic constants were evaluated as $\varepsilon_1 = -(1.0 \pm 0.3)$, $\varepsilon_2 = -(0.25 \pm 0.1)$, and $\varepsilon_3 = -(2 \pm 1)$. The good agreement of the observed and calculated wave forms shows that the improved theory correctly describes the process of nonlinear transformation, although not all features were reproduced quantitatively by the simulated wave forms.

The nonlinear acoustic constants were calculated using elastic moduli of second and third order. The elastic moduli of third order were measured previously with the hydrostatic compression and uniaxial loading methods: $C_{111} = (526 \pm 40)$ GPa, $C_{144} = (93 \pm 8)$ GPa, and $C_{456} = -(11 \pm 3)$ GPa [16]. Using these values we

calculated the following set of nonlinear acoustic constants: $\varepsilon_1 = -1.37$, $\varepsilon_2 = -0.25$, and $\varepsilon_3 = -2.9$. A somewhat different set of values of third-order constants was measured employing harmonic generation combined with an ultrasonic beam-mixing technique: $C_{111} = (648 \pm 50)$ GPa, $C_{144} = (54 \pm 3)$ GPa, and $C_{456} = -(13.2 \pm 0.8)$ GPa [17]. This set yields the following values: $\varepsilon_1 = -0.84$, $\varepsilon_2 = -0.25$, and $\varepsilon_3 = -4.2$. As can be seen, the values of ε_2 coincide, because this constant depends only on the moduli of second order. The main difference of the sets is in constant ε_3. This, as well as the bending in the in-plane velocity profile, could be caused by higher than second-order nonlinear processes not being taken into account. This hypothesis is supported by the observed variation of nonlinear acoustic constants with wave amplitude for bulk waves in fused silica [21].

In conclusion, the first experimental observation of the nonlinear transformation of SAW pulses during propagation and formation of shock fronts in the pulse profile is reported. The employed method for the generation of very high amplitude SAW pulses via a strongly absorbing layer has a broad range of applicability and can be used for a wide range of materials. A change in the propagation velocity of up to 0.6% was registered for the generated shock fronts. The three nonlinear acoustic constants used in the theoretical description of the nonlinear SAW profiles can be uniquely determined by fitting the simulated wave form to the measured SAW pulse. This method will contribute to a better understanding of the nonlinear elastic behavior of materials.

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