Excitation of higher harmonics in transient laser gratings by an ablative mechanism

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Laser-induced transient gratings were excited at the surface of an aluminum film by picosecond laser pulses. The counterpropagating surface acoustic waves with a fundamental frequency of 70 MHz that were launched were monitored outside the source region by laser probe beam deflection. The second and third harmonics were observed for ablative interaction of the laser radiation with the aluminum surface. A simple model is presented that describes the effects on the basis of nonlinear photoacoustic signal generation at the source due to a nonsinusoidal driving force. © *1998 American Institute of Physics.* [S0003-6951(98)03838-8]

The laser-induced transient grating (LITG) technique has been widely used to investigate, for example, transport properties in a number of different materials, such as liquids, on the basis of the counterpropagating ultrasonic waves launched by the optical interference pattern (see, for example, Refs. 1 and 2). Recently, a doubling of the acoustic frequency corresponding to the optical fringe spacing of the grating has been observed in experiments with carbon suspensions.[3] It was suggested that this nonlinearity in the generation of sound waves was due to high-temperature chemical reactions occurring between the surface carbon and the surrounding water.[3,4]

In a LITG surface experiment two temporally coincident laser pulses are crossed at the surface of a strongly absorbing material (a metal film in the present experiments). Absorption of radiation in the sample causes prompt spatially periodic heating and thermal expansion, which gives rise to counterpropagating narrow-band surface acoustic wave (SAW) trains.[5–9] For the detection of these SAWs usually Bragg diffraction of a delayed third laser pulse from the induced acoustic grating is monitored. Nonlinear SAWs were first observed in piezoelectric materials (e.g., $LiNbO_3$), where narrow-band nonlinear SAWs were generated by interdigital transducers. For the optical probing of harmonic generation usually the variation of the first and higher order diffracted probe laser intensities have been monitored as a function of acoustic power.[10–13]

In the present experiments we launched coherent SAWs with picosecond laser pulses and demonstrated that in the nonlinear excitation regime SRWs with higher harmonics can easily be excited. For optical recording outside the source we used a probe beam deflection technique for detection in the time domain.[14] Fourier transformation of the measured waveform was employed to visualize the frequency spectrum with higher harmonics originating from the nonlinear photoacoustic driving force at the source.

A frequency-tripled Nd:YAG laser (355 nm) with a pulse duration of 180 ps full width at half maximum (FWHM) was used to generate the transient gratings. The sample investigated was a 700 nm thick aluminum film evaporated onto a fused silica substrate (Herasil).

The laser generation of LITGs and detection of SAWs is shown schematically in Fig. 1. The two laser pulses were crossed at the sample surface, producing an optical interference fringe pattern of approximately 2 mm length. The width of the grating was reduced to about 0.5 mm by using a cylindrical quartz lens with a focal length of 63 mm. The spacing between two fringes, given by $\Lambda = \lambda/2\sin(\theta/2)$, was measured to be 48 μm, where θ is the angle between the two laser beams and λ the laser wavelength. To detect the SAWs we used the probe beam deflection method described in Ref. 14. The beam of a stabilized frequency-doubled cw Nd:YAG laser (532 nm) was focused by a gradient-index lens to a spot size of about 4 μm at the sample surface. The light reflected from the surface was collimated by a second lens and directed to a position-sensitive detector consisting of two fast PIN photodiodes (Hamamatsu S4751) with a cutoff frequency of 500 MHz and a fast microwave differential amplifier (BFI IBEXSA INA-10386). The detected signal measures the slope of the transient surface movement, and therefore represents the vertical surface velocity. For data acquisition a digitizing oscilloscope (Textronix TDS 680B) with an analog bandwidth of 1 GHz was used.

The probe spot was located outside the grating area in the direction of acoustic wave propagation, as indicated by the arrow in Fig. 1. The distance between the probe spot and

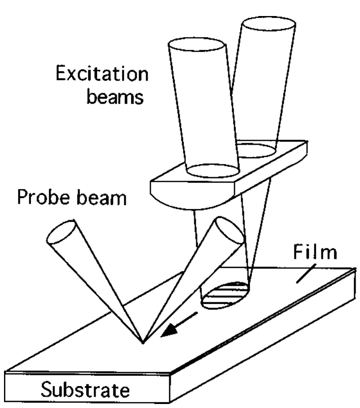

FIG. 1. Scheme of the experimental setup with two interfering ps laser beams and probe beam deflection monitoring of the acoustic wave outside the source.

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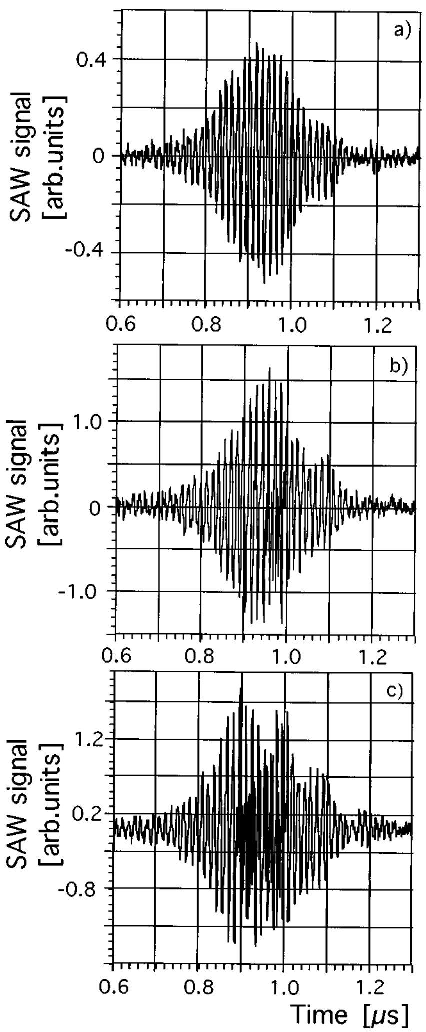

FIG. 2. Detected nearly linear and nonlinear transient grating signals for increasing laser pulse energies: (a) 0.5 mJ; (b) 2 mJ, and (c) 3.5 mJ.

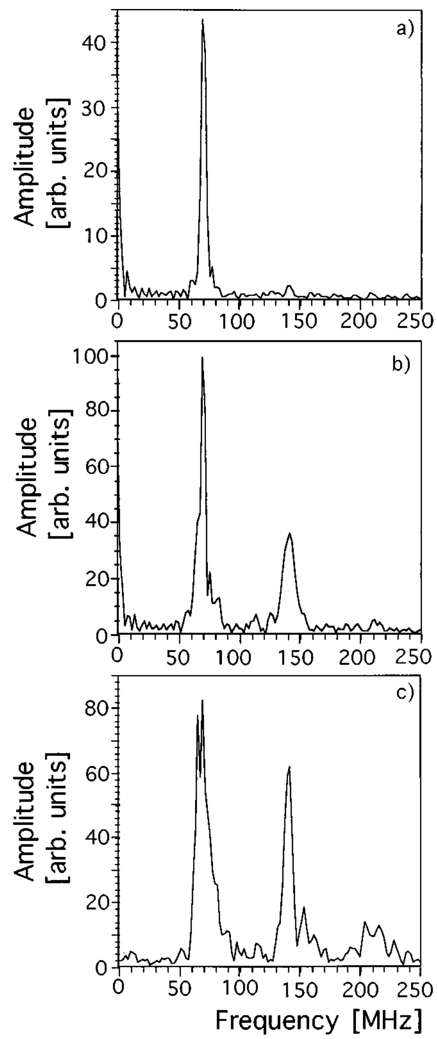

FIG. 3. Frequency spectra obtained by Fourier transformation of the corresponding grating signals of Figs. 2(a)–(c).

the edge of the grating pattern was determined to be 2 mm using the phase velocity of the fundamental frequency ($v = 3391$ m/s at 70 MHz) in the aluminum film and the arrival time of the grating signal (0.644 μs after the trigger).

The measured LITG-induced propagating SAW signals are shown in Figs. 2(a)–2(c). The energy of the pump beam was 0.5, 2, and 3.5 mJ, respectively. Due to the pulse-to-pulse instability of the laser pulse energy, these values are correct only to within 15% (average over ten shots). With increasing pulse energy the shape of the grating signal changes significantly. The rather smooth envelope of the double bell-shaped oscillatory signal at low intensity becomes distorted when the laser intensity is increased. The spatial intensity distribution of the laser radiation in the grating region is far from uniform and as a result the generation of higher harmonics was observed only in the central area with maximum power density. Note the increase of the signal strength with increasing laser pulse energy.

Figures 3(a)–3(c) show the frequency spectra of the propagating SAW signals obtained by Fourier transformation of the whole gratings of Figs. 2(a)–2(c). In the purely thermoelastic regime (laser intensity below the ablation threshold) only one frequency appears, the first harmonic at 70 MHz. In this case the width of the frequency profile should be relatively narrow. The excitation geometry used in this setup produced about forty visible interference fringes, fixing the SAW frequency theoretically to within about 2.5%. The frequency distribution observed experimentally at the lowest intensity [see Fig. 3(a)] is already larger, probably due to the influence of the higher harmonics just visible at the assumed threshold intensity of 3×10^8 J/cm^2 s and/or the dispersion effect observed in a film system.

As soon as the intensity of the pump pulses exceeds the ablation threshold of 3×10^8 J/cm^2 s in the aluminum system, the second harmonic at 140 MHz and, at somewhat higher intensity, the third at 210 MHz grow in the spatial region of maximum power density. As can be seen from the spectra the nonlinear generation process affects the first harmonic such that the peak at 70 MHz is not only broadened but split into two or more peaks, e.g., one at 70 MHz and another at 66 MHz [see Fig. 3(c)]. The nonlinear part of the transient grating in the irradiated area grows with laser intensity [see Figs. 2(a)–2(c)].

The deposition of thermal energy into the solid by the fast nonradiative relaxation processes exactly mimics the optical interference pattern at low intensities and leads to a corresponding impulsive thermal expansion of the material. The resulting spatially harmonic force excites a single-frequency surface acoustic mode. This thermoelastic SAW signal, observed for small laser intensities, below 3

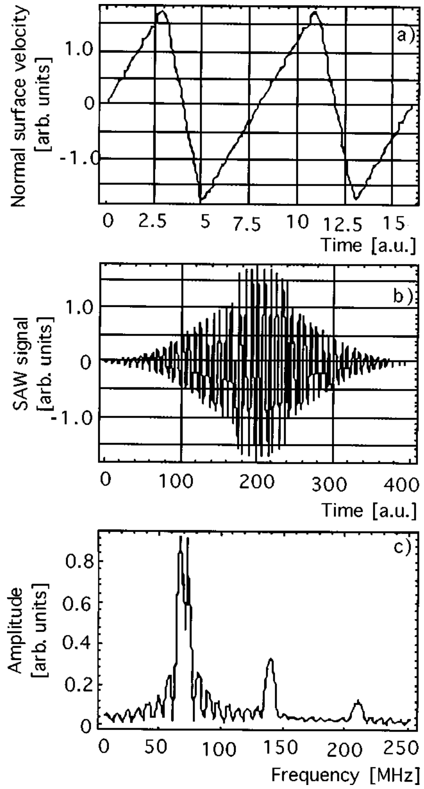

FIG. 4. Nonsinusoidal periodic function assumed for the nonlinear part of the source (a) corresponding grating signal obtained by combining the nonlinear with the linear parts (b) and resulting frequency spectrum (c).

$\times 10^8$ J/cm^2 s for aluminum, is assumed to be proportional to the laser intensity and the optical absorption coefficient.

For laser intensities above this value the light intensity exceeds the threshold for ablation in the center of the irradiated area, changing the mechanism of SAW generation there from thermoelastic to ablative. This nonlinear excitation process, caused by the recoil momentum of the ablating species, may modify the symmetry, polarity, phase, and strength of the periodic forces generating the acoustic waveform. The nonsinusoidal distribution of the acoustic field, with its extended frequency spectrum, is responsible for the formation of the overtones. The harmonics coincide with the higher acoustic modes of the grating, and therefore these frequencies survive from the extended frequency spectrum.

The total periodic acoustic field distribution of the grating, used for the present simulations, is composed of a linear part at the beginning, a nonlinear part in the middle and a linear part at the end. The intensity distribution in the direction of sound propagation is represented by a Gaussian envelope, which is shifted at the transition points between the linear and nonlinear behavior. Figure 4 gives an example of such an assumed nonsinusoidal periodic function describing the surface velocity [Fig. 4(a)], the total oscillatory signal with its linear and nonlinear parts [Fig. 4(b)] and the corresponding frequency spectrum resulting from the Fourier transform [Fig. 4(c)]. The purely thermoelastic parts in the leading and trailing edges of the oscillatory signal are described by pure sinusoidal functions. The nonlinear part in the middle of the grating was simulated by the simple sawtooth function shown in Fig. 4(a), which is suitable to describe the asymmetry, phase, and strength in the nonlinear region.

With this very simple ''ansatz,'' the effects of the nonlinear photoacoustic source distribution and its interaction with the linear parts of the grating can be studied. Due to the steep portions in the asymmetric periodic function, high frequency components are generated in the corresponding spectra. The sharply inclined transition region between the negative and the positive part of the periodic profile also influences the relative intensity distribution of the harmonics. The width and shape of the frequency profile of the first harmonic strongly depend on the phase relation between the linear thermoelastic parts and the nonlinear ablative part of the signal. The goal of this simulation was not a quantitative description of the measured frequency spectrum but to show on the basis of a very simple periodic function how the main features of the nonlinear behavior can be explained.

In conclusion, the easily observable generation of higher harmonics in surface LITGs and the resulting counterpropagating SAWs allows the accurate measurement of the threshold of ablation in strongly absorbing materials such as metals. As laser excitation is increased more energy is directed into harmonics of the fundamental grating frequency, distorting the wave. Since in a frequency up-conversion process the elastic energy is concentrated in a smaller depth, strong nonlinear behavior is indeed expected for surface waves. Compared to the use of interdigital transducers, the laser method for launching nonlinear SAW gratings is also applicable to nonpiezoelectric materials. As demonstrated, time-resolved detection of SAWs is possible outside the source region with the probe beam deflection method. Therefore, this relatively simple probe technique allows the attenuation of the propagating acoustic wave to be measured. The generation of the second and third harmonics at the source could be simulated by assuming a simple asymmetric sawtooth function with a steep transition in the profile.

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