

GENERATION OF ULTRASOUND BY SHORT LASER PULSES AND ITS APPLICATION IN PHYSICS AND NDE¹⁾

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In this paper I review the generation of short ultrasonic pulses via the thermoelastic effect in solids. Besides the basic physical principle of sound wave generation, I discuss also its spectrum, the absolute magnitude of the sound amplitude, and the conversion efficiency for longitudinal plane waves. Since the gradient of the temperature is at the origin of ultrasound generation, focussed laser beams cause shear waves and surface waves. At least for surface waves there exists a certain ratio of wavelength/laser-spot diameter such as non-destructive grain-size determination in thin metal sheets, crack depth determination by surface waves, and at large laser powers the exploitation of the resulting short ultrasonic pulses for the measurement of the electric charge distribution in polymers.

ГЕНЕРИРОВАНИЕ УЛЬТРАЗВУКА ПРИ ПОМОЩИ КОРОТКИХ ЛАЗЕРНЫХ ИМПУЛЬСОВ И ЕГО ПРИМЕНЕНИЕ В ФИЗИКЕ И ПРИ ОЦЕНКЕ МАТЕРИАЛОВ ПОСРЕДСТВОМ АДЕСТРУКТИВНЫХ МЕТОДОВ

В работе дается обзор по генерации коротких ультразвуковых импульсов в твердых телах при помощи термоупругого эффекта. Кроме основных физических принципов генерирования звуковых волн, обсуждается также их спектр, абсолютная величина амплитуды звука и коэффициент полезного действия преобразования для продольных плоских волн. Так как градиент температуры находится в точке генерирования ультразвука, сфокусированные лазерные пучки вызывают поперечные и поверхностные волны. По крайней мере для поверхностных волн существует определенное отношение длины волны к диаметру лазерного пятна, для которого существует максимальный коэффициент полезного действия. Приводятся также обзор применений, таких как адеструктивное определение размеров зерен в тонких металлических пленках, определение глубины трещины при помощи поверхностных волн и использование резонансирующих коротких ультразвуковых импульсов при больших лазерных мощностях для измерения распределения электрического заряда в полимерах.

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I. INTRODUCTION

The generation of sound by thermoelasticity has been known for a century [1, 2, 3]. However, at that time one could not explain the origin of the sound generated. With the advent of lasers the effect gained new interest, and one of the first papers which reappraised on this problem outlined the basic physical mechanism involved [4]. The energy of a short laser or microwave pulse absorbed within the skin depth of a solid or liquid surface is transformed into heat which diffuses a certain distance, the so-called thermal skin depth into the solid or liquid. Due to rapid thermal expansion ultrasonic pulses are generated. In this paper, I will restrict the discussion to solids because this is most important in the nondestructive evaluation (nde) of materials, although interesting phenomena occur in liquids as well [5—10].

II. BASIC THEORETICAL CONSIDERATIONS

Figure 1 displays the principle of pulsed thermoelastic sound generation. A short pulse of radiation such as a pulsed electron beam, a laser or a microwave pulse

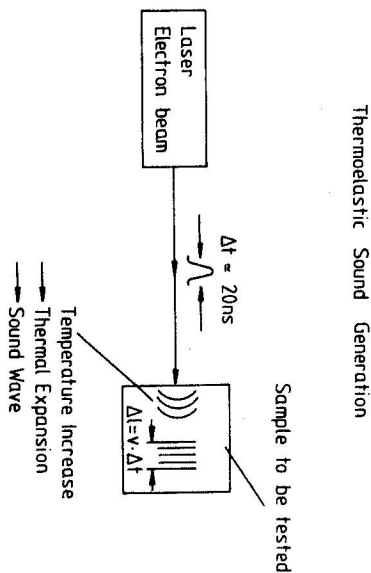


Fig. 1. Principle of thermoelastic sound generation using a laser pulse.

impinges onto the surface of a solid. In most practical cases the absorption length is quite small. For example in metals, the optical skin depth at wavelengths in the visible region is of the order of a few hundred Å. In contrast, the energy transformed into heat diffuses much further into the solid. It is possible to treat the problem in a simplified one-dimensional way provided the diameter of the electron or laser beam is large enough, and the solid is homogeneous within the diffusion length. The one-dimensional heat-diffusion equation describes the spatial and

temporal dependence of the temperature rise of the solid due to the incident radiation [11]:

$$\frac{\partial^2 T(x, t)}{\partial x^2} - \frac{1}{\kappa} \frac{\partial T(x, t)}{\partial t} = - \frac{A(x, t)}{K}. \quad (1)$$

Here, $T(x, t)$ is the temperature rise above equilibrium temperature, K is the thermal conductivity, κ is the thermal diffusivity, and $A(x, t)$ is the heat input per unit volume and per unit time. Let us assume now that we use laser pulses whose temporal shapes are gaussian. The only relevant frequency components of the problem are then the Fourier components of the envelope of the laser pulses because at the optical frequency the return to thermal equilibrium is so rapid that it is of no concern here. The solution of Eq. (1) for a sinusoidal heat input $A(\omega)$ of angular frequency is well known:

$$T(x, t) = (P_0/K) \sqrt{\kappa/\omega} \exp(i(\omega t - kx)) \exp(-kx). \quad (2)$$

$$\delta = 1/k = \sqrt{2\kappa/\omega}$$

Here, P_0 is the optical intensity absorbed on the surface of the material under consideration. Eq. (2) describes a temperature wave which is damped. Its attenuation length δ — also called thermal skin depth or diffusion length — is equal to its inverse wave-vector k . Typical values of thermal diffusion lengths are listed in Table 1. Within δ the solids expands, and the rate at which this expansion occurs is closely related to the frequency components of the laser pulse envelope.

Table 1
Thermal parameter of selected materials relevant to thermoelastic sound generation (from [36])

Material	Thermal diffusivity (cm ² /sec)	Thermal skin depth (μm) (at ω/2π = 10 MHz)	Thermal expansion (10 ⁻⁶ /K)
stainless steel	0.04	0.35	14.8
copper	0.95	1.7	16.8
silver	1.70	2.4	19.7
aluminum	0.98	1.8	23.8
Si ₃ N ₄ ceramic	0.16	0.7	3.2

The source term for the ultrasound is the thermal expansion. In order to account for this, one considers Hook's law including the stress σ_x caused by the temperature rise $T(x, t)$, and the equation of motion:

$$\frac{\partial^2 u}{\partial t^2} = \frac{1}{\rho} \frac{\partial \sigma_x}{\partial x} = \frac{1}{\rho} (cS - c\beta T(x, t)). \quad (3)$$

Here, β is the thermal expansion, c is the appropriate elastic modulus, S is the strain, ρ is the density, and u is the displacement. This equation can be solved taking into account the boundary conditions (uniform material for $x \geq 0$):

$$\sigma_x(x=0) = 0 \quad (4a)$$

i) for mechanically free surfaces:

$$u(x=0) = 0. \quad (4b)$$

The resulting stress as a function of sound frequency is then given by [4]:

$$\sigma'_x = \rho v_l^2 \frac{\beta P_0 \sqrt{\kappa}}{K} \frac{\sqrt{\omega}}{\omega + v_l^2/2\kappa} \quad (5a)$$

$$\sigma'_x = \rho v_l^2 \frac{\beta P_0 v_l}{K} \frac{1}{\omega + v_l^2/2\kappa}. \quad (5b)$$

Here, v_l is the longitudinal sound velocity. These equations allow one to make a detailed comparison with experiments. The limitations due to the one-dimensional treatment of the problem can be considered in actual experiments.

III. EXPERIMENTAL RESULTS ON PULSED THERMOELASTIC SOUND GENERATION

Many experiments have been reported of thermoelastic sound generation. It is obvious from Eq. (3) that the spatial gradient of the temperature causes the ultrasonic strain. Consequently, focusing the laser beam causes shear stresses and hence shear waves and surface waves [12, 13] (although not included in the one-dimensional treatment). In this respect thermoelasticity is analogous to piezoelectricity where the gradient of the electrical field is also at the origin of sound generation [14]. However, piezoelectric sound generation is an always linear effect with the ultrasonic strain proportional to the electric field strength, whereas in thermoelasticity the strain is proportional to the optical intensity (Eq. (6)). This has been verified by several groups [13, 15], and a typical experimental result is shown in Fig. 2 [16]. It displays the longitudinal ultrasonic strain S generated in the ceramic material Si_3N_4 as a function of absorbed optical intensity using a Q-switched ruby laser with a total pulsewidth of 50 nsec. It can be clearly seen that for low optical intensity the strain increases linearly. At optical powers larger than $\sim 20 \text{ MW/cm}^2$ this dependence becomes stronger than linear. At this optical power ablation of the surface of the solid sets in, and also non-linear effects in the

optical absorption of the solid may play a role. This critical intensity is different for different materials. It has been shown experimentally that the momentum transfer due to ablation enhances the longitudinal strain components [13].

Eqs. (5) also predict that the ratio of the strain levels for clamped and unclamped mechanical surfaces differ by almost a factor of 300 corresponding to $\sim 50 \text{ dB}$. This was indeed observed experimentally [17] (see Fig. 3). It originates from the fact that on a mechanical clamped surface the backward travelling wave generated within the thermal skin depth is reflected without a phase jump, and therefore adds constructively to the forward travelling part.

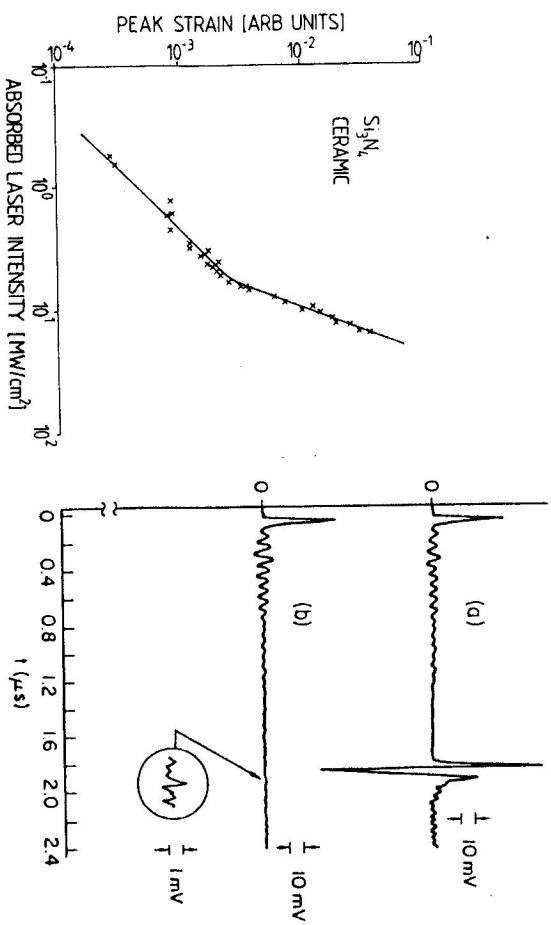


Fig. 2. Ultrasonic strain versus absorbed optical intensity in the ceramic material Si_3N_4 . At low optical power P_0 and in the thermoelastic regime the strain is linearly proportional to P_0 whereas for large P_0 ablation of the surface sets in and momentum transfer dominates the sound generation.

Fig. 3. Ultrasonic signals received by a piezoelectric transducer. The signals were generated by a laser impinging on a constrained surface (a) and on a free surface (b). There is $\sim 40 \text{ dB}$ difference in efficiency between the two. The energy of the laser pulse was rather small in the experiment (from [17]).

Only a few experiments related to the spectrum of the ultrasound generated have been reported. Mostly, this question was dealt with indirectly by observing the temporal shape of the ultrasonic signals. It is of importance, however, to know the spectrum exactly because it enables one to determine the minimal bandwidth of the detection systems applied. In particular using optical interferometers, it is desirable to limit the bandwidth as much as possible in order to increase their sensitivity. We

have therefore concentrated on this question. A backwall-echo sequence was generated for example in a Si_3N_4 sample of approximately 3 cm thickness. The echo-train was received by a piezoelectric transducer with a large bandwidth. Its output was amplified and digitized and the data were handled by a computer. Details of the experimental set-up can be found elsewhere [18].

The Fourier transform of any received backwall-echo can be written as:

$$G_i(\omega) = \tilde{S}(\omega) IL(\omega) \exp(-\alpha(\omega)x_i) \quad (6)$$

Here, $\tilde{S}(\omega)$ is the frequency spectrum of the strain, $IL(\omega)$ is the insertion loss of the transducer, $\alpha(\omega)$ is the absorption as a function of angular frequency, and x_i is the corresponding path length. $IL(\omega)$ can be accurately measured by using the reciprocity principle [19] and $\alpha(\omega)$ by the standard pulse echo-method. By Fourier transformation of a given backwall echo and deconvoluting it, it is therefore possible to determine experimentally the ultrasonic strain per frequency-band $\tilde{S}(\omega)$ as a function of frequency using Eq. (6). A typical experimental result is shown in Fig. 4 for Si_3N_4 . $\tilde{S}(\omega)$ first increases, passes through a maximum and then decreases rapidly as a function of frequency. Quite similar curves can be observed in polycrystalline aluminium [16], although its thermal skin depth differs by a factor of 2.5 from that of Si_3N_4 at the same thermal frequency. This behaviour can be understood quite easily. Since Eqs. (5) describe the ultrasonic strain at a given thermal frequency component, they only have to be multiplied by the Fourier transform of the laser pulse envelope:

$$S_L^i(\omega) = \frac{\beta P_0}{K} \frac{\sqrt{\kappa}}{\omega + v_i^2/2\kappa} \frac{\sqrt{\omega}}{\sqrt{\pi}} \tau \exp(-\omega^2\tau^2/4) \quad (7a)$$

$$S_L^e(\omega) = \frac{\beta P_0 v_i}{K} \frac{1}{\omega + v_i^2/2\kappa} \frac{1}{\sqrt{\pi}} \tau \exp(-\omega^2\tau^2/4).$$

One obtain from Eq. (7a) the dashed line in Fig. 4 which reproduces the shape of the experimental curve quite accurately. It is more problematic to reproduce its absolute magnitude because this requires the accurate determination of the optical intensity P_0 absorbed, which is rather difficult.

Eq. (5a) reflects the fact that at low frequencies the efficiency of sound generation is low because of the small ratio of the thermal skin depth to the ultrasonic wavelength. With increasing ultrasonic frequency, this ratio increases as can be easily seen from Eq. (5a). It would be maximal at $\omega = v_i^2/2\kappa$ corresponding to frequencies at least of the order of 100 GHz. The pulse width of the laser pulse, however, is far too large to contain any appreciable amount of power at these frequencies. From this argument it is obvious that only its absolute value but not the shape of the spectrum depends on the material under consideration. We have verified this by comparing the spectrum obtained in Si_3N_4 (low thermal diffusivity)

and aluminium (high thermal diffusivity). As mentioned above the same spectrum was obtained in the two materials. As can be easily seen from Eq. (7a) the maximum occurs at $\omega = 1/\tau$ where 2τ is the laser pulse width ($1/e$ points of the intensity).

For constrained surfaces we have not measured the spectrum up to now. In principle, there should be no increase of $\tilde{S}(\omega)$ as can be seen from Eq. (7b) in comparison with Eq. (7a) resulting in that only a pressure pulse should be generated.

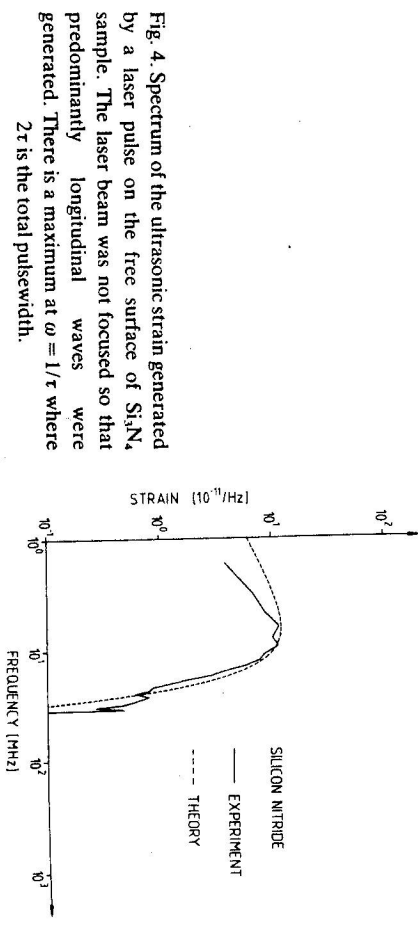


Fig. 4. Spectrum of the ultrasonic strain generated by a laser pulse on the free surface of Si_3N_4 sample. The laser beam was not focused so that predominantly longitudinal waves were generated. There is a maximum at $\omega = 1/\tau$ where 2τ is the total pulsewidth.

It should be mentioned that Eqs. (7) are valid provided $\lambda < L$ for all wavelengths where L is the length of the sample. If $\lambda \sim L$, then the boundary conditions have to be modified [20].

For interferometric detection it is also of great importance to know the absolute amplitudes u_0 obtained. We have estimated this quantity by carrying out a comparison experiment. At the frequency where the maximum in $\tilde{S}(\omega)$ as shown in Fig. 4 is observed, we generated normal ultrasonic backwall echoes using a rf-transmitter. The power P_r of the transmitter was adjusted so that the first echo displayed the same absolute amplitude on the crt as that one thermoelastically obtained at a certain P_0 . By measuring P_r and knowing $IL(\omega)$ it is straightforward to estimate u_0 using the reciprocity principle [19]. At an optical power just at the onset of ablation such as $\sim 20 \text{ MW/cm}^2$ in Si_3N_4 and at $\sim 5 \text{ MW/cm}^2$ in aluminium one obtains an amplitude of $\sim 5 \text{ \AA}$ and $\sim 3 \text{ \AA}$, respectively. Considering the large optical powers involved, this leads to a very low conversion coefficient of the order of $\sim 90 \text{ dB}$ [16]. As noted earlier, thermoelasticity is a non-linear effect, and hence the conversion efficiency itself depends on the optical power and renders the application of this quantity in this context less meaningful. Additionally, the large optical powers readily available with Q-switched lasers still result in appreciable signal/noise ratios of the ultrasonic signals.

As pointed out above, focusing the laser beam causes lateral thermal expansion and hence shear stresses. By focusing the laser diameter down to a diffraction limited spot one eventually ends up with an ultrasonic point source radiating all polarizations: longitudinal, shear and surface waves. This has been verified experimentally in detail [21, 22]. Due to the elastic boundary conditions and diffraction effects, it leads to a peculiar angular radiation pattern which can be

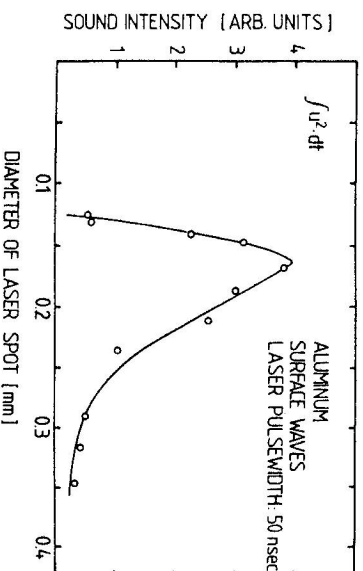


Fig. 5. Ultrasonic intensity (arb. units) of surface waves generated on a aluminium sample as a function of laser spot diameter. As can be seen clearly, a pronounced maximum in the efficiency occurs at a certain diameter d_m .

explained physically quite reasonably. On the theoretical side, there have been only a few attempts to extend the mathematical description beyond the one-dimensional theory [23].

Upon focusing the laser beam one expects the efficiency of shear and surface waves generation to increase because the lateral thermal stresses also increase. This has been verified quite recently [24]. Fig. 5 shows the ultrasonic intensity of surface waves on aluminium as a function of the laser spot diameter. The intensity of the surface wave was measured by a piezoelectric transducer using a wedge. Its position was kept constant on the surface of the sample and in addition its band-width was large enough to be sensitive to the whole spectrum. Fig. 5 clearly shows that the intensity first increases with the decreasing laser spot diameter d (but at constant optical intensity). It passes through a maximum and then decreases rapidly. The maximum occurs approximately when $\lambda_m \sim d$ where λ_m is the wavelength of the maximum in the spectrum of the ultrasonic pulses. This effect can be understood at least qualitatively: From each angular ring of the laser spot a surface wave is launched. The overall integrated intensity radiated from the spot depends on the relative phase of each radial component, and must therefore exhibit a maximum when for most of them constructive interference occurs.

At very large laser powers beyond the thermoelastic regime one can clearly observe the spectrum of the ultrasonic pulses broaden [18]. It is not yet clear in detail what causes this effect, and it might as well be the several factors contribute: momentum transfer by the material evaporated, enhanced optical absorption to plasma formation, increasing thermal expansion with increasing temperature, and eventually generation of higher harmonics by nonlinear elasticity.

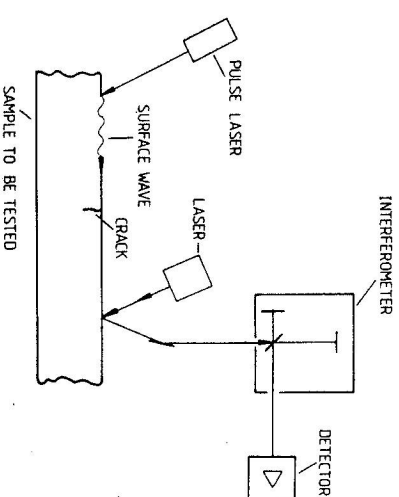


Fig. 6. Principle of the technique to estimate the depth of a crack by thermoelastically produced surface waves. The crack acts as low-pass filter on the spectrum of the surface waves which can be observed by the detection system in this case an interferometer (from [26]).

IV. APPLICATIONS

Together with optical detection, thermoelastic sound generation allows remote non-destructive testing of components particularly at high temperatures where conventional transducers would be destroyed [25]. Additionally, it is quite easy to scan optical beams over the surface of a component to be inspected. Besides this application there are several others which exploit the broad frequency spectrum of the ultrasound generated. Recently, a method has been proposed to estimate the depth of a crack extending from the surface into a material [26]. As can be seen from Fig. 6 a laser beam focused with a cylindrical lens launches a surface wave. A crack possibly present acts as a low-pass filter because of the finite penetration depth of the surface waves. With an interferometric detection system one observes the change of the spectrum received relative to other scan positions on the surface. This allows one not only to detect but also to estimate its depth.

In thin polycrystalline metal sheets it is interesting to know the average size d of the grains. It is well known that by exploiting the Rayleigh-scattering one can

determine the grain size nondestructively [27]. However, for components thinner than a few millimeters conventional ultrasound becomes increasingly difficult to apply because of lack of axial resolution. In contrast, by using the short ultrasonic pulses obtained by thermoelastic sound generation one can overcome this problem [28]. By a short laser pulse one generates a backwall-echo sequence in the metal sheet to be examined. Then with a transducer one observes the change in spectrum of any two subsequent echoes. For the spectrum of these echoes Eq. (6) holds. In this case the total attenuation $\alpha(\omega)$ is caused by two attenuation mechanisms. The inelastic part is caused by various effects such as thermal conduction loss, dislocation and magnetic losses, and varies approximately linearly with frequency in technical materials [27]. The elastic part is due to scattering by the ensemble of grains. Provided $\lambda \gg d$, one can therefore write for $\alpha(\omega)$:

$$\alpha(\omega) = a\omega + bd^3\omega^4. \quad (9)$$

Here a is a constant and b is the so-called scattering parameter. At least b is generally known for polycrystalline metals [28]. By comparing the experimentally obtained $\alpha(\omega)$ to Eq. (9) it is then straightforward to obtain d by a least square fit. We have verified this technique for polycrystalline metal sheets whose grain sizes extended from $\sim 10 \mu\text{m}$ to $\sim 100 \mu\text{m}$. For the best accuracy one has to adjust the laser pulse and hence the spectrum in order to obtain an appreciable amount of scattering within the limits given by the available signal/noise ratio of the detection system used.

In the so-called plasma regime when ablation of the surface sets in an interesting application in solid state physics has been reported [29, 30]. Depending on the experimental conditions almost step-like ultrasonic signals are generated whose rise-time can be of order of one nsec. Applied to polymers this would correspond to a spatial extent of the sound pulse of the order of a few μm . By placing a polymer foil in a condenser and generating such a step-wave on one of its faces, one can study the spatial distribution of electrical changes and dipole moments within the foil. Due to their presence the density variation accompanying the step-wave causes an opposite charge at the condenser electrodes which can be displayed on an oscilloscope. Knowing the sound velocity it is then possible to deduce deviations from homogeneity of the electrical charges present with μm resolution. In this way, for example, it has been possible to shed more light on the nature of piezoelectricity in PVDF induced by static electrical polarization [31].

There are other interesting applications and modifications of ultrasonic generation by thermoelasticity as well as by ablation which I shall not discuss here due to lack of space (see, eg. [32, 33, 34, 35]).

Summarizing I should like to point out that sound generation by short laser pulses is an interesting technique both from a physical and practical point of view.

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REFERENCES

- [1] Bell, A. G.: *Am. J. Sci.* 20 (1880), 305; *Phil. Mag.* 11 (1881), 510.
- [2] Tyndall, J.: *Proc. Roy. Soc. London* 31 (1881), 307.
- [3] Röntgen, W. C.: *Phil. Mag.* 11 (1881), 308.
- [4] White, R. M.: *J. Appl. Phys.* 34 (1963), 3559.
- [5] Carome, E. F., Clark, N. A., Moeller, C. F.: *J. Appl. Phys. Lett.* 4 (1964), 95.
- [6] Bunkin, F. V., Mikhailovich, V. G., Shipulo, G. P.: *Sov. J. Quant. Electr.* 6 (1976), 238.
- [7] Bozhkov, A. I., Bunkin, F. V., Gyrdev, L. L.: *Sov. J. Quant. Electr.* 6 (1976), 809.
- [8] Sigrist, M. W., Kneubühl, F. K.: *J. Acoust. Soc. Am.* 64 (1978), 1652 and references contained therein.
- [9] Lyamshev, L. M., Sedov, L. V.: *Sov. Phys. Acoust.* 27 (1981), 4 and references contained therein.
- [10] Lai, M. H., Young, K.: *J. Acoust. Soc. Am.* 72 (1982), 2000.
- [11] see for example Ready, J. F.: *J. Appl. Phys.* 36 (1965), 462.
- [12] Ledbetter, H. M., Moulder, J. C.: *J. Acoust. Soc. Am.* 65 (1979), 840.
- [13] Aindow, A. M., Dewhurst, R. J., Hutchins, D. A., Palmer, S. B.: *J. Acoust. Soc. Am.* 69 (1981), 449.
- [14] Jacobsen, E. H.: *J. Acoust. Soc. Am.* 32 (1966), 946.
- [15] Kaule, W.: private communication.
- [16] Betz, B.: *Diploma thesis*, Univ. of Saarbrücken (FRG) 1983. (unpublished).
- [17] von Gutfeld, R. J., Melcher, R. L.: *J. Appl. Phys. Lett.* 30 (1977), 257.
- [18] Betz, B., Arnold, W.: *J. de Physique* 44 (1983), C6—61.
- [19] Bömmel, H. E., Dransfeld, K.: *Phys. Rev.* 117 (1960), 1245.
- [20] Tronconi, A. L., Amato, M. A., Morais, P. C., Neto, K. S.: *J. Appl. Phys.* 56 (1984), 1462.
- [21] Hutchins, P. A., Dewhurst, R. J., Palmer, S. B.: *J. Acoust. Soc. Am.* 70 (1981), 1362.
- [22] Scruby, C. B., Dewhurst, R. J., Hutchins, D. A., Palmer, S. B.: in *Research Technique in Non-destructive Testing*, Ed. by Sharpe, R. S., Academic New York, 5 (1982), 281.
- [23] Lyamshev, L. M., Chelnokov, B. I.: *Sov. Phys. Acoust.* 29 (1983), 220.
- [24] Arnold, W., Betz, B., Hoffmann, B.: *J. Appl. Phys. Lett.* 47 (1985), 672.
- [25] Kaule, W.: *Proc. 8th World Conf. Non-destructive Testing*, 1976.
- [26] Kaule, W.: German patent application No. DE 32 17947 A1.
- [27] Goebels, K.: in *Research Techniques in Non-Destructive Testing*, Ed. by Sharpe, R. S., Academic, New York, 4 (1980), 87.
- [28] Arnold, W., Betz, B.: German patent application No. P34 12 615.5.
- [29] Alquie, C., Dreyfus, G., Lewiner, J.: *Phys. Rev. Lett.* 47 (1981), 1483.
- [30] Sessler, G. M., West, J. E., Gerhard, G.: *Phys. Rev. Lett.* 48 (1982), 563.
- [31] Gerhard-Multhaup, R., Sessler, G. M., West, J. E., Holdik, K., Haardt, M., Eisenmenger, W.: *J. Appl. Phys.* 55 (1984), 2769.

- [32] Brienza, M. J., de Maria, A. J.: Appl. Phys. Lett. 11 (1967), 44.
- [33] Scruby, C. B., Wadley, H. G., Dewhurst, R. J., Hutchins, D. A., Palmer, S. B.: Mat. Ev. Dec. 81 (1981), 1250.
- [34] Tam, A. C., Coufal, H.: J. de Physique 44 (1983), C6—9.
- [35] Cielo, P., Nadeau, F., Lamontagne, M.: to be published in Ultrasonics.
- [36] American Inst. of Physics Handbook. McGraw-Hill 1972; Ziegler, G., Hasselman, D. P.: J. Mat. Sci. 16 (1981), 495.

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