ARCHIVES OF ACOUSTICS
19, 3, 381–393 (1994)

ON GENERATING ULTRASOUNDS BY LASER IN POLYMERS

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The paper reports an experimental investigation of the possibility of generating ultrasonic waves by a *Q*-switched Nd:YAG laser of small power in polymers by the example of polyvinyl chloride. The possibility of creating acoustic-point-sources in visco-elastic materials is confirmed. Explanation of the mechanism of generation of ultrasonic waves is proposed. Directivity amplitude patterns of the acoustic sources are determined using ultrasonic broadband of resonance frequencies: 1 MHz, 2.25 MHz, 5 MHz, and a head for acoustic emision. The directivity frequency pattern for a head 2.25 MHz is also determined.

1. Introduction

Although the idea of the research on the application of light to the generation of acoustic waves, which comes from BELL [1], appeared quite a long time ago (1881), intensive experimental investigations were started in the early 1960 s, after the invention of the laser. The present paper is a contribution to this research. However, in order to have a better understanding of the validity and purposefulness of the investigation presented in this paper, it will be probably to provide at the outset a brief historical overview of the research in question. Thus, quite an extensive introduction to the present article is motivated by the fact that it contains an approach to this research which is not often met in the literature.

The problem of thermally generated elastic impulses, which is always involved in the generation of ultrasound waves by laser radiation, was considered for the first time by DANILOVSKA [2], who employed equations of classical thermoelasticity to the description of the phenomenon. Appearance of new sources of intensive light, lasers, which make possible generation of a strong photo-acoustic effect, drew more interest to this phenomenon in recent years. Very accurate sensors (capacitance transducer, mode of PZT ceramic) have become available for the measurement of elastic waves. Application of lasers for generation of elastic waves in solids was described for the first time by WHITE in 1963 [3]. He considered the problem of the way in which the change in the temperature of the surface of an elastic half-space exerts an influence on stresses in the investigated point of a half-space depending on time and distance from its surface. A theoretical description (one-dimensional model of wave generation in solid body by a laser) was given by READY in 1965 [4] and BUSHNELL and MCCLOSKEY in 1968 [5]. In the first experimental investigations lasers of a very high power (greater than 10 MW) were used. It always caused a significant damage of the metal surface. The metal surface on which the waves were generated was either not modified (e.g. PENNER and SHARMA — 1966 [6]) or modified (e.g. FELIX — 1974 [7]). A common feature of these first works was also the fact that not much attention was given to the kinds of waveform which could be generated. Although LEE and WHITE showed in 1968 [8] that Rayleigh's waves could also be generated by a Q-switched ruby laser, until 1979 there were no publications which dealt with simultaneous laser generation in metal of longitudinal, shear and Rayleigh's waves. It was LEDBETTER and MOULDER in 1979 [9] who showed that neodymium laser of high energy (0.3–1 J) and focused light beam could simultaneously generate volume and surface waves in metals. AINDOW *et al.* demonstrated in 1981 [10] that a Q-switched ND:YAG laser of a much lower energy (ca 30 mJ) could be used to generate ultrasound pulses, both in aluminium and in steel. He observed generation of pulses of volume and surface waves with densities of the power of laser beam both below and above the ablation threshold, after which damage of a metal surface follows. It was perhaps the first work in which the mechanisms of wave generation were divided into the thermoelastic and ablation mechanisms, and their relative efficiencies was determined. In that investigation a piezoelectric transducer was employed as the receiver. It distorted, to a certain degree, the output electric signal which was the image of the recorded wave. It is sufficient in many applications that a thermoelastic source of ultrasound can be treated as a point dipole and the ablation source- as a point force normal to the free surface SCRUBY *et al.* 1980 [11]. Equations of directivity patterns for the thermoelastic and ablations sources were presented by SCRUBY *et al.* in 1982 [12]. Theoretical calculations and experimental measurements of the shape of the impulse on the surface were presented by DEWHURST *et al.* in 1982 [13]. In 1980, Scruby proposed to use, as model of the thermoelastic source on free surface of metal pair of equivalent forces acting parallel to this surface (Fig. 1a). In 1984 [14] ROSE expounded Scruby's model basing on point temperature source acting on the metal surface, and provided a precise analytical solution. In 1984 [15] WADLEY showed that experimental measurements were in good agreement with the above models, with the exception for a small positive "precursor" in the epicenter, whose source of origin was not well-known. The calculated and experimental directivity patterns complied with each other, in particular if finite dimensions of both the acoustic source and the receiver were taken into consideration. In 1986 [16] DOYLE extended Rose's work by integrating Green's elastic function in the whole material volume in which temperature was growing. He showed that the finite dimensions of the source followed from thermal diffusion inside the plate and they were the main cause of the "precursor" occurring in the waveform in the epicenter. SCHLIECHERT [17] and AUSSEL [18], using different calculation methods, confirmed independently Doyle's results. In 1989 [19] CONANT and TELSCHOW applied the Hankel-Laplace transform to the calculation of laser-generated thermoelastic disturbance in a plate (they neglected thermal conductivity, but took into account finite depth where electromagnetic wave was absorbed). Those authors suggested that finite dimensions of the source in time and space could be the cause of the "precursor" to appear. In 1989 [20], MCDONALD provided a new description of the source of laser-generated ultrasounds. He was basing on the generalized theory of thermoelasticity, and calculated numerically the inverse Henkel-Laplace transform. This author suggested that the "precursor" resulted from conversion of thermal wave to the acoustic one at the plate surface. An essentially new element resulting from the generalized theory of thermoelasticity is the adoption of the hyperbolic equation of heat transport, which causes finite velocity of heat propagation (normally a little higher than the velocity of longitudinal wave).

The ablation source of ultrasounds is described in a less precise way, but more intuitively. Physical phenomena occurring during that mode of laser work were described by READY in 1971 [21] and next by KREHL *et al.* in 1975 [22]. Directivity pattern of ablation source was provided by HUTCHINS *et al.* in 1981 [23] for longitudinal wave and COOPER in 1985 [24] for shear wave. The shape of ablation-generated waves in the epicenter was given by DEWHURST *et al.* in 1982 [13]. Approximate model calculations for ablation source were given by AUSSEL *et al.* in 1988 [18], as well as by SCRUBY and DRAIN in 1990 [25]. Problems of laser generation and ultrasound propagation were much less frequently analyzed for plastics and composite materials than for metals. In 1971 [21], READY determined the temperature distribution in non-metal (a one-dimensional model) after absorption of an electromagnetic wave sent from the laser. In 1985 [26], BOURKOFF and PALMER were perhaps the first to investigate the possibility of a small power laser generation (using visible light) of ultrasounds in non-metals (polyamid reinforced by glass fibres and epoxy reinforced by carbon fibres). They used a PZT transducer or interferometer as a receiver. In 1988 [27] BUTTLE applied a pulsed laser and point piezelectric transducer to investigate the propagation of elastic waves in composites of epoxy resin reinforced by glass or carbon fibers. In 1989 [28] CASTAGNEDE *et al.* investigated the elastic properties of polyester reinforced by glass fibers. A pulsed laser was the source of ultrasounds and a point transducer from PZT-ceramic was used as the receiver. In 1990 [29] TAYLOR *et al.* applied an industrial laser TEA CO_2 for ultrasounds generations in polymers. He obtained a good conformity of the results of the waveform measurements with Rose's theory [14], under the assumption that the ultrasound source is at a long distance ca 200 μm from the material surface. Taylor's results are confirmed by [19], in which an experimental and theoretical investigations was made on the effect of the position of the thermoelastic source of ultrasounds under the surface of material, up on the waveform. In 1993 [30] POUET and RASOLOFOSAON applied a laser to the generation and reception of ultrasound waves in different polymers, in order to measure internal friction and dispersion of the velocity of longitudinal wave. In 1993 [31] CORBEL *et al.* measured and calculated the directivity amplitude pattern of laser — generated waves in composites of various degrees of an isotropy, made from epoxy resin reinforced by carbon fibers.

The object of interest of the present Author is the propagation of laser-generated ultrasound waves in plastics, and therefore this paper refers to the investigations [19, 29, 30] and to some, degree, it is their continuation. The purpose of this work was to carry out experiments which would enable: 1) examination of the possibilities of a small power pulsed laser generation of ultrasound waves in non-metals of visco-elastic properties, on the example of isotropic polyvinyl chloride; 2) determination of the directivity patterns of the acoustic point source using commercial ultrasonic heads made from PZT-ceramic. There is a need to provide more precise interpretation of the results of acoustic measurements in non-metals and composite materials by the acousto-ultrasonic and acoustic emision methods. Therefore, the subject of this investigation will be the problems connected with optical generation and propagation of elastic waves. It seems relevant to provide a short discussion in Section 2 of the basic mechanisms of this generation in order to familiarize the Reader with the description of experimental investigations provided in Section 3.

2. Basic mechanisms of optical generation of elastic waves

2.1. Thermoelastic excitation

A laser beam, incident on a free surface of a optically opaque solid body, is partly reflected and partly absorbed. In metals, absorption of an electromagnetic wave is mainly caused by interaction with its electrons and it occurs on the so-called skin layer of thickness of about 0.1 nm. A significant role is also played by heat diffusion during generation of an elastic wave. It causes the source of sounds to be "displaced" inside the material. In polymers the coefficient of absorption γ of the electromagnetic wave, caused by vibrations of atoms or group of atoms excited by the electromagnetic wave, is relatively small contrary to the case of metals. It causes the wave to penetrate inside the material to a significant depth ($d = 1/\gamma = 0.1$–1 mm). For polymers the phenomenon of heat diffusion can be negligible and heat conductivity is very small. As a result, heat flow from the area where the electromagnetic wave was absorbed is negligible (in the scale of the source time) and it is about 0.1 nm. The energy of laser radiation absorbed is transformed into heat. In this way the volume element in which absorption of electromagnetic radiation occurs becomes heated and deformed. Instantaneous spatial deformation of this volume element leads to radiation of the elastic waves (Fig. 1a). In the heated volume element a compressions wave (L) is initially created. Shear (S) and surface (R-Rayleigh) waves are created on the surface of a solid as a result of transformation of the compressions waves. The compression wave falls on the surface of a solid also generates a head wave (H), its front having the form of a spherically truncated cone and which is tangential, under a precisely defined angle, to the shear wave (Fig. 2). Volume waves generated in the isotropic medium are propagated approximately as spherical waves. The mechanism of thermoelasticity of generating acoustic wave occurs most frequently under densities of laser light powers smaller than 10 MW/cm^2. Increasing the power density over this value, we can trigger an additional mechanism which generates acoustic waves, the so-called ablation mechanism. Further growth of the power density up to about 1 GW/cm^2 will cause a continuous change of the signal structure and a systematic increase in its amplitude. Above this power value, the plasma formed on the surface of the sample screens the access of laser radiation to the material and the signal amplitude decreases.

2.2. Ablation excitation

Under the density of power specific for each material the ablation mechanism, which consists in rapid evaporation of a part of the material, leaves behind a small crater in the surface layer. It follows from the principle of conservation of momentum and impulse force that the above loss of material in the surface later is the cause of the effect of a certain instantaneous pressure exerted on the crater surface — in this way an acoustic

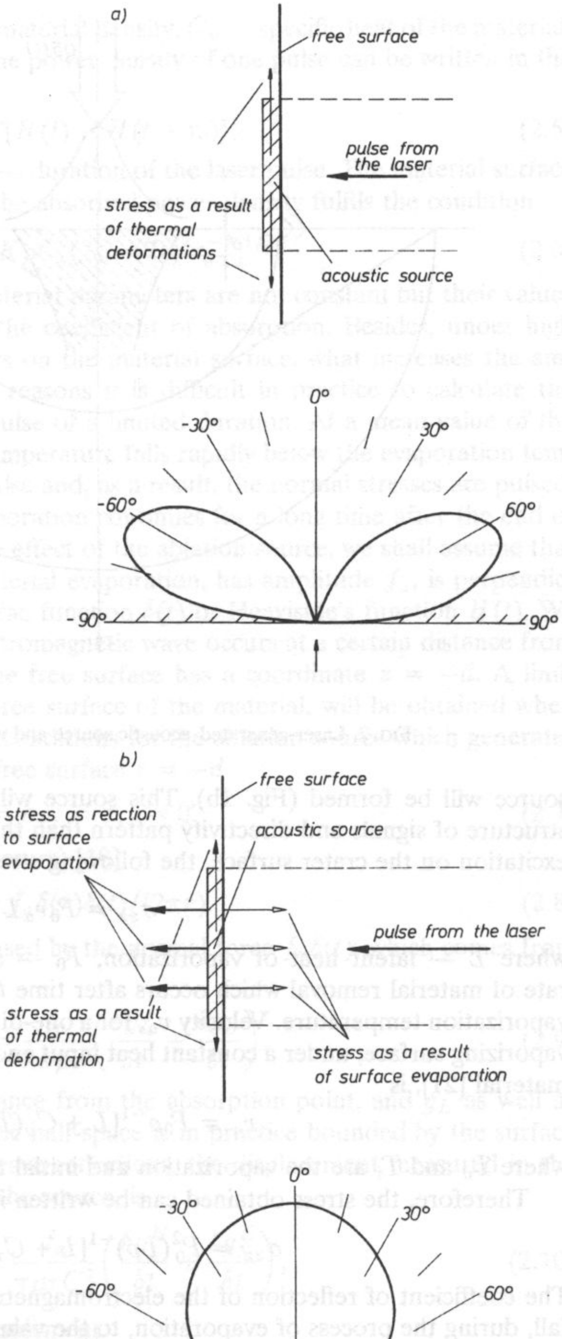

FIG. 1. Model of acoustic source and its directivity pattern for dilatation component of acoustic field according to [12] a) thermoelastic source, b) ablation source.

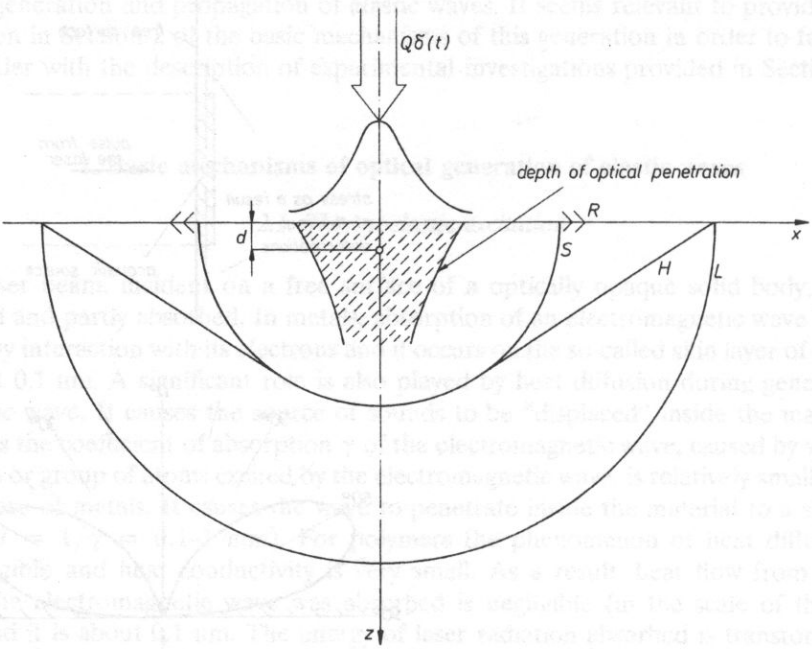

FIG. 2. Laser-generated acoustic source and wave-fronts of the wave formed.

source will be formed (Fig. 1b). This source will generate elastic waves of a different structure of signals and directivity pattern than the thermoelastic source. Under ablation excitation on the crater surface, the following stresses [32] are formed:

$$\sigma_{zz} = P_0 v_a L^{-1}, \tag{2.1}$$

where L — latent heat of vaporization, P_0 — absorbed power density and v_a is the rate of material removal which occurs after time t_v necessary to heat the material to the vaporization temperature. Velocity v_a, for a one-dimensional heat flow with a continuously vaporizing surface, under a constant heat input and a continuous removal of the vaporized material [21], is

$$v_a = P_0 \rho^{-1}[L + C_v(T_v - T_i)]^{-1}, \tag{2.2}$$

where T_v and T_i are the vaporization and initial temperature, respectively.

Therefore, the stress obtained can be written in the form

$$\sigma_{zz} = P_0^2(L\rho)^{-1}[L + C_v(T_v - T_i)]^{-1} \tag{2.3}$$

The coefficient of reflection of the electromagnetic wave which initially is 0.7 to 0.9 may fall, during the process of evaporation, to the value of 0.1. This causes an increase of the absorbed power density P_0. The time during which the surface temperature will increase to reach the evaporation temperature is about

$$t_v = 0.25\pi K \rho C_v (T_v - T_i)^2 P_0^{-2}, \tag{2.4}$$

where K — thermal conductivity, ρ — material density, C_v — specific heat of the material. If the laser works in the pulse mode, the power density of one pulse can be written in the following way

$$p(t) = P[H(t) - H(t - \tau_0)], \tag{2.5}$$

where $H(t)$ — Heaviside unit step, τ_0 — duration of the laser pulse. The material surface will undergo rapid evaporation when the absorbed power density fulfils the condition

$$P_0 > 0.25\pi[K\rho C_v(T_v - T_i)^2\tau_0^{-1}]^{1/2}. \tag{2.6}$$

Under such conditions the thermal material parameters are not constant but their values depend on temperature, particularly the coefficient of absorption. Besides, under high power densities a liquid phase appears on the material surface, what increases the amplitude of normal stresses. For these reasons it is difficult in practice to calculate the amplitude of stresses δ_{zz} for a laser pulse of a limited duration. At a mean value of the incident power density, the material temperature falls rapidly below the evaporation temperature after the end of each laser pulse and, as a result, the normal stresses are pulsed. However, at high power densities evaporation continues for a long time after the end of each laser pulse. In order to model the effect of the ablation source, we shall assume that the power pulse, equivalent to the material evaporation, has amplitude f_a, is perpendicular to the surface, and varies as a Dirac function $\delta(t)$ or Heaviside's function $H(t)$. We can assume that absorption of the electromagnetic wave occurs at a certain distance from the material surface (Fig. 2). Then the free surface has a coordinate $z = -d$. A limit case, when absorption occurs on the free surface of the material, will be obtained when we assume that $d \to 0$. The boundary conditions for the ablation source which generates a normal force are the following: for free surface $z = -d$

$$\sigma_{zz} = \sigma_{rz} = \sigma_{rr} = 0, \tag{2.7}$$

for surface $z = 0$ (where absorption occurs) [18]

$$\sigma_{zz} = f_a\delta(r)\delta(t)/(2\pi r). \tag{2.8}$$

Displacement on the acoustic axes caused by the normal force $f_a\delta(t)$, which comes from the ablation source, is [23]

$$u_z(r,\theta,t) = \frac{f_a}{4\pi\mu r}\left(\frac{\delta g_L}{\delta t} + \frac{\delta g_S}{\delta t}\right), \tag{2.9}$$

where μ — Lamé constant, r — distance from the absorption point, and g_L as well as g_S are given in the Supplement. Elastic half-space is in practice bounded by the surface $z = r$; if we take into consideration wave reflections, the displacement measured in the epicenter, i.e. on the acoustic axes of the source, is

$$u_z^E(r,\theta,t) = \frac{f_a}{\pi\mu r C_s^2}\left(\frac{\delta g_L^E}{\delta t} + \frac{\delta g_S^E}{\delta t}\right), \tag{2.10}$$

where g_L^E and g_S^E are given in the Supplement.

In reality, the acoustic source which is formed after exceeding the material ablation threshold, can be modelled by superimposing the thermoelastic point source over the point normal force, as well as by an adequate selection of the form of the excitation pulse. In this way it is possible to build a model which enables us to recreate the results of experimental investigations. However, the model does not provide any relations between the parameters of the electromagnetic wave and the material parameters.

3. Measuring stand and measurements

In Fig. 3 the diagram of the measuring stand is shown. In the investigations, for excitation of ultrasounds, a Q-switched ND:YAG laser with a built-in nonlinear crystal was used. The duration of a single pulse was 10 ns, the diameter of the laser beam was 1.5 mm, and its energy — 1.7 mJ. The frequency at which the pulses were emitted was 2 Hz. An electromagnetic wave of length $\lambda = 532$ nm and power density 10 MW cm^{-2} illuminated, at the point of intersection of diagonals, the flat surface of a semi-cylinder from polyvinyl chloride (PVC) of radius 45 mm and height 40 mm. This flat surface was free (one time it was constrained with glycerine and plexiglass plate). The wave power density was selected in such a way that the ablation threshold was only slightly exceeded and the PVC surface was slightly damaged. Piezolectric ultrasonic heads were used to measure the deformation caused by the ultrasound dilatation wave, in the direction normal to the cylinder surface. The measurements were made using the following ultrasonic heads; a head for the measurement of acoustic emision (EA) of resonance frequency 160 kHz, and the following ultrasound broadband heads; 5 MHz, 2.25 MHz, 1 MHz of bandwidth about 100%. The diameter of the transducers in all the heads was 12.7 mm. The medium which coupled the head with a semi-cylinder from PVC was an aqueous solution of glycerine. During the amplitude measurements of directivity patterns the head moved with a constant velocity on one plane along a parallel back and forth, in the angular range of $\pm 90°$, and it stopped at the measurements points. Measurement of the maximum amplitude of the first pulse of the dilatation wave performed every $1°$, within 4 seconds. The mean value of amplitude calculated from eight measurements at one point was taken as a result. The plane of measurements intersected the epicenter of the acoustic source. Both the control of the head movement and recording of the results were performed by a PC 386 computer. In the first stage of the investigation using all the heads, the amplitude directivity patterns $A =$

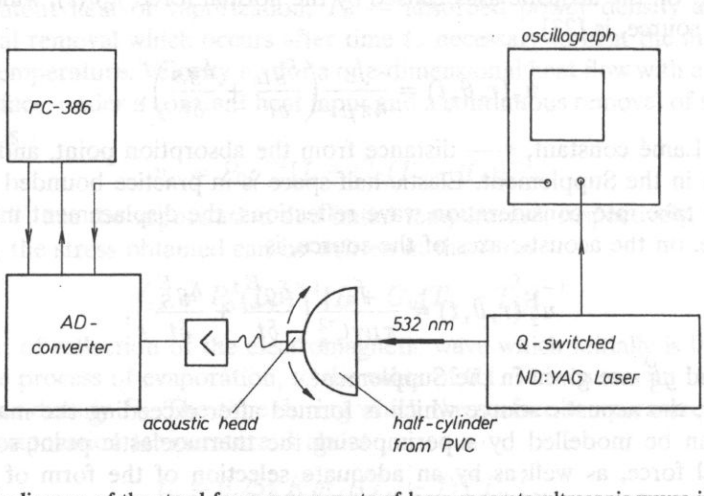

FIG. 3. Schematic diagram of the stand for measurements of laser-generate ultrasonic waves in PVC.

$f(\alpha)$, were measured, where α — angle of the head position with respect to the optical axis of laser beams radius. Four similar amplitude directivity patterns were obtained. They differ from each other mainly by absolute values of amplitudes. The highest amplitudes were obtained by the head 1 MHz ($A_{max} = 11$ V), and the smallest — by the head 5 MHz ($A_{max} = 1$ V). Amplitudes measured by the head 2.25 MHz ($A_{max} = 9$ V) were higher than those measured by the head for acoustic emission ($A_{max} = 5.5$ V). After an analysis of the results obtained, an ultrasound head 2.25 MHz of bandwidth 94.4% was selected for further investigation. Measurements of amplitude and frequency directivity patterns were performed using this head. During the measurements of frequency directivity patterns the head moved stepwise every 2.5° along a parallel line in the angle range ±90°, on the same plane where measurements of amplitude directivity patterns were performed. The first pulse of a dilatation wave was recorder at the points of measurement. Next a frequency spectrum was determined using a rapid Fourier transformer. The mean value (for two measurements made at one point) of frequency with the pulse of dilatation wave of maximum amplitude changes was taken as the result of measurements.

4. Result and discussion

The result of the measurements of maximum amplitude of the first pulse of the dilatation wave, depending on angle α, for different heads, are shown in Fig. 4. In order to compare the relations obtained $A_i = f(\alpha)$, where $i = 1, 2, 3, 4$, each curve was normalized to the form $A_{norm} = A_i/A_{i\,max} = f(\alpha)$, and next they were represented in a system of polar coordinates. It follows from the directivity patterns that, using the head 1 MHz, 2.25 MHz and a head to EA, we obtain as a matter of fact the same relations. The maximum value of amplitude occurs in the direction 0°, i.e. in the direction of the laser beam. A systematic decrease in amplitude occurs with increasing angle to the value of about 50°. In the range from 50° to 90°, a slight relative amplitude increase with a local maximum for $\alpha = 68°$ can be observed. This slight amplitude increase in the range 50°–90° is the effect of the thermoelastic mechanism in the acoustic source, for which, according to the

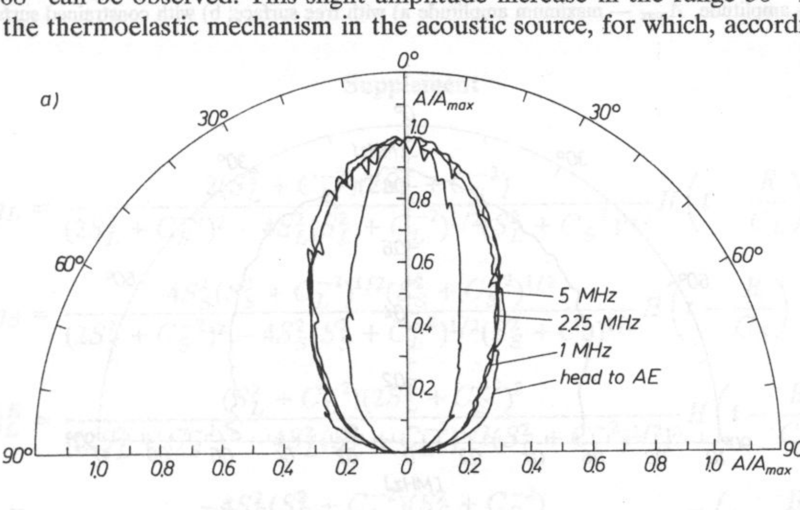

FIG. 4. Amplitude-normalized directivity pattern of laser-generated acoustic source in PVC determined by different heads; A — amplitude, A_{max} — maximum amplitude.

theory, maximum amplitude occurs for angle $\alpha = 64°$. As has been already stated, in Section 3, depending on the applied measurement head, we obtain different amplitude values, which results mainly from their different sensitivity. In the case of the head for the measurements of acoustic emission, its resonance frequency is 160 kHz and it is well below the frequency ca 1 MHz at which the maximum-amplitude disturbances occur. For this reason, the amplitude measured by this head is smaller than those measured by heads 1 MHz and 2.25 MHz.

A directivity amplitude pattern obtained from the second measurements made by the head 2.25 MHz, is represented in Fig. 5. It is evident from an analysis of the result obtained (in the case of the free surface) that the amplitude falls by 6 dB in the range of angle $\alpha = \pm 55°$, whereas in the range $\alpha = \pm 75°$, the amplitude falls by 12 dB. Such a form of the directivity pattern confirms that we have to do with an ablation mechanism.

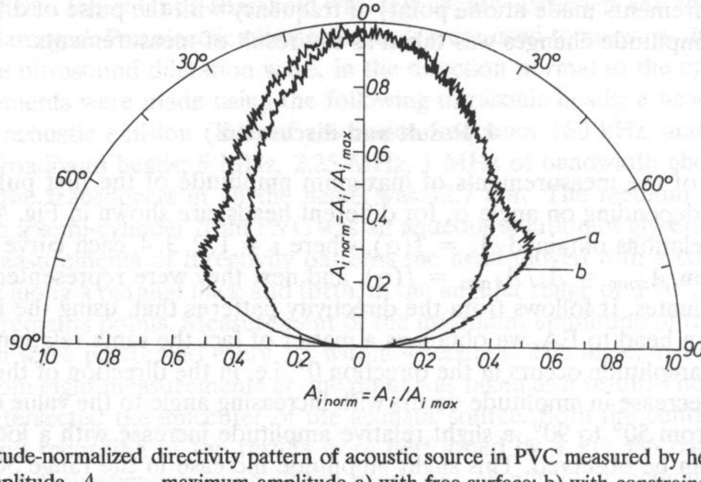

FIG 5. Amplitude-normalized directivity pattern of acoustic source in PVC measured by heads 2.25 MHz; A — amplitude, A_{max} — maximum amplitude a) with free surface; b) with constrained surface.

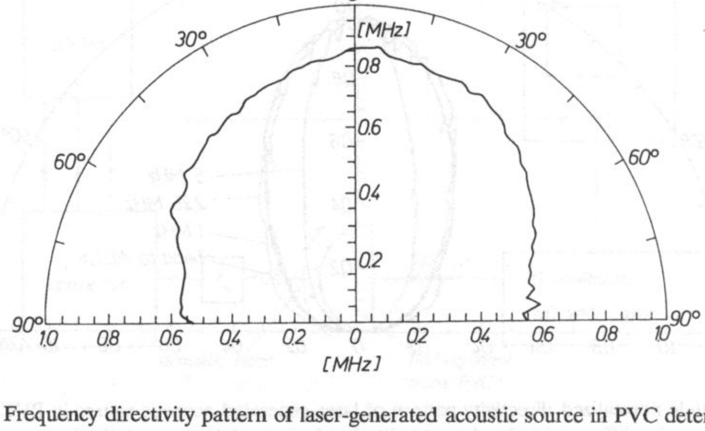

FIG 6. Frequency directivity pattern of laser-generated acoustic source in PVC determined by head 2.25 MHz MHz: f — frequency.

A directivity frequency pattern obtained from measurements by the head 2.25 MHz is represented in Fig. 6. It follows that the measured pulse of a dilatation wave of maximum amplitude varies with a frequency of about 0.85 MHz for $\alpha = 0°$. With an increasing value of this angle to $\alpha = 60°$, the frequency of changes of this pulse decreases systematically to the value of about 0.70 MHz. In the range of angles $\alpha = 60°–90°$, this frequency decreases more intensively than in the previous range, to the value of about 0.55 MHz for $\alpha = 90°$. Such a characteristic of the optically-generated acoustic source has not been known before. Variation in the vibration frequency, as a function of angle and PVC-viscoelasticity, is responsible for the form of the directivity amplitude pattern measured by the head 5 MHz. For his head, vibration amplitude is reduced by 6 dB in the range of angles $\alpha = \pm 20°$.

5. Conclusions

The investigation has shown applicability of the Q-switched Nd:YAG laser of wavelength $\lambda = 532$ nm, energy 1.7 mJ and duration of single pulse 10 ns, to the generation of ultrasounds in PVC. The investigation of directivity amplitude pattern has shown that laser light of power density 10 MW cm^{-2}, when incident on a free surface of PVC, is the cause of formation of an acoustic source, mainly due to ablation mechanism and, to a small degree, to the thermoelastic mechanism.

Normalized results of the investigation of the directivity amplitude pattern by heads 1 MHz, 2.25 MHz and EA represented in polar coordinates differ negligibly from each other. In the range of angles $\alpha = \pm 55°$, the amplitude decreases by 6 dB. Investigation of the directivity frequency pattern have shown that the frequency of the disturbances measured is not constant, and changes non-uniformly as a function of angle α. This property of the analyzed acoustic source and visco-elastic properties of PVC are the reason that the results of measurements by the head 5 MHz are markedly different from those obtained by the other heads.

Supplement

$$g_L = \frac{2(S_L^2 + C_L^{-2})(2S_L^2 + C_S^{-2})}{(2S_L^2 + C_S^{-2})^2 - 4S_L^2(S_L^2 + C_L^{-2})^{1/2}(S_L^2 + C_S^{-2})^{1/2}} H\left(t - \frac{R}{C_L}\right)$$

$$g_S = \frac{-4S_S^2(S_S^2 + C_L^{-2})^{1/2}(S_S^2 + C_S^{-2})^{1/2}}{(2S_S^2 + C_S^{-2})^2 - 4S_S^2(S_S^2 + C_L^{-2})^{1/2}(S_S^2 + C_S^2)^{1/2}} H\left(t - \frac{R}{C_S}\right)$$

$$g_L^E = \frac{(S_L^2 + C_L^{-2})(2S_L^2 + C_S^{-2})^2}{[(2S_L^2 + C_S^{-2})^2 - 4S_L^2(S_L^2 + C_L^{-2})^{1/2}(S_L^2 + C_L^{-2})^{1/2}]^2} H\left(t - \frac{R}{C_L}\right)$$

$$g_S^E = \frac{-4S_S^2(S_S^2 + C_L^{-2})(S_S^2 + C_S^{-2})}{[(2S_S^2 + C_S^{-2})^2 - 4S_S^2(S_S^2 + C_L^{-2})^{1/2}(S_S^2 + C_S^{-2})^{1/2}]^2} H\left(t - \frac{R}{C_S}\right)$$

$$S_L^2 = \left(\frac{t^2}{R^2} - C_L^{-2}\right) \qquad S_S^2 = \left(\frac{t^2}{R^2} - C_S^{-2}\right)$$

were C_L and C_S are velocities of longitudinal and shear waves respectively.

Acknowledgments

I am grateful to Dr E. NEUMANN and S. GRIPP of BAM in Berlin, Germany, for financial and scientific assistance during the investigations, as well as for discussion of the results. I am grateful to Professor J. OSIECKI for reading the manuscript and making numerous remarks.

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