Laser excitation of ultrashort acoustic pulses: New possibilities in solid-state spectroscopy, diagnostics of fast processes, and nonlinear acoustics

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The experimental results in the field of laser generation of acoustic pulses of duration less than 1 ns are reviewed. The various physical mechanisms of optoacoustic conversion are analyzed theoretically. Possibilities are shown for shortening the duration of optoacoustic pulses by increasing the intensity of the laser exposure. The prospects of initiating ultrashort, strong shock pulses with high-power femtosecond light pulses, are discussed.

1. INTRODUCTION. HIGH-POWER PICOSECOND AND FEMTOSECOND LASER PULSES IN OPTOACOUSTICS. ON THE PATH TO GENERATING EXTREMELY SHORT ACOUSTIC PULSES

The studies and developments of recent years in the field of the physics and technology of generating pico- and femtosecond laser pulses have led to truly record-breaking advances. The lengthy and demanding studies of many research groups over nearly 25 years following the quest of the shortest possible light pulse have been followed almost to the end. In the visible range of the optical spectrum pulses have been obtained with a length of $\tau_L \approx 6\times 10^{-15}$ s, i.e., pulse lengths of only three periods of the light oscillations.[1] In the far infrared region, experiments have been performed on the generation of short bipolar pulses—pulses of duration of "one period of oscillation," which amounts to $\sim 10^{-12}$–10^{-13} s.[2]

At the same time, the progress of the technology of generating femtosecond laser pulses, "focusing radiation energy in time," has led also to a new jump in the intensity scale, a breakthrough into the region of light intensities $I \approx 10^{19}$–10^{20} W/cm^2, energy fluxes $W_S \approx 10^4$ J/cm^2, and volume concentration of light energy reaching $W_V \approx 10^{10}$ J/cm^3 (Ref. 3).

Femtosecond lasers have revolutionized the optical spectroscopy of fast processes; the extremely short light pulses have made it possible to measure directly the time-dependent linear and nonlinear response of atoms, molecules, and condensed media.[4,5]

Experiments on the interaction of high-power femtosecond pulses with atoms, molecules, and condensed media have led to new and, in many cases unexpected results. These include the discovery of "above-threshold" effects of ionization of atoms and generation of high (of ~20–30th order) optical harmonics in gases in superstrong light fields,[6,7] "cold" melting of semiconductor single crystals,[8,9] and generation of dense femtosecond laser plasmas having an electron temperature reaching $T_e \approx 1$ keV.[10–12]

We can naturally expect that the invention of generators of short, high-power radiation pulses in other regions of the electromagnetic spectrum, of short electron clusters, and short and powerful elastic pulses will lead to no less significant results. Substantial results have already been obtained along this line. The laser plasma created by the fast heating of strongly absorbing semiconductor and metal targets by high-power femtosecond laser pulses is an effective source of pico- and subpicosecond pulses of soft x rays.[13,14] The high efficiencies of conversion of laser radiation into x rays attained in these experiments (now one speaks of efficiencies exceeding $\eta \approx 10\%$; see Refs. 14 and 15) make it practical to obtain soft x ray intensities of $I \approx 10^{12}$–10^{14} W/cm^2, i.e., intensities sufficient for performing experiments in the field of nonlinear x-ray optics.

The prospects of the physics and technology of ultrashort acoustic pulses also look very attractive. The spatial extent of an acoustic pulse of duration $\tau_a \approx 10^{-12}$ s is

$$L_a \approx c_a \tau_a \approx 5\cdot 10^{-7} \text{ cm} = 50 \text{ Å}.$$

In a solid this is larger than a lattice constant by only an order of magnitude, which is of fundamental interest for acoustic spectroscopy and diagnostics.

In spectroscopic language the transition to picosecond acoustic pulses means the mastery of the previously inaccessible range of acoustic frequencies exceeding $\nu_a \gtrsim 100$ GHz. The concentration of acoustic energy in such small time intervals enables one to rely on obtaining high sonic pressures exceeding $P_a \sim 10^9$ bar. Undoubtedly, as in the technology of short, ultrahigh-frequency electric pulses and that of ultrashort x-ray pulses, the exposure of a condensed medium to pico- and femtosecond laser pulses to excite ultrashort acoustic pulses is a highly promising route, especially in the laser excitation of coherent deformation pulses.

The mechanisms of optical excitation of such pulses are varied. Acoustic pulses are excited in optical breakdown, and in the ablation of matter by heating a solid target with a laser beam. In addition, methods based on nondestructive exposures of matter to optical radiation are effective. Figure 1 illustrates schematically the principal mechansims of optoacoustic interaction pertaining to generation of short coherent deformation pulses and coherent packets of optical and acoustic phonons. Figure 1(a) shows a nondestructive mechanism of thermoelastic sound generation practically universal for laser optoacoustics. Heating of a near-surface layer of thickness $l_p \sim \alpha^{-1}$, where α is the optical absorption coefficient, leads to non-steady-state thermoelastic stresses and to the excitation of a bipolar coherent deformation pulse. The relative slowness of the processes of heat dissipation is the reason for the slow response of this mechanism.

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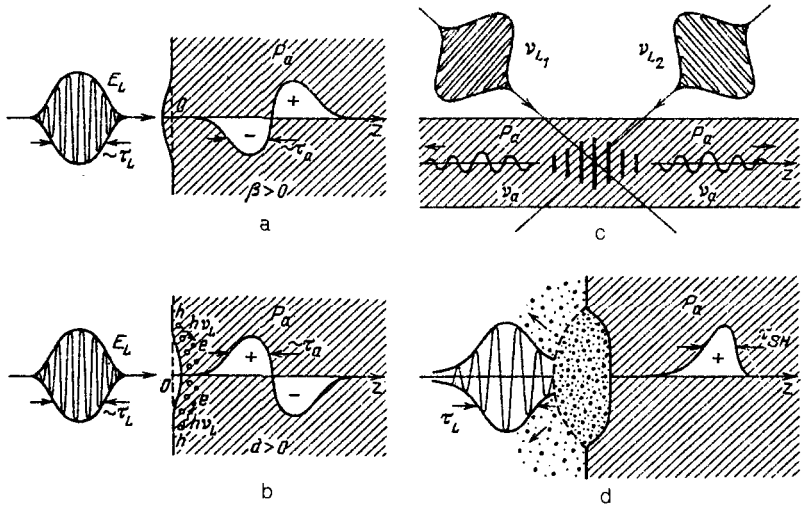

FIG. 1. Diagram of the optoacoustic conversion of short laser pulses. (a)—Generation of a bipolar acoustic pulse via deformation of the absorbing cyrstal by optical heating. b—Optoacoustic generation of a coherent deformation pulse in a semiconductor. The photoexcited carriers deform the crystal lattice; the polarity of the acoustic pulse of "one-period" duration depends on the sign of the deformation potential ($d \gtrless 0$). c—Generation of optical and acoustic phonons in a transparent medium by biharmonic laser pumping. d—Generation of shock waves arising from the pressure of the expanding laser plasma near the surface of the metal target.

For subnanosecond laser pulses, the duration of the acoustic pulse generated by the thermoelastic mechanism usually exceeds the time of passage of sound through the region of light absorption [$\tau_a \gtrsim (\alpha c_a)^{-1}$].

One can obtain substantially shorter acoustic pulses in semiconductor crystals by using the so-called concentration-deformation mechanism: carriers excited by the laser light induce stresses in the crystal lattice (Fig. 1b).

The corresponding external force in the acoustic wave equation for the lattice displacements has the form

$$F_n \approx -\frac{d}{\rho_0}\frac{\partial n}{\partial z}.$$

Here d is the deformation-potential constant and n is the density of the electron-hole plasma.

The rate of generation of photoexcited carriers at the *leading edge of the laser pulse is determined by the rise time* and intensity of the optical field. Moreover, in contrast to the temperature, the relaxation of the concentration of free carriers is determined not only by diffusion, but also by recombination of electron-hole pairs. The rate of the latter process can be increased by increasing the concentration of carriers, i.e., altering the density of light energy. The ensuing possibilities of controlling the evolution of the electron-hole plasma give promise of obtaining acoustic pulses of duration $\tau_a \approx \tau_L$ with $\tau_L \approx 10^{-12}$ s.

Optoacoustic sound generation is possible also in a transparent medium. Here the most effective mechanism is electrostriction. The corresponding coercive force in the acoustic wave equation has the form

$$F_S \sim \nabla\left[\rho_0\left(\frac{\partial \varepsilon}{\partial \rho}\right)E^2\right].$$

In a biharmonic optical field we have

$$\mathbf{E} = \mathbf{E}_{L1}\exp[i(2\pi\nu_{L1}t - \mathbf{k}_1\mathbf{r})] + \mathbf{E}_{L2}\exp[i(2\pi\nu_{L2}t - \mathbf{k}_2\mathbf{r})] + \text{c.c.} \quad (1.1)$$

Electrostriction results in the generation of phonons at the difference frequencies $\Omega = 2\pi(\nu_{L1} - \nu_{L2})$. The corresponding diagram of generation of short coherent bursts of acoustic (and optical) phonons is shown in Fig. 1c–the envelopes *of the components of the biharmonic wave of* (1.1) represent the short light pulses $E_{L1}(t)$ and $E_{L2}(t)$.

Finally, Fig. 1d illustrates one of the methods of generating elastic shock waves excited by an expanding laser plasma formed at the surface of a metal target. The duration of the acoustic pulse is largely determined by the rate of cooling of the laser plasma. Here the problems of reducing the pulse length are actually the same as in the generation of short x-ray pulses. It must be noted that the diagrams shown in Fig. 1 have been already studied for a rather long time in laser optoacoustics.

However, the recently developed technology of high-power femtosecond laser pulses, forms the basis for studies directed at revealing the ultimate potentialities of optoacoustic conversion and the ultimate characteristics of optoacoustic sound generators. At least three lines of study seem especially promising:

1. *Optoacoustic converison of femtosecond laser pulses* is absolutely the most effective method of obtaining *extremely short acoustic* pulses (here we must bear in mind the fact that now such pulses can be obtained in the far infrared, throughout the visible, in the near ultraviolet, and in the soft x-ray ranges of the electromagnetic spectrum). Theoretical estimates show that this is a question of coherent pulses of duration $\tau_a \gtrsim 100$ fs: several specific schemes for generating them have been proposed.

2. By using high-power femtosecond laser pulses one can realize the ultimate potentialities of optoacoustic generation of short coherent bursts of acoustic and optical phonons.

Femtosecond pulses enable one to realize the limiting optical strength of a transparent medium as determined by the tunneling ionization of the atoms (see Ref. 3 and Fig. 2, which shows the dependence of the optical-breakthrough threshold of a medium on the pulse duration [1)]). The ideas involve experiments on the nondestructive interactions of short laser pulses with a condensed medium, which can be performed at a light-intensity level reaching $I \approx 10^{13}$–10^{14} W/cm^2. We note that even experiments on coherent generation of phonons by means of biharmonic pumping performed at the level $I \approx 10^9$ W/cm^2 have recently enabled researchers to detect reliably the *nonlinear relaxation* of strongly nonequilibrium excitations of the crystal lattice; see Ref. 17 (*for the earlier history of studies on nonlinear relaxa-*

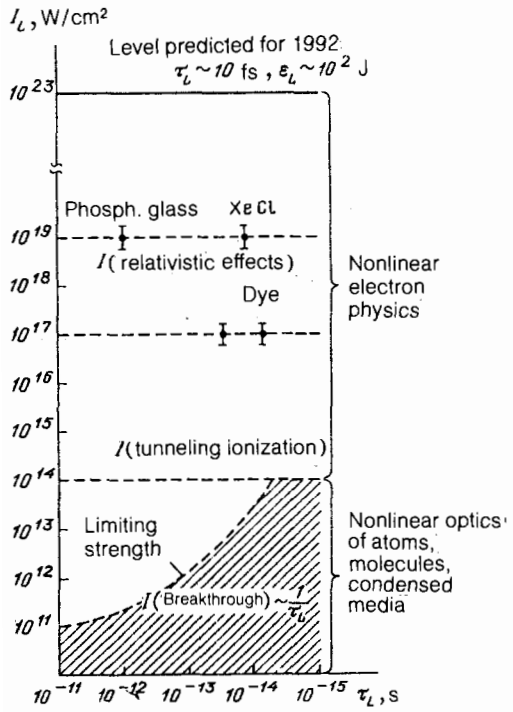

FIG. 2. Intensity-time diagram and the physics of the interaction of laser radiation with matter. The region of nondestructive interaction of laser pulses with a transparent medium is hatched. Some characteristic intensity scales are indicated here.

tion of nonequilibrium phonons, see Ref. 18).

3. Finally, high-power femtosecond laser pulses bring to acoustics, and especially to nonlinear acoustics, new schemes of optoacoustic interaction and stimulate new formulations of the physical problems. At an intensity $I \approx 10^{18}$ W/cm^2, the light pressure on the absorbing medium reaches $P_L \sim I_L/c_L \sim 3 \times 10^8$ bar. Laser melting of the surface of a strongly absorbing semiconductor crystal in the field of laser pulses with $\tau_L \approx 10^{-13}$ s occurs at an energy density at the target $W_S \approx 0.2$ J/cm^2, i.e., at an intensity $I \approx 10^{12}$ W/cm^2. At quite accesible energy flux densities $W_S \approx 10^4$ J/cm^2, the amplitude of the ultrashort shock pressure pulse, as the estimates show (see also Sec. 4 of this review), can reach $P_a \approx 10^9$ bar. We note also a unique new state of matter that arises as a result of the fast heating and ionization of the absorbing medium by femtosecond laser pulses–the femtosecond laser plasma.[19] It is heated to temperatures of 1 keV ($T \sim 10^7$ K) at the solid-state density $n_i \sim 10^{22}$–10^{23} cm^{-3}, and can possess also unusual acoustic properties As we see, the advances of the technology of ultrashort laser pulses pose completely new problems for physical and applied acoustics.

This review presents an analysis of the results already obtained in these new fields, and attempts to sketch the promising directions of study. We begin with a review of (Sec. 2) the physical mechanisms of excitation of short acoustic pulses and a summary of the experimental results; particular emphasis is placed on the technology of detecting short acoustic pulses. Section 3 presents the contemporary thoery of optoacoustic converters that employ the nondestructive action of short laser pulses on matter; the various physical mechanisms are compared from the standpoint of obtaining extremely short pulses. Section 4 examines the features of sound generation in the process of ablation of targets irradiated with high-power pico- and subpicosecond laser pulses. Major attention is paid to the factors that determine the duration and pressure of the shock elastic pulse excited under these conditions. Section 5 is devoted to analyzing the physical factors that influence the duration of ultrashort acoustic pulses. Some problems of nonlinear acoustics are discussed that are very closely associated with the progress in the field of pulsed hyperacoustics. We also discuss the possibilities of using nonlinearities in an optoacoustic converter (nonlinear absorption, screening of electric fields, nonlinear recombination of carriers, etc.) to shorten the acoustic pulse. Finally, in conclusion, we briefly discuss the prospects of using extremely short acoustic pulses in spectroscopy, diagnostics of short-lived nonequilibrium states in condensed media, in nonlinear acoustics, and the physics and technology of the action of high-power short acoustic pulses on matter.

2. PHYSICS OF LASER EXCITATION OF ULTRASHORT ACOUSTIC PULSES. METHODS OF GENERATION AND MEASUREMENT

The aim of this section is to present a view of the experimental technology of present-day subnanosecond and picosecond optoacoustics. As was pointed out in the Introduction, femtosecond lasers enable one to reach the limits imposed by the principle of physics in generation of short acoustic pulses and in creating high-frequency acoustic sources. The limiting frequency for acoustics $\nu_a \sim 10$ THz (at this frequency the acoustic wavelength is comparable to the interatomic distance in matter) can be excited by using optoacoustic conversion of a single laser pulse of duration $\tau_L \approx 10^{-13}$ s, or a light wave modulated in amplitude at terahertz frequencies.

The main problem here is the optimal choice of a fact-response mechanism of conversion that will provide a high efficiency in generating coherent deformation pulses.

Of greatest interest from this standpoint are the "nondestructive" methods of optoacoustic generation of bipolar (also called acoustic pulses of one oscillation period) or unipolar acoustic "videopulses" (see Fig. 1).

In addition, the ablation of strongly absorbing targets in the field of high-power femtosecond laser pulses leads to generation of short shock pulses (with a duration of several tens of picoseconds).

Just as in femtosecond laser optics, one of the key problems in picosecond hyperacoustics proves to be the development of adequate methods for measuring the duration and shape of the ultrashort pulses. The obvious difficulties of the direct methods of measuring picosecond times shift the emphasis to various indirect methods. Therefore the role of femtosecond laser technology in picosecond hyperacoustics is in no way restricted to exciting short acoustic pulses: the high sensitivity of the optical methods of measuring deformations can serve as the basis of the metrology of short acoustic pulses. The use of femtosecond laser pulses to measure short acoustic pulses enables one to develop systems unique in time resolution and to reconstruct the profiles of ultrashort acoustic pulses.

Pulsed (time-domain) acoustic spectroscopy based on optoacoustic methods of generation and acoustooptic methods of detection of sound is effectively used for determining the anisotropy of elastic moduli[20] and the spacecharge distri-

bution in dielectrics,[21] for profiling the rear (hidden) side[22] and measuring the thickness[23] of plates.[2)] The creation of optical generators of ultrashort acoustic pulses enables one to enhance the spatial resolution in acoustic spectroscopy and defectoscopy.[27] The technique opens up possibilities of studying elastic properties[28–30] and measuring thicknesses[30–32] of thin films, and determining the absorption coefficients of acoustic waves in the giga- and terahertz range of frequencies.[30,32–35]

The growth in recent years in the number of experimental[28,29,31–37] and theoretical[3,26,38–42] studies directed at developing optical methods of exciting and measuring ultrashort coherent deformation pulses is undoubtedly associated with the prospects of applying this contact-free, remote, and when necessary, nondestructive method for fast diagnostics of various materials.

2.1. Nondestructive and destructive methods of generating coherent deformation pulses

The experimental optoacoustics generation of sound pulses in a solid was obtained almost immediately after the invention of lasers.[43] By using a ruby laser, White[43] was able to obtain thermoelastic excitation of acoustic pulses with a duration of the order of tens of microseconds. In 1970 Brienza and De Maria proposed using ultrashort optical pulses for generating ultrashort acoustic pulses; at the same time the corresponding experiment was performed.[44] The radiation of a picosecond neodymium-glass laser was absorbed in a metal film deposited onto the end of an $LiNbO_3$ crystal. The exposure was made with a train of 100–150 optical pulses, the duration τ_L of each being $\tau_L \sim 1$–10 ps. The repetition period of the pulse in the train was 5 ns, which corresponds to a repetition frequency $\nu_r \approx 200$ MHz. The 15-mm-thick crystal simultaneously acted as a piezotransducer. In the experiment,[44] done at room temperature, acoustic waves were detected up to a frequency of 2 GHz, which corresponds to the tenth harmonic of the repetition frequency ν_r of the pulses in the laser train. Of course, these data do not yet suffice for an unambiguous conclusion on the characteristic duration of the optoacoustic pulses. The generation of sound at frequencies that are multiples of the repetition frequency in the train of subnanosecond laser pulses was realized also in the development of an optoacoustic microscope.[27] In this expeirment[27] acoustic signals were observed at the frequency $\nu_a \approx 5\nu_r \approx 1$ GHz. Yet, the profiles of ultrashort deformation pulses were determined experimentally only after more than 15 years had passed since the study of De Maria.[32,44] This delay is explained mainly by the difficulties of detecting high-frequency, broad-band acoustic signals.

The most often used nondestructive methods of laser excitation of ultrashort acoustic pulses, is the thermoelastic mechanism of generating deformation waves, which is associated with thermal expansion of the medium by optical heating.[28–34,40] Subnanosecond acoustic pulses were first measured experimentally in the study of Tam.[31] The main elements of the experimental setup used in Refs. 31 and 37 are shown in Fig. 3. The source of the pulsed optical radiation was an atmospheric-pressure nitrogen laser, which generated pulses of duration $\tau_L \approx 0.5$ ns of energy $\mathscr{E}_L = 1.5$ mJ at a wavelength $\lambda_L \approx 0.337$ μm. The radiation was loosely focused into a spot of dimensions 2×0.5 mm on the

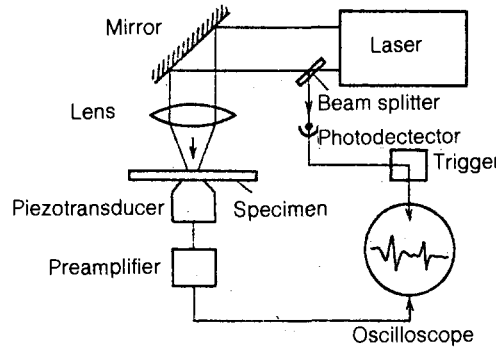

FIG. 3. Experimental diagram[31] of optoacoustic generation and detection of 1-ns acoustic pulses in solids.

surface of the specimen, which was a steel plate of thickness d from 12 to 260 μm. The longitudinal acoustic waves were detected with a thin piezoelectric transducer, a ZnO film of thickness $a \approx 5$ μm between two gold electrodes (0.1-μm thick). The piezotransducer was deposited onto one of the ends of a sapphire sound guide, the opposite end of which was in contact with the specimen being irradiated. The signal from the transducer passed through a preamplifier of bandwidth 5–100 MHz to an oscilloscope with a frequency band up to 500 MHz. Figure 4 shows the acoustic signal formed as a result of multiple reflections of the ultrashort deformation pulses in a stainless-steel foil of thickness $d \approx 14$ μm.[31] The full-width at half-height of the positive phase of the first of the signals entering the detector was $\tau_a \approx 0.8$ ns. However, we must note that the time of passage of sound through the ZnO film was of the order of the duration of the recorded pulses ($a/c_a \approx 0.8$ ns, $c_a \approx 6 \times 10^5$ cm/s is the velocity of longitudinal acoustic waves in ZnO). Therefore, the profiles of unidirectional deformation pulses are distorted during recording. In other words, for a given experiment in the frequency range $\nu_a \lesssim \tau_a^{-1} \sim \tau_L^{-1}$, the spectral sensitivity of the piezotransducer is quite nonuniform. The form of the reproduced signals can also be affected by the insufficient band width of the preamplifier and the oscillascope. The experimental apparatus (see Fig. 3) was subsequently automated.[37]

In the experiment of Ref. 36, laser pulses of duration $\tau_L \approx 70$ ps at a wavelength $\lambda_L \approx 1.06$ μm and energy $\mathscr{E}_L \approx 1$–10 mJ were used to excite ultrashort acoustic pulses. When the radiation was focused on the target, intensitites of exposure I_0 were attained in the range 0.14 GW/cm$^2 \lesssim I_0 \lesssim 1.4$ GW/cm^2. A diagram of the experiment is shown in Fig. 5. A condenser method of detecting the acoustic signal was used, in which the sample and the air gap formed two series-cou-

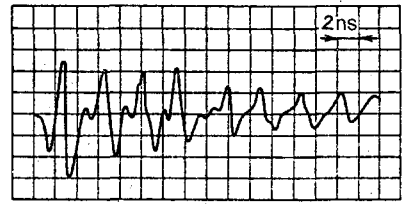

FIG. 4. Experimentally observed[31] optically excited acoustic pulse formed as a result of multiple reflections in a steel film (14-μm thick). Horizontal scale–2 ns div.

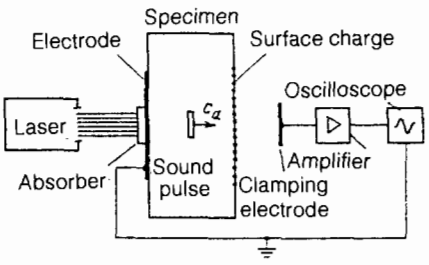

FIG. 5. Experimental diagram of the method of laser-induced pressure pulses[36] for specimens with one metallized surface.

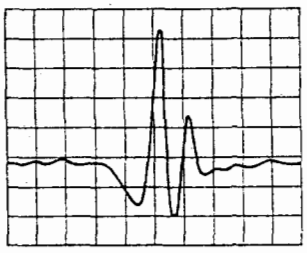

FIG. 7. Oscilloscope trace corresponding to recording[35] of acoustic pulses in water. Sweep rate 2 ns/div. Downward deflection of the oscillograph beam corresponds to positive pressure.

pled condensers. In the case of piezoelectric specimens (quartz), an electrode was deposited only on the front (irradiated) surface of the specimen. In addition, in some cases the rear surface of the specimen (Aclar) was first charged electrically, or an electrode deposited on the rear surface (Teflon) was charged. Over the electrode on the irradiated surface was deposited a layer of a material that strongly absorbed the laser radiation: either a graphite coating 2-μm thick or a layer of metal (Zn, Cd, Pb, Bi) 0.1–0.5-μm thick. The signal was viewed with an oscilloscope with a bandwidth of 1 GHz (rise time of the transient characteristic 380 ps). Signals up to 3 GHz could be preamplitifed. Figure 6 shows typical recorded profiles of short-circuit current pulses, which resembled the pressure profiles in the acoustic wave when electric charges existed only on the surface of the specimen. Depending on the material of the specimen and its thickness, the pulse lengths of the acoustic signals observed in Ref. 36 varied from 0.5 to 0.75 ns. The authors of Ref. 36 especially note that nonparallelism of the plates of the condenser and spatial delocalization of the electric charges can broaden the signals in this scheme of recording. The validity of the last statement is indicated by the results of experiments performed earlier,[45–47] in which the optoacoustic apparatus described in Ref. 36 was used specifically for determining the spatial distribution of charges in polymer films. The authors of Refs. 45–47 attributed the broadening of the recorded electric signals to 2–5 ns (and also their characteristic shape) to the inhomogeneous depth distribution of charges near the surface of films subjected, e.g., to irradiation with an electron beam. Thus, the broadening of the signal to $\tau \sim 2$ ns in Telfon corresponds to delocalization of electric charges in a layer of thickness $\sim \tau c_a \sim 2.6$ μm ($c_a \approx 1.3 \times 10^5$ cm/s is the velocity of sound in Teflon[47]). We note that the maximum of the charge density distribution can lie beneath the surface of the film being studied.[47]

Vodop'yanov *et al.*[35] reported laser generation of high-power sound pulses of subnanosecond duration in liquids.

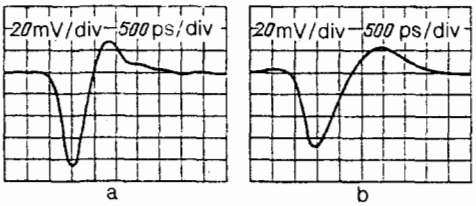

FIG. 6. Current pulses recorded at the emergence of a pressure wave at the rear surface of a quartz plate 190-μm thick 9(a), and at the front surface of an Aclar film 25-μm thick (b).

Single pulses from an erbium garnet laser ($Y_3Al_5O_{12}:Er^{3+}$, $\tau_L \approx 80$ ps, $\lambda_L \approx 2.94\,\mu$m, $\mathscr{E}_L \lesssim 60\,\mu$J) were used. The optical interaction occurred in hydroxyl-containing liquids with hydrogen bonds (water, ethanol, glycerol), which have extremely high absorption coefficients at the wavelength of the laser that was used. The sound receiver was a piezoelectric transducer based on a film of a polycyclic oriented organic compound of thickness $a \approx 1.2\,\mu$m. Figure 7 shows the profile of a signal recorded when the laser radiation was absorbed in water. The signal of negative polarity corresponds to compression at the leading edge of the acoustic wave. The duration of the first positive phase of the signal is 0.75 ns. The profile of the signal (see Fig. 7) is determined by the simultaneous affect of the ultrashort deformation pulse on the two surfaces of the transducer film (since $a/c_a \approx 0.46$ ns, where $c_a \approx 2.6 \times 10^5$ cm/s is the velocity of longitudinal sound in the material of the piezotransducer). On the whole, the situation today is that the use of different types of thin-film transducers and electric recording circuits does not enable one adequately to reproduce the form of acoustic pulses. Thus, in the experimental study[48] single laser pulses having the parameters: $\tau_L \approx 30$ ps. $\lambda_L \approx 1.06\,\mu$m, and $\mathscr{E}_L \lesssim 100$ mJ were used to excite ultrashort defomration pulses in a Si wafer ($d \approx 2$ mm). The signal from the ZnO piezotransducer was applied via a preamplifier of bandwidth 0.1–1300 MHz to an oscilloscope (500 MHz). In the opinion of the authors of this study, the duration of the recorded acoustic pulses ($\tau_a \approx 3$ ns) was determined by the response of the piezotransducer to an ultrashort pulse. Dewhurst and Al Rubai[23] were not able to reconstruct accurately the form of the acoustic signal. The leading edge of the deformation excited in an aluminum foil ($d \approx 500$ μm) by radiation having $\tau_L \approx 30$ ps, $\lambda_L \approx 1.06$ μm, and $\mathscr{E}_L \approx 8$ mJ had a duration $\tau_f \approx 1.7$ ns, which characterizes the speed of the acoustoelectric converter. A polarized piezopolymeric film of thickness $a \approx 9\,\mu$m was used as the receiver of the acoustic signal in this experiment.[23]

We note that as the intensity of the optical exposure was increased the experiments of Refs. 23, 36, and 48 marked a transition from thermoelastic sound generation to excitation of sound by the recoil pressure in the process of ablation of the target material. Importantly, according to the results of Ref. 36 the generation of ultrashort deformation pulses was also observed in the latter case.

However, this situation is apparently realized only at relatively low intensities of optical exposure. Actually, a considerable number of experiments have been performed for the purpose of exciting strong shock waves in condensed media by the use of picosecond laser pulses.[49–69] Shortening

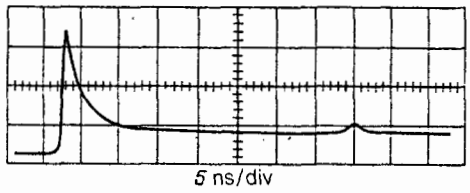

FIG. 8. Profile of the pressure initiated by laser exposure to the metallized surface of a quartz transducer.[51]

the deformation pulses was not an aim of these experiments. Nevertheless, analysis of the obtained results shows that the generated acoustic perturbations are not ultrashort, presumably because of the broadening of the shock pulses during nonlinear propagation.[49,50]

In the experiment of Ref. 51, a high-power picosecond optical pulse was applied ($\tau_L \sim 2$–4 ps, $\mathscr{E}_L \sim 10$–20 J) to one of the electrodes of a quartz piezotransducer (x-cut) of thickness $d \sim 0.25$–0.5 cm. With neodymium glass laser system the illumination of the surface of the transducer (area ~ 1 cm^2 [Ref. 52]) was uniform with intensity $I_L \sim 3 \times 10^{12}$ W/cm^2. Despite the strong reflection of the radiation of wavelength $\lambda_L \approx 1.06\,\mu$m from the aluminum and gold electrodes (reflection coefficients $R \approx 0.94$ and $R \approx 0.99$, respectively), ablation of the material led to generation of deformation waves with pressures of several kilobars. A typical profile of a deformation pulse in shown in Fig. 8. The duration of the recorded signals at half-height was $\tau_a \sim 2$ ns. The length of the leading edge of the pulse did not exceed 1 ns ($\tau_f \lesssim 1$ ns), and in the opinion of the authors,[51] was determined by the response time of the electrical system for taking the signal from the transducer. The use of piezoelectric detection also did not allow adequate reproduction of the profile of the shock waves (pressure $P_a \lesssim 3$ kbar) excited by radiation with $\lambda_1 \approx 0.53\,\mu$m, $\tau_L \approx 6$ ps, $I_L \lesssim 2 \times 10^{11}$ W/cm^2 in the semiconductor cyrstals GaAs and Si.

These circumstances motivated the active adoption of various optical methods of recording. It is true that some of these methods do not provide the necessary time resolution either. For example, Leuna *et al.*[54] employed laser interferometric detection of the displacement of a mirror surface as the shock wave arrived at the surface. Despite the use of ultrashort light pulses for generating the deformation waves ($\lambda_L \approx 1.054\,\mu$m, $\tau_L \approx 10$ ps), the duration of the leading edge of the recorded signals exceeded 1 ns: $\tau_f \sim 2$ ns (Fig. 9).

In the experiment of Ref. 54 the optical exposure was applied to an absorber (copper foil) pressed between two optical windows made of fused quartz. The broadening of the shock wave front may be partially due to reflections of the deformation waves in the thin layer of oil used to make acoustic contact between the back side of the absorber and the quartz window. The spatial restriction of the possibility of fast expansion of the plasma formed by ablation of the target (Cu) leads (as was pointed out earlier[51]) to a substantial increase in the efficiency of optoacoustic conversion. When the radiation is focused into a spot of diameter 0.5 mm, shock waves with pressures $P \sim 20$ kbar are excited when one uses generally available lasers with energy $\mathscr{E}_L \sim 20$ mJ per pulse ($I_L \lesssim 10^{12}$ W/cm^2). However, we note that in similar schemes of action on a target the slow expansion of the plasma leads to an increase in the time of its effective pressure on the target, and hence, to an increase in the shock pulse length (see Fig. 9). This situation is also indicated by the results of the experiment of Ref. 55, in which the shock waves were excited in an aluminium foil by radiation with about the same parameters as in Ref. 54. The rear side of the absorber (Al ~ 0.12 cm thick) was in contact with a layer of GaSe 20 μm thick. The entire system was clamped between optically transparent plates (Fig. 10). The second harmonic of the fundamental radiation ($\lambda_L \approx 0.527$ μm, $\tau_L = 6$ ns) was used to initiate luminescence on the surface of the GaSe[56] abutting the optical window. Luminescence was excited with a delay that could be adjusted with respect to the time of generation of the shock wave in the ablation of the aluminium to a time near when the shock wave arrived at the probed surface of the GaSe. The recording of the frequency shift of the luminescence line under the action of pressure enables one to reconstruct the time dependence of the characteristic pressure in the luminescing region of the GaSe. In Ref. 55 a slow decline in pressure was actually detected with a characteristic time of ~ 50 ns. The observed duration of the leading edge of the shock wave proved to be ~ 3 ns, which may be partially governed by the time of passage of the shock-wave front through the photoexcited region of the GaSe (the depth of the luminescence region was $\sim 5\,\mu$m [Ref. 55]. Subsequently, similar schemes in which picosecond laser pulses are used to initiate pres-

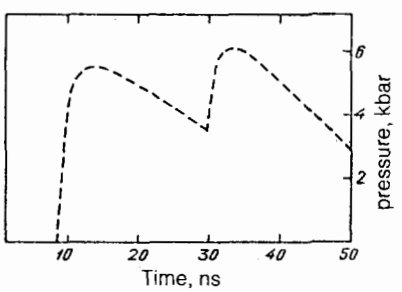

FIG. 9. Profiles of the pressure in a shock wave excited with a picosecond laser as the shock wave emerges at the rear surface of a 40-μm film.[54] The second peak in the profile is due to the reflected shock wave.

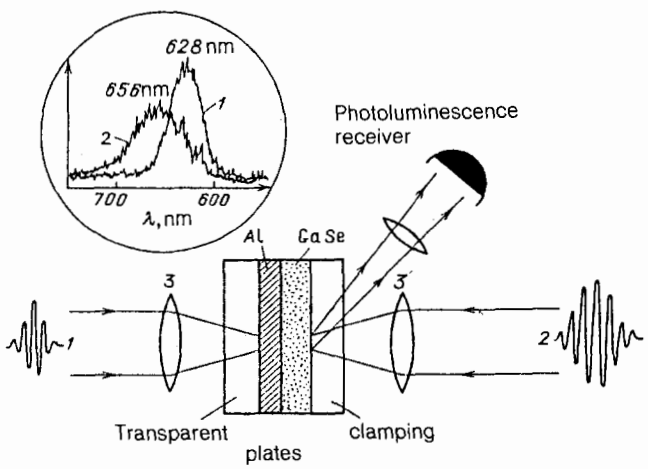

FIG. 10. Optoacoustic cell used in Refs. 55 and 58. *1*—radiation that excites the shock wave. *2*—radiation that initiates luminescence. *3*—lenses. The inset shows the spectra of spontaneous luminescence in without (*1*) and with (*2*) shock compresseion.

sures $P \sim 1$–10 kbar and the photoluminescence is detected have been widely applied for diagnostics of materials undergoing shock compression (solids,[57–59] solutions[60,61]).

The use of high-power subnanosecond laser pulses ($\mathscr{E}_L \sim 1$–100 J) has enabled the excitation of shock waves with pressures of several megabars ($P \sim 1$–10 Mbar) by the ablation of targets. The condensed matter is heated to a high temperature in the shock compression in such a wave. Correspondingly, the emergence of the shock wave to the surface initiates luminosity of the surface that can be detected.[62–64] In Ref. 62, laser pulses with parameters $\lambda_L = 1.06$ μm, $\tau_L \approx 300$ ps, and $\mathscr{E}_L \sim 20$–40 J were applied to an aluminum foil. The rear surface of the target was profiled into a stepped form. A streak camera (50-ps resolution) recorded the luminescence formed as the shock wave emerged onto the two displaced surfaces at the rear of the specimen. Measurement of the delay in the appearance of luminescence from these surfaces yields the velocity of the shock wave and the pressure in it. At intensities $I_L \sim 3 \times 10^{14}$ W/cm^2 pressures of $P \sim 2$ Mbar were attained. According to the results,[62] the width of the leading edge of the luminescence pulse does not exceed 50 ps. Thus the duration of the shock-wave front is shortened by the strong nonlinearity to less than the smallest durations of laser exposure. The experiment of Ref. 63 employed a laser apparatus with parameters similar to those of Ref. 62, but with a faster streak camera (with a time resolution ~ 10 ps). Here the recorded width of the shock-wave front was determined also by the time of passage of the front of the shock wave through the skin layer from which the luminescence was measured (~ 10 ps). In the experiments of Refs. 63 and 64 laser generation of shock waves with pressures $P \sim 20$–30 Mbar was realized.

The nonlinear elastic effects steepen the fronts of strong shock waves as they propagate. This enables one to obtain shock waves with subnanosecond rise times, even when one uses nanosecond laser exposures. Such an effect was observed experimentally in Refs. 67–69 ($\lambda_L \approx 0.53$ μm, $\tau_L \approx 2$ ns, $I_L \lesssim 5 \times 10^{13}$ W/cm^2). Figure 11 shows the time dependence of the temperature of the rear of the aluminum target as reconstructed from the results of the experiments.[68] The time to establish the maximum temperature was $\tau_f \sim 0.1$–0.3 ns. In the experiment of Ref. 50 a shock-wave front propagating in water was observed ($P \sim 40$ kbar) to become steeper to a rise time $\tau_f \sim 20$ ps. To initiate the shock wave, the absorber was exposed to radiation with $\lambda_L \approx 1.06$ μm, $\tau_L \approx 15$ ns, and $\mathscr{E}_L \approx 1$ J. The pressure and velocity of the shock wave were determined interferometrically by reflecting the probe radiation of a helium-neon laser from it. The experimental results agree with the hypothesis that the spatial extent of the front of the shock wave is negligible in comparison with the wavelength of the probe light ($\tau_f \lesssim 20$ ps).

2.2. Optical methods of detecting short acoustic pulses

Analysis of the experiments with strong shock waves certainly indicates the advantages of using optical methods for recording fast processes initiated by the deformation of crystals. At the same time, as is implied by the results of Refs. 31, 35, 36, 52, and 53, the use of piezoelectric transducers and visualization of the signals with oscilloscopes having a bandwidth under 1 GHz does not allow us to claim adequate reproduction of the shape of even subnanosecond sound pulses. Therefore, we can conclude that the future of detecting high-frequency broad-band acoustic signals (picosecond and *a-fortiori* subpicosecond deformation pulses) will involve only the optical methods.

At present the progress attained in the generation and detection of ultrashort acoustic pulses[28–30,32–34] is practically completely determined by the refinement of piscosecond and femtosecond laser technology. Primarily, this is a question of using picosecond and femtosecond lasers operating at a high repetition rate for optoacoustic excitation and acoustooptic detection of deformation waves, techniques that have been successfully applied to the study of various fast processes in solids.[70–74] The main advantage of such laser systems (Fig. 12) is that the use of an additional low-frequency modulation of the radiation exciting the acoustic pulses and tuned amplification in the processing the probe radiation enables one to increase substantially the sensitivity of reception of the useful signals. It is possible, to measure the relative variations of the reflection coefficient of the probe radiation (including changes caused by the nonsteady-state deformation of the specimen) at the level of 10^{-6}–10^{-7} (Ref. 72). The time resolution of such an optical recording system is determined by the length of the probe laser pulses. The correct reconstruction of the profile of the ultrashort acoustic pulse enables one to identify the mechanisms of generation of sound in the process of relaxation of

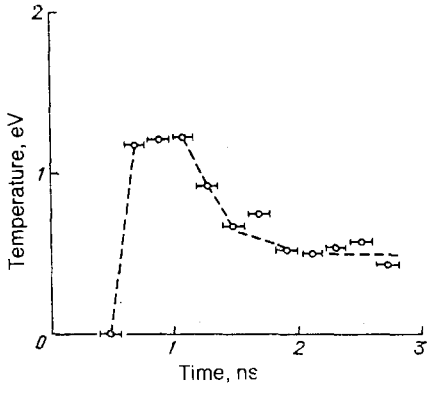

FIG. 11. Variation in time of the temperature of the rear surface of an aluminum film 38-μm thick after absorbing radiation of intensity $I_i \approx 5 \times 10^{13}$ W/cm^2 (Ref. 68). The peak intensity occurs at the zero instant of time.

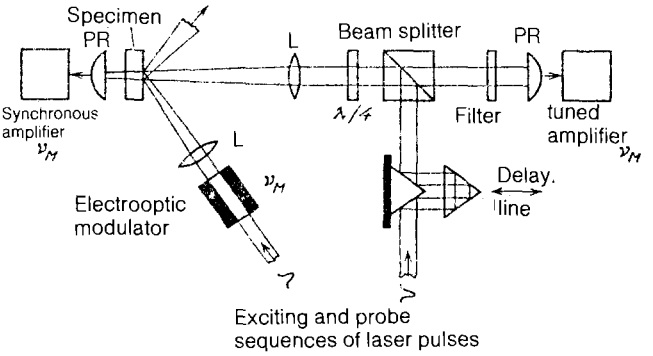

FIG. 12. Completely contact-free scheme for picosecond optoacoustic spectroscopy. PR—photoreceiver, L—lenses, ν_M—modulation frequency.

the energy of the absorbed exciting optical pulse.[33] At the present time ultrashort longitudinal acoustic pulses have been generated only by thermoelasticity[28–35,37] and the concentration-deformation mechanism[33] involving the change in density of the material induced by a change in the number of free charge carriers (in this case as a result of interband light absorption).

Coherent acoustic oscillations with a period shorter than 100 ps were recorded for the first time in the study by Thomsen *et al.*[32] of films of a-As_2Te_3. Amorphous films of As_2Te_3 of varying thicknesses ($a = 470, 900, 1200$ and 1600 Å) were deposited onto a sapphire substrate. To excite deformation waves, pulses of a dye laser with passive mode locking were used (photon energy $h\nu_L \approx 2.0$ eV, $\tau_L \approx 1$ ps, repetition rate of pulses $\nu_r \approx 0.5$ MHz, $\mathscr{E}_L \approx 1$ nJ). These pulses were focused onto the substrate-film boundary into a spot 50 μm in diameter. This same radiation, but substantially weakened in intensity, was used for probing the changes in time of the transmission and reflection coefficients of the a-As_2Te_3 film. A sequence of probe laser pulses was also focused onto the film from the substrate side with a variable delay time τ_d with respect to the probe pulses. Figure 13 shows the results[32] of the measurements of the transmission T of films of different thicknesses. The variation in transmission induced by absorption of the exciting laser pulses contains two components. The first of them (the discountinous decrease in transmission $\Delta T/T \sim 10^{-3}$ at the instant of photoexcitation and the subsequent monotonic increase in transmission) is due to photoexcitation of minority carriers and their relaxation. We note that the absorption of light in a-As_2Te_3 is interband absorption, since $h\nu_L$ exceeds the bandgap of the semiconductor ($E_g \approx 0.8$ eV). The second component, which corresponds to damped oscillations with an amplitude $\Delta T/T \sim 3\times 10^{-5}$–$3\times 10^{-4}$ involves the propagation of coherent acoustic waves in the film. The deformation of the semiconductor in the field of the acoustic wave causes a change in the band gap and in the light absorption coefficient α:

$$\Delta\alpha = (\partial\alpha/\partial E_g)(\partial E_g/\partial U)U. \qquad (2.1)$$

In Eq. (2.1) $U = \partial u/\partial z$ is the longitudinal deformation, u is the mechanical displacement in the acoustic wave, and z is the spatial coordinate along the normal to the surface of the film. It is assumed that the deformation does not cause a substantial change in the shape of the fundamental absorption edge of the semiconductor:

$$\partial\alpha/\partial E_g \approx -\partial\alpha/\partial(h\nu_L).$$

The constant $\partial E_g/\partial U$ is directly associated with the deformation potentials of the electrons (conduction band) and holes (valence band): $\partial E_g/\partial U = d = d^e + d^h$. If we assume that the changes in the absorption coefficient induced by the sound are small, we obtain the following expression for the relative changes in the transmission of the film:

$$\frac{\Delta T}{T} = \exp\left(-\int_0^a \Delta\alpha dz\right) - 1$$

$$\approx \frac{\partial\alpha}{\partial E_g} a(u(0) - u(a)) \approx \frac{1}{\hbar}\frac{\partial\alpha}{\partial\nu_L} d\,(u(a) - u(0)). \qquad (2.2)$$

In the case of strong interband absorption ($\alpha_L^{-1} \approx 300$ Å in As_2Te_3 [Ref. 32]) we can neglect interference effects. We assume that the changes in the refractive index in the region of localization of the acoustic wave do not substantially affect the transmission of the film.[32] Owing to the relationship (2.2), the change in transmission of the film is determined by the relative displacement of its surfaces.

According to the results of the experiments of Ref. 32, the thinnest films of a-As_2Te_3 exhibited acoustic vibrations at the frequency $\nu_a \sim 14$ GHz that corresponded to multiple reflections of the excited ultrashort acoustic pulses in the film. The authors of Ref. 32 also performed experiments with films of polyacetylene and a-SiO_2. In Ref. 30 a ring dye laser with passive mode locking and unloading of the resonator ($\lambda_L \approx 0.625$ μm, $\tau_L \approx 0.5$ ps, $\mathscr{E}_L \approx 2$ nJ, $\nu_r \approx 0.2$ MHz) was used to excite and record sound in submicron films of GaAs and InGaAsP, which were cemented with epoxy resin between the substrate and a glass cover. In subsequent studies[29,33] the laser exposure was performed on the free surface of films. The reflection of the probe radiation from this same surface was measured with a time delay. The sensitivity of the optical recording system was substantially enhanced. For this purpose an additional acoustooptic modulation of the exciting radiation from the ring dye laser was carried out with colliding pulses under conditions of passive mode locking ($h\nu_L \sim 2$ eV, $\tau_L \approx 0.2$ ps, $\mathscr{E}_L \approx 0.2$ nJ) at the frequency $\nu_M \approx 4$ MHz. The signal from the photodiode that received the reflected pulses of the sequence of probe pulses was applied to a phase-sensitive amplifier synchronized with the modulation frequency ν_M. Consequently it was possible to record the relative variations in the light reflection coefficient at the level of $\Delta R/R \sim 10^{-6}$–10^{-3}. In a film of a-As_2Te_3 of thickness $a = 2200$ Å echo signals were distinctly observed to occur when the exciting deformation pulse was positioned near the free (probed) surface of the film (Fig. 14). The first echo pulse is shown on a larger scale in Fig. 15. According to the theoretical views developed in Ref. 29, the profile of the signal $\Delta R(\tau_d)$ is determined by the changes in the absorption coefficient α_L and the refractive index n_L of the light throughout the volume of penetration of the probe radiation. The relative contribution of the variation of the refractive index Δn_L and the extinction coefficient depend

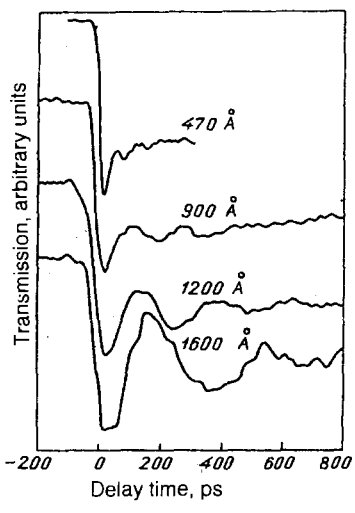

FIG. 13. Photoinduced changes in transmission of films of a-As_2Te_3 of various thicknesses at room temperature.[32]

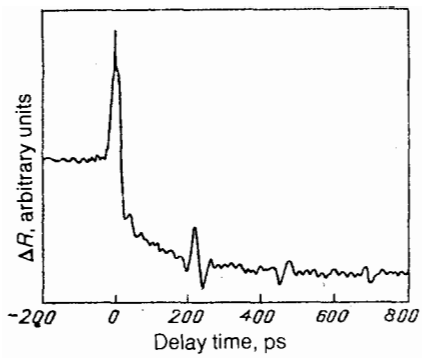

FIG. 14. Photoinduced changes in reflection from an As_2Te_3 film 2200-Å thick deposited on a sapphire substrate.[33]

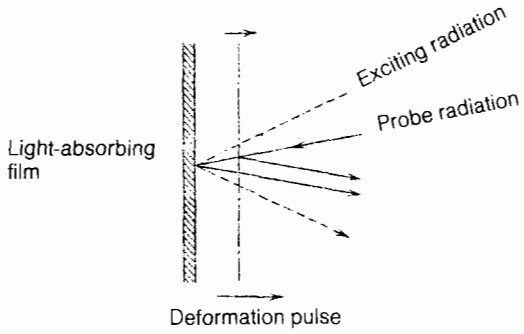

FIG. 16. Diagram of an experiment[34] on optical detection of harmonic hyperacoustic waves.

on the frequency of the probe light (in particular, on the closeness of $h\nu_L$ to the fundamental absorption edge of the semiconductor).

The analysis performed in Ref. 33 showed that the recorded tripolar signal (see Fig. 15) corresponds to reflection of the bipolar acoustic deformation pulses from the free surface of the film. The duration of each of the phases of the pulse is determined by the time of passage of the sound through the region of absorption of the exciting radiation $\tau_a \sim (\alpha_L c_a)^{-1} \sim 15$ ps ($c_a \sim 2\times 10^5$ cm/s is the velocity of sound in a-As_2Te_3). In Ref. 33 the optical generation and detection of sound were also obtained in a-Ge, a-As_2Se_3, and the metals Ni, Cu, and Al. In the work of Ref. 29 films of a-As_2Te_3 of thickness $a \lesssim 0.5\alpha_L^{-1}$ were used as generators and detectors of ultrashort acoustic pulses propagating in amorphous films of silicon oxynitride (SiON), which are optically transparent to radiation of wavelength $\lambda_L \approx 0.62$ μm ($h\nu_L \approx 2$ eV). In this experiment films with different relative contents of nitrogen [from $N/(N+O) = 0$ in SiO_2 to $N/(N+O) = 1$ in Si_3N_4] were deposited onto a sapphire substrate, and films of a-As_2Te_3 were deposited on top of them. The optical part of the experimental apparatus[29] was the same as that used in Ref. 33.

In the experiment of Ref. 28 the optical scheme shown in Fig. 12 was used to excite and detect the ultrashort acoustic pulses in films of the metals Ni, Zr, Ti, and Pt. Two dye lasers were pumped with a mode-locked argon laser. To generate sound, radiation was used with the wavelength $\lambda_L \approx 0.633$ μm, $\tau_L \approx 5$ ps, $\mathscr{E}_L \approx 0.3$ nJ, while the probe pulses had the parameters $\lambda_L \approx 0.595$ μm, $\tau_L \approx 5$ ps, and $\mathscr{E}_L < 75$ pJ. The repetition rate of the pulses was $\nu_r = 246$ MHz. In nickel films signals were detected with the relative amplitude $\Delta R/R \sim 1.6\times 10^{-6}$. The shape of the first echo pulse resembled that shown in Fig. 15, but differed in that the characteristic time scale was smaller by approximately half. The use of optical radiation with different frequencies to excite and to record the sound naturally allows one to increase the noise shielding of the detection circuit, and correspondingly, the sensitivity of detection of the acoustic signals. It is also important that it be possible to optimize independently the generation and detection of the acoustic pulses by tuning the frequencies of the dye lasers.

In Ref. 75, Wright *et al.* reported dividing the probe beam into two independent beams to attain a sensitivity $\Delta R/R \sim 10^{-7}$. One of the beams, after reflection from the unperturbed surface of the specimen, provided a reference signal. In this way the fluctuations of energy in the probe laser pulses were compensated.

Figure 16 shows a diagram of the experiment of Ref. 34, in which coherent acoustic waves at the frequency $\nu_a \approx 26$ GHz were detected optically. Broad-band acoustic pulses were excited by the absorption of a sequence of laser pump pulses ($h\nu_L \approx 2$ eV, $\tau_L \approx 0.2$ ps, $\mathscr{E}_L \approx 0.1$ nJ, $\nu_r \approx 110$ MHz) in films of a-Ge or Al and propagated in optical glass. The delayed sequence of probe pulses was directed at an angle to the normal. The magnitude of the reflected signal was determined by the interference of the part of it reflected from the surface of the film with the part reflected from the inhomogeneities of the refractive index n in the region of localization of the traveling deformation pulse. Figure 17 shows the experimental variation of the reflection coefficient of the probe pulses as a function of their delay time with respect to the incident pulses for an absorber made of amorphous germanium. The theoretical model proposed in Ref. 34 associates the damping of the oscillations ΔR (see Fig. 17) with the propagation and absorption of the component of the acoustic signal at the oscillation frequency. The frequency of the acous-

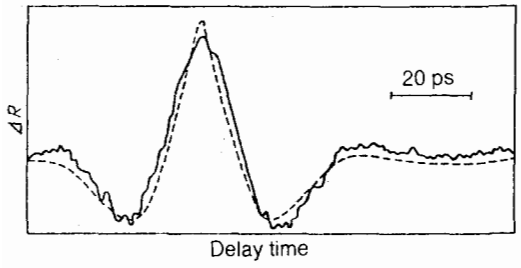

FIG. 15. First echo pulse in a-As_2Te_3. Solid line—experiment, dashed line—theory.[33]

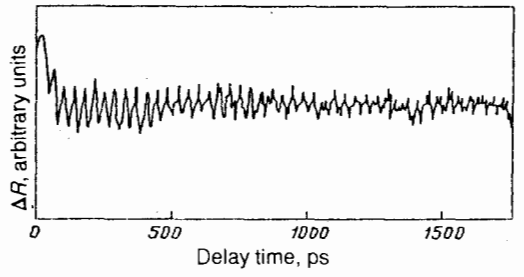

FIG. 17. Variation of the reflection of probe laser pulses as a function of their time delay with respect to the exciting pulses.[34]

tic component being probed depends on the angle of incidence θ of the probe pulses: $\nu_a = 2\nu_L n(c_a/c_L)\cos\theta$. By making measurements at different angles of incidence, in principle one can try to reconstruct the spectrum of the acoustic signal and thus obtain information on the shape of the profile of the deformation pulse and its duration. However, the existence in the spectrum of a separate high-frequency component ($\nu_a \sim 26$ GHz [Ref. 34]) still does not allow us to say that the excited acoustic pulses are ultrashort. The frequency of the acoustic waves accessible to recording by this method is bounded from above by the inequality $\nu_a \lesssim 2\nu_L n(c_a/c_L)$, as are the frequencies of the acoustic waves excited in stimulated Brillouin scattering.[76] For the conditions of the experiment of Ref. 34 we find $n \approx 1.5$, $c_a \approx 5.2\times 10^5$ cm/s, and hence $\nu_a \lesssim 26$ GHz. Thus, a substantial advance into the higher-frequency region in the optical scheme being discussed (see Fig. 16) requires lasers in the x-ray range.

To summarize the review of the experimental studies, we point out that the minimal duration of deformation pulses recorded up to now is $\tau_a \sim 5$–15 ps.[28,33]

3. THEORY OF OPTOACOUSTIC CONVERTERS. MECHANISMS OF GENERATION OF ULTRASHORT DEFORMATION PULSES

In the absorption of optical radiation in a target material, acoustic waves are excited by sources distributed through the volume of the medium. The wave equation that describes the generation of plane longitudinal acoustic waves can be written as[26,77]

$$\frac{\partial^2 u}{\partial t^2} - c_a^2\frac{\partial^2 u}{\partial z^2} = -\frac{1}{\rho}\frac{\partial G}{\partial z}. \tag{3.1}$$

Here u is the mechanical displacement in the direction of the z axis, c_a is the velocity of longitudinal sound in this direction, and ρ is the equilibrium density of the material. The function $G(z,t)$ has the physical meaning of the component σ_{zz} of the stress tensor σ_{ij}, from which the purely elastic part linear in the deformations has been subtracted. In other words, $\partial G/\partial z$ is the density of volume forces not directly involving the linear elasticity of the material. For simplicity and compactness of further analysis, we assume the possibility of propagation of a pure longitudinal wave along the z axis. The condition of absence of stresses at the mechanically free, uniformly irradiated surface $z = 0$ has the form

$$-c_a^2\frac{\partial u}{\partial z}\Big|_{z=0} = -\frac{1}{\rho}G. \tag{3.2}$$

If we employ a Laplace transform in the coordinate, by using a Fourier transform in time of all the functions

$$\tilde{F}(\omega, z) \equiv \int_{-\infty}^{\infty} F(t, z)\exp(i\omega t)\mathrm{d}t,$$
$$\hat{F}(\omega, p) = \int_0^{\infty} \tilde{F}(\omega, z)\exp(-pz)\mathrm{d}z, \tag{3.3}$$

and also the condition of radiation into the half-space $z > 0$, then we can describe the deformation $U = \partial u/\partial z$ that satisfies the problem (3.1) and (3.2) (outside the sound-generation region) by the following relation:[26,78]

$$U = \frac{1}{2\pi}\int_{-\infty}^{\infty} \tilde{U}(\omega)\exp(-i\omega\tau)\mathrm{d}\omega; \tag{3.4}$$

Here $\tau = t - (z/c_a)$ is the time in the accompanying system of coordinates, while we can represent the frequency spectrum of the deformation $\tilde{U}(\omega)$ in the plane acoustic wave propagating from the boundary in the form:

$$\tilde{U}(\omega) = -\frac{1}{2\rho c_a^3}(-i\omega)\left(\hat{G}\left(\omega, i\frac{\omega}{c_a}\right) - \hat{G}\left(\omega, -i\frac{\omega}{c_a}\right)\right) \tag{3.5}$$

We note that, when the boundary of the target is mechanically clamped ($u = 0$ for $z = 0$), then for describing the acoustic signal it suffices to change the sign inside the square [*sic*] brackets in (3.5).[77]

According to the solution of (3.4) and (3.5), the problem of determining the spectral composition of $\tilde{U}(\omega)$, and then the profile $U(\tau)$ of the deformation pulse, reduces to finding the space-time spectrum $\hat{G}(\omega,p)$ of the sources of the acoustic waves. Consequently one must specify the mechanism of excitation of the sound. To represent clearly the physical problems that stand in the way of reducing to the maximum extent the duration of acoustic pulses generated in laser action on matter, we shall systematically examine all the fundamental mechanisms of exciting them.

In this regard we note that the very widespread view that the characteristic duration of optoacoustic pulses is determined either by the time of laser action or by the time of passage of sound through the region of light absorption (as is implied by the treatment of thermoelastic sound generation under conditions of slow phonon heat conductivity in Sec. 3.1) is not correct in general. It is demonstrated in this section that the possibilities realized in optoacoustic conversion are indeed far more varied. As the duration of the laser exposure is shortened it is necessary to take into account the noninstantaneous processes of thermalization of the absorbed optical energy, especially in those cases in which photogeneration of non-equilibrium charge carriers occurs. Here the duration of the excited optoacoustic pulses can depend strongly on the crystal parameters that determine the space-time dynamics of the photogenerated electrons and holes.

A theoretical analysis enables one to discover the conditions under which the duration of the optoacoustic pulses equals the time of volume recombination of the electron-hole plasma, or the time of passage of sound through the region of recombination heating (Sec. 3.2). In the case of piezoelectric excitation of sound the duration of the optoacoustic pulses can equal the time of detachment of the diffusion wave of electrons, the time of surface recombination of electrons, or the time of passage of sound through the region of spatial separation of electrons and holes (Sec. 3.3). We note also that acoustic pulses can be shortened by the action of external fields that facilitate the confinement of photogenerated carriers near the surface. In particular, this can involve the drift of electrons in the electric field of the positively charged surface (in the region of surface bending of the bands of the semiconductor) (Sec. 3.3).

3.1. Thermoeleastic excitation of acoustic waves

When the generation of deformation waves is initiated by thermoleastic stresses (thermal expansion or compression of the material), the acoustic sources can be described

by[79,80]

$$G = G_T = K\beta T, \tag{3.6}$$

where K is the bulk modulus, β is the bulk expansion coefficient, and T is the temperature increase of the crystal. In the process of laser generation of sound the change in temperature of the material is associated with the thermalization of part of the absorbed optical energy.

A very simple description of the diffusion heat conductivity initiated by absorption of a laser pulse has the form

$$\frac{\partial T}{\partial t} = \chi\frac{\partial^2 T}{\partial z^2} + \frac{1}{\rho c}W,$$
$$\frac{\partial T}{\partial z}_{z=0} = 0, \quad T \underset{z\to\infty}{=} 0; \tag{3.7}$$

where χ is the thermal diffusivity, c is the specific heat, and $W(z,t)$ is the volume density of thermalized energy. With the use of the integral transformations (3.3), it suffices to represent the solution of the problem (3.7) in the form

$$\hat{T}(\omega, p) = \frac{1}{\rho c\chi(p^2 - p_\chi^2)}\left(\frac{p}{p_\chi}\hat{W}(\omega, p_\chi) - \hat{W}(\omega, p)\right), \tag{3.8}$$

where $p_\chi = \sqrt{-i\omega/\chi}$; from now on we shall use the symbol $\sqrt{\ }$ for the root with the positive real part.

To proceed further in the analysis we shall use again a very simple model to describe the spatial distribution of the intensity I of the laser radiation

$$I = (1 - R)I_i f(t)\exp(-\alpha z);$$

where α and R are the reflection and absorption coefficients of light, I_i is the intensity of radiation incident on the surface $z = 0$, and the function $f(t)$ describes the envelope of the laser pulse. Then, in the case when the absorbed optical energy is instantaneously thermalized, we have $W = \alpha I$, and

$$\hat{W}(\omega, p) = (1 - R)\, I_i\frac{\alpha}{\alpha + p}\tilde{f}(\omega). \tag{3.9}$$

By using Eqs. (3.8) and (3.9), we can represent the deformation spectrum (3.5) in the case of thermoelastic sound generation as

$$\tilde{U}_T(\omega) = -\frac{(1 - R)K\beta}{c\rho^2 c_a^3}\frac{(-i\omega)m_\chi\omega_\chi}{\omega_\chi + i\omega}$$
$$\times\left[\frac{\omega_\chi}{\omega^2 + m_\chi^2\omega_\chi^2} + \frac{1}{\sqrt{-i\omega}(m_\chi\omega_\chi^{1/2} + \sqrt{-i\omega})}\right]I_i\tilde{f}(\omega). \tag{3.10}$$

In writing Eq. (3.10) we have introduced the symbol $\omega_\chi = c_a^2/\chi$ for the frequency at which the moduli of the wave vectors of the acoustic and temperature waves become comparable. This characteristic frequency does not depend on the parameters of the optical exposure. In the system being studied there exist other "internal" characteristic frequencies: $\omega_\alpha = \alpha c_a$ is the reciprocal time of passage of sound through the region of light absorption, and the frequency $\omega_T = \alpha^2\chi$ is the reciprocal time of cooling of this region. Since the light absorption coefficient can depend strongly on its wavelength, the frequencies ω_T and ω_α can vary with the frequency of the laser radiation. An important factor is that they are associated with ω_χ by the single dimensionless parameter[77,81] $m_\chi = \alpha\chi/c_a\,(\omega_\alpha = m_\chi\omega_\chi, \omega_T = m_\chi^2\omega_\chi)$, which was used in the solution (3.10). The parameter m_χ is the ratio of the characteristic times of escape of sound and heat from the region of light absorption.

It is easy to see that, by virtue of Eq. (3.10), we can represent the spectrum $\tilde{U}(\omega)$ of the optoacoustic pulse as the product

$$\tilde{U}(\omega) = K^0K^1(\omega)I_i\tilde{f}(\omega) \tag{3.11}$$

of the spectrum $I_i\tilde{f}(\omega)$ of the time envelope of the intensity of the laser pulse multiplied by the function $K(\omega) \equiv K^0K^1(\omega)$, which is determined by the parameters of the medium (some of which, indeed, can depend on the photon energy $h\nu_L$). The coefficient K^0, which does not depend on the frequency ω, can always be chosen in such a way that the function $K(\omega)$ is dimensionless and normalized to a value of the order of unity ($\max|K^1(\omega)| \lesssim 1$). It is this function $K^1(\omega)$ that determines the relative efficiency of transformation of the various frequencies of the spectrum $I_i\tilde{f}(\omega)$ into acoustic frequencies. In this review we shall use the term "spectral transfer function of optoacoustic conversion,"[77,82,83] both for $K(\omega)$ and for $K^1(\omega)$.

In accordance with expression (3.11), in order that the spectrum of the optoacoustic signal $\tilde{U}(\omega)$ contain high-frequency components (and hence, the acoustic pulse will be short), it is necessary at least that frequencies equally high should exist in the spectrum of the intensity of the laser pulse $I_i\tilde{f}(\omega)$. Actually this means that to generate deformation pulses with a characteristic pulse length τ_a, one must always use laser pulses of length $\tau_L \lesssim \tau_a$. However, this is not sufficient. It is also necessary that the high-frequency components of the envelope of the light intensity be efficiently transformed by the spectral transfer function $K(\omega)$ into acoustic components.

If we assume for definiteness that the laser pulse has a Gaussian form with the characteristic pulse length τ_L ($f(t) \sim \exp[-(2t/\tau_L)^2]$), then the spectrum of the envelope $\tilde{f}(\omega) \sim \exp[-(\tau_L\omega/4)^2]$ will contain components up to the frequency $\omega_L \equiv 4\tau_L^{-1}$. Consequently, to generate acoustic pulses of length $\tau_a \sim \tau_L$, the function $K(\omega)$ must efficiently "transfer" frequencies of the order of $\omega_L = 4\tau_L^{-1}$.

According to Eq. (3.11) the characteristic shape of the spectral transfer function of thermoelastic optoacoustic conversion depends on the single parameter m_χ. Its shape can be substantially simplified in two limiting physical situations ($m_\chi \ll 1$ and $m_\chi \gg 1$).

If the region of light absorption is thermally thick at the frequency ω:

$$\omega \gg \omega_T = m_\chi^2\omega_\chi, \tag{3.12}$$

then we have

$$K_T^1(\omega) \approx \frac{(-i\omega)m_\chi\omega_\chi}{\omega^2 + m_\chi^2\omega_\chi^2} = \frac{(-i\omega)\omega_\alpha}{\omega^2 + \omega_\alpha^2}. \tag{3.13}$$

In accordance with Eq. (3.13), the most efficient optoacoustic conversion is obtained at the frequency $\omega = \omega_\alpha$ (in the frequency band $e - \sqrt{e^2 - 1})\omega_\alpha \leqslant \omega \leqslant (e + \sqrt{e^2 - 1})\omega_\alpha$ at the $1/e$ level). Consequently, Eq. (3.13) correctly de-

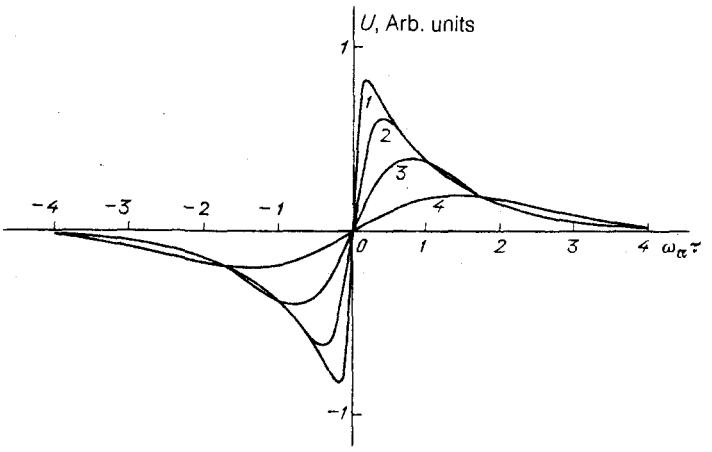

FIG. 18. Profiles of optoacoustic pulses excited thermoelastically near the free boundary for different values of the parameter $\omega_\alpha/\omega_L = 0.2$ (*1*), 0.6 (*2*), 1.4 (*3*), and 3 (*4*).

scribes the transfer function if ω_α satisfies inequality (3.12), i.e., $m_\chi \ll 1$. In the regime being discussed, laser generation of deformation pulses of length τ_a requires that the function $K_T^1(\omega)$ does not cut off the high-frequency components. To ensure this, we must include the condition $\omega_\alpha \gtrsim \omega_a$ by increasing α and c_a.

The profile of the acoustic pulse for $m_\chi \ll 1$ is determined by using Eqs. (3.13) and (3.11) and the inverse Fourier transformation (3.4). It has a characteristic symmetric bipolar form (Fig. 18). The shape of the optoacoustic signals is described particularly simply in the case $\omega_L \gg \omega_\alpha$:

$$U_T(\tau) = -\frac{K_T^0}{2} I_i(\omega_\alpha \tau_L) \frac{\tau}{|\tau|} \exp(-\omega_\alpha|\tau|). \tag{3.14}$$

The characteristic duration of each phase of the pulse Eq. (3.14) equals the time of passage of sound through the region of light absorption $\tau_a = \omega_\alpha^{-1}$. We note that the condition $\omega_L \gg \omega_\alpha$ is almost always realized when one uses subpicosecond light pulses, since when $\tau_L \lesssim 1$ ps we estimate correctly that $\omega_L \gtrsim 4\times 10^{12}$ s^{-1}, while with the typical restrictions on the quantities $\alpha \lesssim 10^6$ cm^{-1} and $c_a \lesssim 5\times 10^5$ cm·s^{-1} we find that $\omega_\alpha \lesssim 5\times 10^{11}$ s^{-1}.[30,32,33] The profiles of acoustic pulses of the form (3.14) were reconstructed by analyzing the experimental results.[33] The profiles of pulses for an arbitrary ratio between ω_α and ω_L in the regime $m_\chi \ll 1$ were calculated in Refs. 84–86. Figure 18 illustrates the character of the transformation of the profiles of the deformation pulses with increase in the parameter ω_α/ω_L for a laser pulse of Gaussian form (and $\beta > 0$). As the parameter ω_α/ω_L increases, the relative duration of the transition region from the compression phase to the rarefaction phase ($\sim\tau_L$) increases. In the limiting case $\omega_\alpha/\omega_L \gg 1$ the profile of the optoacoustic pulse, according to Eqs. (3.11) and (3.13), is determined by the derivative of the profile of the laser pulse

$$U_T(\tau) = K_T^0 \frac{I_i}{\omega_\alpha} \frac{\partial}{\partial\tau} f(\tau),$$

while its length is determined by the duration of the optical exposure.

If the absorption region is thermally thin at the frequency ω:

$$\omega \ll \omega_T = m_\chi^2 \omega_\chi, \tag{3.15}$$

then we have

$$K_T^1(\omega) = \frac{\sqrt{-i\omega}\,\omega_\chi^{1/2}}{\omega_\chi + i\omega} \tag{3.16}$$

and the maximum of the transfer function is obtained at $\omega = \omega_\chi$ ($|K_T^1| \sim \omega^{1/2}$ for $\omega \ll \omega_\chi$ and $|K_T^1| \sim \omega^{-1/2}$ for $\omega \gg \omega_\chi$). Therefore, in view of Eq. (3.15), the description (3.16) is correct when $m_\chi \gg 1$, i.e., when heat propagates to distances of the order of α^{-1} faster than longitudinal sound. The electronic heat conduction can be supersonic in metals. In this case, to shorten thermoelastic optoacoustic pulses, one must simultaneously increase ω_L and $\omega_\chi = c_a^2/\chi$. The profile of the deformation pulse described by the relations (3.4), (3.11), and (3.16) depends strongly on the ratio between ω_L and ω_χ, i.e., on whether heat conduction is subsonic or supersonic at the frequencies ω_L. If the heat conductivity at these frequencies is slow ($\omega_\chi/\omega_L \gg 1$), then in calculating the profile of the optoacoustic pulse one can use the approximation $K_T^1 \approx \sqrt{-i\omega/\omega_\chi}$. The profile of the acoustic pulse proves to be bipolar (Fig. 19, curve *1*). The duration of the first phase of the acoustic pulse does not exceed the duration of the laser exposure ($\tau_a \lesssim \tau_L$). However, if the heat conduction is fast ($\omega_\chi/\omega_L \ll 1$), then instead of (3.16) we can use $K_T^1 \approx -\sqrt{\omega_\chi/(-i\omega)}$, and the deformation pulse proves to be unipolar (Fig. 19, curve *3*). Figure 19 shows the results of the analytical calculations of the pro-

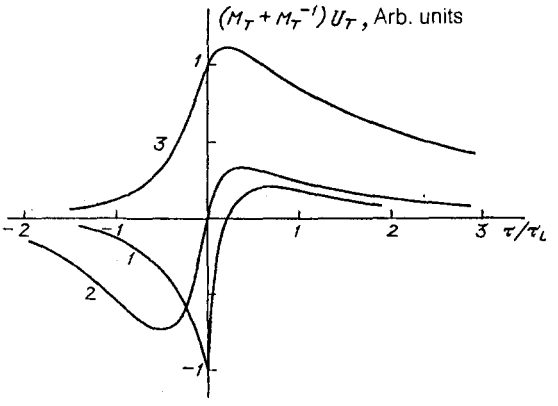

FIG. 19. Profiles of optoacoustic pulses excited thermoelastically for different values of the parameter $M_T = (\omega_\chi/\tau_L)^{-1/2} \ll 1$ (*1*), $= 1$ (*2*), and $\gg 1$ (*3*).

files of optoacoustic pulses for a laser exposure modulated in the following way $f(t) = \exp(-2|t|/\tau_L)$.[77,87]

The deformations are normalized to the quantity $(-K_T^0 I_i) = (1-R)K\beta I_i/c\rho^2 c_a^3$. Analysis[87] shows that the transformation of the profile of the deformation pulse as the parameter $M_T = (\omega_\chi \tau_L)^{-1/2}$ increases occurs via the predominant decline of the efficiency of excitation of the first phase of the acoustic signal when $M_T > 1$. In its physical meaning M_T is the Mach number of a temperature wave having the characteristic frequency τ_L^{-1}. For supersonic velocities of expansion of the heated region ($M_T > 1$) the efficiency of excitation of acoustic waves propagating into the depth of the crystal immediately after generation sharply declines ($\sim M_T^{-2}$ for $M_T \gg 1$). These waves form the leading edge of the deformation pulse. When $M_T \gg 1$ the shape of the signal ceases to depend on M_T. However, its pulse length exceeds severalfold (a factor of 4 to 5) the duration of the laser pulse (Fig. 19, curve *3*). One can show that the profile of the optoacoustic pulse in this limit resembles the time dependence of the surface temperature ($z = 0$). Actually the fast heat conduction smooths out the spatial inhomogeneities of the temperature so quickly that the generation of sound in the bulk of the crystal becomes inefficient.

If pure phonon heat conduction occurs in the material, then one cannot use Eq. (3.7) at frequencies $\omega \gtrsim \omega_\chi$, since it does not describe the existence of a limiting velocity of phonon heat conduction c_T. In any case c_T does not exceed the velocity of the fastest (longitudinal) acoustic waves: $c_T \lesssim c_a$. In this case on ordinarily uses[88,89] the generalized heat conduction equation[90]

$$\frac{\partial T}{\partial t} + \frac{\chi}{c_T^2}\frac{\partial^2 T}{\partial t^2} = \chi\frac{\partial^2 T}{\partial z^2} + \frac{1}{\rho c}W. \tag{3.17}$$

The solution of Eq. (3.17) with the boundary conditions from Eq. (3.7) is described as before by Eq. (3.8). However, the number p_χ, which is proportional to the wave number of the temperature wave, is modified: $p_\chi = \sqrt{(-i\omega/\chi) - (\omega^2/c_T^2)}$. The role of this refinement is seen most clearly in analyzing the thermoelastic generation of sound near a rigid boundary ($u(z=0) = 0$) in the limiting case of surface absorption of light ($\alpha \to \infty$). In this case the spectral transfer function takes the form

$$K_T^1 \approx \frac{\omega_\chi}{\omega_\chi + i\Delta\omega}; \tag{3.18}$$

Here, as before, we have used the notation $\omega_\chi = c_a^2$, while $\Delta = 1 - (c_a^2/c_T^2)$ is the relative mismatch of the velocities of the temperature and sound waves. However, the characteristic frequency ω_χ has not kept its previous physical meaning, since when $c_T \lesssim c_a$ the wave numbers of the temperature and sound waves can be comparable only when $\omega \to \infty$ (and only in the case $c_T = c_a$). In view of Eq. (3.18), when the ratio between c_T and c_a changes, the location of the pole of the transfer function in the complex plane of ω changes. This leads to substantial transformation in the profiles of the acoustic pulses. Let the laser exposure be sufficiently short

$$\omega_L \gg \omega_\chi/|\Delta|.$$

Then, if the limiting velocity of propagation of heat is supersonic (e.g., in the case of Eq. (3.7) $c_T = \infty$), the profile of the optoacoustic pulse is described as follows:

$$U_T(\tau) = \frac{1}{2}K_T^0 I_i \frac{\omega_\chi}{\Delta}\left(1 - \frac{\tau}{|\tau|}\right)\exp\frac{\omega_\chi}{\Delta}\tau.$$

The characteristic pulse length $\tau_a = \Delta/\omega_\chi$ is determined by the fast thermal expansion of the region of localization of the thermoleastic sources in the case $\Delta > 0$.

However, if the limiting velocity of propagation of heat is subsonic (e.g., to allow for the possible existence of second sound,[90] one assumes that $c_T = c_a/\sqrt{3} < c_a$), then the profile of the optoacoustic signal is described by

$$U_T(\tau) = \frac{1}{2}K_T^0 I_i \frac{\omega_\chi \tau_L}{|\Delta|}\left(1 + \frac{\tau}{|\tau|}\right)\exp\frac{\omega_\chi}{\Delta}\tau.$$

The acoustic pulse length is determined now by the slow cooling of the subsurface region in the case $\Delta < 0$. Here the thermoelastic sources rapidly (or instantaneously) are turned on by the laser effect, and disappear only after a finite time.

At low (liqiud-helium) temperatures ballistic phonon heat condution[91,92] can occur, even in crystals of macroscopic dimensions. In this process the energy of thermal lattice vibrations is transported by acoustic phonons propagating without collisions. Upon pulsed heating of the surface of the crystal, the front of the flux of non-equilibrium, longitudinally polarized phonons propagates into the interior of the specimen with the velocity c_a. However, to analyze the thermoelastic generation of sound in non-steady-state ballistic phonon heat conduction, the description discussed above Eqs. (3.4), (3.6), and (3.17) must be substantially altered. Instead of the heat-conduction Eq. (3.17), we must use the kinetic equations for the distribution function of phonons of different polarizations,[77,93] since the concept of temperature in the traditional sense for non-equilibrium phonon distributions cannot be introduced. In this situation it is especially useful to treat the excitation of regular deformation waves as resulting from interaction of random (thermal) waves through the quadratic nonlinearity of the elastic medium.[38,77] The sources of the deformation waves prove to depend directly on the distribution function of the non-equilibrium phonons. For example, if we take account only of phonons with longitudinal polarization, then instead of Eq. (3.6) we should use[38,77,94]

$$G_T = (2\pi)^{-3}\int \hbar c_a k(\varepsilon_2 + \varepsilon_1\xi^2)N(\mathbf{k}, t)\mathrm{d}^3\mathbf{k}, \tag{3.19}$$

Here $\mathbf{k}$ is the wave vector of the phonon, $k \equiv |\mathbf{k}|$, ξ is the cosine of the angle between $\mathbf{k}$ and the z axis, N is the distribution function, and the dimensionless parameters ε_1 and ε_2 are related to the second- and third-order elastic moduli.

The most interesting feature of ballistic phonon heat conduction is the following: outside the region of laser generation of phonons, after a time greater than their time of generation and the time of passage of sound through the region of phonon generation, the synchronous component predominates in the sources of coherent deformation waves of Eq. (3.19). We bear in mind that under these conditions the right-hand side of the wave equation for the deformation in the coordinate system moving with the velocity of sound has a constant form. Indeed, in the plane of the geometry of the problem the amplitude of the sources falls off with distance from the irradiated surface ($\sim z^{-1}\partial\Phi/\partial\tau$).

Thus, there are sources propagating exactly with the

velocity of sound (synchronous). The excited deformation waves do not break away from the leading edge of the flux of non-equilibrium phonons. Therefore the solution Eq. (3.4), which is valid outside the region of localization of the sources, cannot be used in this case. However, the solution Eqs. (3.4), (3.5) can be useful also in this case. If we modify the sources $G \to G[1 - \theta(z - z_0)]$, then the expression Eqs. (3.4) and (3.5) will describe all the waves excited in the region $0 \leqslant z \leqslant z_0$. In this approach the synchronously excited component of the deformation wave at the distance z_0 is described exactly. For this same reason, apparently, it is not correct to base on Eqs. (3.4) and (3.5) the study of the thermoelastic generation of sound in the case in which $c_T = c_a$ in the model (3.17).

Analysis of the exact solution of the wave equation[38,77,94] shows that, at distances from the boundary of the crystal substantially exceeding the spatial dimension of the excited deformation pulse, the fundamental contribution to the optoacoustic signal ($\sim \ln z \cdot \Phi(\tau)$) comes from the synchronous sources. The amplitude of the deformation wave localized at the front of the flux of ballistic phonons slowly (logarithmically) increases with distance from the boundary. The duration of the excited optoacoustic pulse is determined either by the time of passage of sound through the region of phonon generation or by their time of generation (actually the larger of these times).[77] Ballistic phonon heat conduction does not broaden optoacoustic pulses. It is also important that the generation of longitudinal sound at the front of the heat pulse of transverse phonons, which propagate at the velocity $\sim c_a/2$, is not synchronous. Therefore, at large distances we can neglect the contribution to the coherent acoustic wave from the transverse phonons.

We note that the synchronous character of the interaction of heat pulses and deformation waves can require that we take into account the self-action of coherent sound and its reverse influence on the propagation of phonons. A very simple model of describing these processes is discussed in Ref. 95.

3.2. The concentration-deformation mechanism of exciting sound

The generation of sound in crystals can arise from modulation of the concentration of free charge carriers.[76] Deep modulation of the carrier concentration can be attained by using interband light absorption, since in this case the simultaneous creation of an electron and a hole at a given point in space does not violate the electrical neutrality of the medium. Consequently, the Coulomb interaction does not prevent the existence of such spatially inhomogeneous carrier ditributions ($n^e = n^h \equiv n$ is the concentration of carriers; hereinafter the index $m = e, h$ is used for quantities pertaining respectively to electrons and holes).

If free carriers with a finite lifetime are created by absorption of photon (and hence, the thermalization of all the absorbed light energy is not instantaneous), then, along with the thermoelastic mechanism, the deformation[96] (electronic,[33,97] concentration-deformation[38,85]) mechanism of sound excitation is involved:

$$G = G_T + G_n = K\beta T - dn. \tag{3.20}$$

In Eq. (3.20), as before, the symbol d is used for the overall deformation-potential constant of the electrons and holes ($d = d^e + d^h$). Here, for simplicity in the model of an isotropic solid, we neglect the tensor character of the deformation potential, $d_{ij} \to d\delta_{ij}$, which is valid for spherical energy surfaces of the charge carriers.[76,98]

A very simple model describing the temperature and concentration fields of charge carriers generated by the interband absorption of light is the following:[39,78,99]

$$\frac{\partial n}{\partial t} = D\frac{\partial^2 n}{\partial z^2} - \frac{n}{\tau_R} + \frac{W}{h\nu_L}; \quad \frac{\partial n}{\partial z}\underset{z=0}{=} 0, \quad n \underset{z\to\infty}{=} 0, \tag{3.21}$$

$$\frac{\partial T}{\partial t} = \chi\frac{\partial^2 T}{\partial z^2} + \left(1 - \frac{E_g}{h\nu_L}\right)\frac{W}{\rho c} + \frac{E_g}{\rho c}\frac{n}{\tau_R}. \tag{3.22}$$

Here D is the coefficient of ambipolar diffusion of the quasineutral electron-hole plasma, and τ_R is the time of nonradiative recombination of the electrons and holes. The fundamental difference between Eq. (3.22) and Eq. (3.7) is in the assumption that only the fraction $(h\nu_L - E_g)/h\nu_L$ of the absorbed optical energy is thermalized instantaneously with the relaxation of electrons to the bottom of the conduction band and of holes to the top of the valence band. The remaining fraction of the energy is transformed into thermal motion of atoms only with the recombination of electron-hole pairs (the last term on the right-hand side of Eq. (3.22)). Consequently the space-time characteristics of thermoelastic sound sources can depend substantially on the dynamics of the electron-hole plasma. The interesting limiting case in which the duration of a thermoelastically excited deformation pulse is determined by the time of passage of sound through the region of recombination heating ($\tau_a \sim \sqrt{D\tau_R}/c_a$) has been analyzed in Ref. 100. Actually the distance $\sqrt{D\tau_R}$ that the non-equilibrium carriers diffuse in their lifetime can play the role of α^{-1} under certain conditions[77] in the spectral transfer function of the form of Eq. (3.13).

The most important assumptions used in the linearized model of Eqs. (3.21) and (3.22) are the neglect of surface recombination of electron-hole pairs[78] and of radiative bulk recombination.[99] Nevertheless it proves highly useful for a qualitative explanation of a number of optoacoustic experiments.[85,101]

Equation (3.21), together with Eqs. (3.20), (3.4), and (3.5), enables one to study the generation of ultrashort optoacoustic pulses via the concentration-deformation mechanism. Importantly, analysis and estimates for typical semiconductors[39,77,99] indicate the predominance of the electronic over the thermoelastic mechanism in the frequency region $\omega \geqslant \omega_R \equiv \tau_R^{-1}$. Consequently the role of the electronic mechanism becomes ever more important as we advance toward laser generation of ever shorter acoustic pulses.

The integral transforms of Eq. (3.3) enable one to write the solution of the problem (3.21) in the form

$$\hat{n}(\omega, p) = \frac{1}{h\nu_L D(p^2 - p_D^2)}\left(\frac{p}{p_D}\hat{W}(\omega, p_D) - \hat{W}(\omega, p)\right), \tag{3.23}$$

where $p_D \sqrt{(\omega_R - i\omega)/D}$. Further, in substituting G_n of Eq. (3.20) into Eq. (3.5), we obtain by using Eq. (3.23)

$$\tilde{U}_n = \frac{(1-R)d}{\hbar\omega_L \rho c_a^3} \frac{(-i\omega)^2 m_D \omega_D}{\omega_D(\omega_R - i\omega) + \omega^2} \times \left[\frac{\omega_D}{\omega^2 + m_D^2\omega_D^2} + \frac{1}{\sqrt{\omega_R - i\omega}(m_D\omega_D^{1/2} + \sqrt{\omega_R - i\omega})} \right] I_i \tilde{f}(\omega). \quad (3.24)$$

Here we have introduced the characteristic frequency $\omega_D = c_a^2/D$ at which the wave numbers of the acoustic and concentration waves become equal in absence of recombination of carriers ($\tau_R = \infty$). The dimensionless parameter $m_D = \alpha D/c_a$ equals the ratio of the times of escape of sound and of freely ($\tau_R = \infty$) diffusing carriers from the region of light absorption. The qualitative form of the spectral dependence of Eq. (3.24) does not coincide with (3.10) with the replacement $D \to \chi$ only because of the presence of the additional characteristic frequency $\omega_R \neq 0$ that arises because of the finite lifetime on non-equilibrium electron-hole pairs. An important fact is that recombintion also restricts the spatial motion of the carriers.

If the region of light absorption is diffusionally thick at the frequency ω:

$$|\omega_R - i\omega| \gg m_D^2\omega_D \equiv \omega_n = \alpha^2 D, \quad (3.25)$$

then Eq. (3.24) determines the following form of the spectral transfer function of concentration-deformation optoacoustic conversion:[42,77]

$$K_n^1(\omega) = \frac{(-i\omega)^2 m_D \omega_D}{(\omega_R - i\omega)(\omega^2 + m_D^2\omega_D^2)} = \frac{(-i\omega)^2\omega_\alpha}{(\omega_R - i\omega)(\omega^2 + \omega_\alpha^2)}. \quad (3.26)$$

When $\omega_R < \omega_\alpha = \alpha c_a$, the situation differs only insignificantly from Eq. (3.13) in the sense that $|K_n^1(\omega)|$ (Eq. 3.26) just like $|K_T^1(\omega)|$ in (3.13), has a maximum at the frequency $\omega \sim \omega_\alpha$. Correspondingly, in view of Eq. (3.25) the description Eq. (3.26) in this case is valid when $m_D \ll 1$. To shorten the optoacoustic pulses in this regime, we must first of all increase ω_α along with increasing ω_L.

However, in the electronic mechanism of the optoacoustic effect, another possibility exsits of shortening τ_a. For $\omega_R > \omega_\alpha$, the falloff in the efficiency of optoacoustic conversion begins, according to Eq. (3.26), only when $\omega > \omega_R > \omega_\alpha$ ($|K_n^1| \sim \omega^2$ when $\omega \ll \omega_\alpha$, $|K_n^1| \sim$ const when $\omega_\alpha \lesssim \omega \lesssim \omega_R$, and $|K_n^1| \sim \omega^{-1}$ only when $\omega \gg \omega_R$). Consequently, in the electronic mechanism, one can use relatively deeply penetrating radiation ($\omega_\alpha \ll \omega_a$) for generating short deformation pulses with a duration τ_a, if the recombination of carriers is fast ($\omega_R \sim \omega_a$).[39] In this case one uses the possibility of fast recombination-induced quenching of the sources of the acoustic waves. Sufficiently short recombination times of photogenerated carriers can be obtained in heavily doped and amorphous semiconductors.[102] We note that, in view of Eq. (3.25) when $\omega_R > \omega_\alpha$, the description (3.26) is valid if $\omega_R \gg \alpha^2 D$, i.e., the diffusion length of the carriers in their lifetime $\sqrt{(D\omega_R^{-1})}$ is substantially smaller than the light-absorption length α^{-1}.

Analytic calculations of the profiles of deformation pulses with an arbitrary ratio between ω_L and the characteristic frequencies ω_α and ω_R in the spectral transfer function (3.26) were performed in Refs. 85 and 86. Here we shall present only a heuristic description of the profile of the optoacoustic pulse in the limiting case $\omega_\alpha \ll \omega_R \ll \omega_L$:

$$U_n(\tau) = \frac{K_n^0}{2} I_i \omega_\alpha \tau_L \left(1 + \frac{\tau}{|\tau|}\right) \exp(-\omega_R \tau). \quad (3.27)$$

Consequently, under these conditions the acoustic pulse length equals the time of recombination of electron-hole pairs ($\tau_a = \tau_R$).

If the region of light absorption is diffusionally thin at the frequency ω:

$$|\omega_R - i\omega| \ll m_D^2\omega_D \equiv \omega_n = \alpha^2 D, \quad (3.28)$$

then we have

$$K_n^1(\omega) \approx \frac{(-i\omega)^2\omega_D^{1/2}}{\sqrt{\omega_R - i\omega}\,[\omega_D(\omega_R - i\omega) + \omega^2]}. \quad (3.29)$$

For $\omega_R < \omega_D$, the spectral transfer function (3.29) actually resembles (3.16): the maximum efficiency of conversion is realized at frequencies $\omega \sim \omega_D$ when the velocity of sound coincides with that of the concentration wave of the nonrecombining ($\omega_R < \omega_D \sim \omega$) carriers. The description (3.29) in this case is valid if $m_D \gg 1$. To shorten optoacoustic pulses in such a regime one must increase ω_D (decrease the carrier mobility). In typical semiconductors, even at room temperature, $D \gtrsim 10$ cm^2/s, and hence, $\omega_D \lesssim 2.5 \times 10^{10}$ s^{-1}. Apparently one can substantially weaken this restriction only in amorphous semiconductors.

However, one can reduce the diffusion length not only by decreasing the mobility of the carriers, but also by decreasing their lifetime. For $\omega_R > \omega_D$ the decline in efficiency of optoacoustic conversion begins only for $\omega > \omega_R$. However, the description (3.29) in this situation is correct under the condition $\omega_R \ll \omega_n = \alpha^2 D$, which is more stringent than $m_D \gg 1$.

Thus, also in this case one can reduce the recombination time to shorten optoacoustic pulses. However, we stress that increasing $\omega_R = \tau_R^{-1}$, according to (3.26) and (3.29), leads to decreased efficiency of optoacoustic conversion (predominantly at frequencies $\omega < \omega_R$). In other words, the deformation pulses are shortened not by increasing the efficiency of conversion of high frequencies, but by suppressing the low frequencies.

Surface recombination of photogenerated carriers can play an analogous role in the optoacoustic excitation of ultrashort acoustic pulses via the electronic mechanism. For the sake of clear description of the influence of surface recombination, we shall neglect in Eq. (3.21) the bulk recombination, assuming that ω_R is smaller than the other characteristic frequencies in the problem. At the same time, in Eq. (3.21) we shall define the flux of carriers at the boundary $z = 0$ by the condition $D\partial n/\partial z = Sn$, where S is the surface recombination velocity. Then in the case of light absorption at the surface ($\alpha \to \infty$), the spectral transfer function can be represented in the form[26,87]

$$K_n^1(\omega) = \frac{(-i\omega)\omega_D^{1/2}}{(\omega_D + i\omega)(\omega_S^{1/2} + \sqrt{-i\omega})}, \quad (3.30)$$

where we have introduced the notation $\omega_S = S^2/D$ for the frequency at which the velocity of the concentration wave equals the surface recombination velocity. When $\omega_S < \omega_D$

the maximum efficiency of optoacoustic conversion is obtained at the frequency ω_D. However, in view of Eq. (3.30), when $\omega_S > \omega_D$, the falloff in efficiency of optoacoustic conversion begins only at $\omega > \omega_S$. The condition $\omega_S > \omega_D$ is equally valid with $S > c_a$. Consequently, supersonic velocities of surface recombination facilitate the excitation of short deformation pulses. We note that very high surface recombination velocities ($S \sim 10^6$–3×10^7 cm/s) have been observed in many experiments (mainly optical).[103–105]

In the most interesting case $\omega_D \ll \omega_L$ where in absence of surface recombination ($\omega_S = 0$) the optoacoustic pulse expands because of supersonic carrier diffusion, we can simplify $K_n^1(\omega)$ of Eq. (3.30) in calculating the profile of the deformation pulse:

$$K_n^1(\omega) \approx -\frac{\omega_D^{1/2}}{\omega_S^{1/2} + \sqrt{-i\omega}}.$$

This representation of the spectral transfer function, together with (3.11), explicitly demonstrates that when $\omega_S \gg \omega_L$ the profile of a deformation pulse excited near the free surface must resemble the profile of the laser pulse. Figure 20 shows how the acoustic pulse becomes shorter as the Mach number M_S of the motion of carriers toward the surface increases ($M_S = S/c_a$). The calculations were performed for the model $f(t) = \exp(-2|t|/\tau_L)$.[87] In Fig. 20 also the Mach number M_D of the concentration wave at the frequency τ_L^{-1} has been introduced [$M_D = (\omega_D \tau_L)^{-1/2}$], and the deformation is normalized to the quantity $K_n^0 I_i = (1-R) dI_i/h\nu_L \rho c_2^3$. According to Fig. 20, as the surface recombination velocity is increased from $M_S \ll M_D$ (curve *1*) to $M_S \gg M_D$ (curve *3*), the optoacoustic pulse length decreases approximately fourfold at half-height.

We note that situations are possible in which the slowing of free carriers by scattering from defects, ionized impurities, and phonons does not compensate locally the actions of the forces of the internal pressure of the photogenerated electron-hole plasma.[106] The realization of hydrodynamic (rather than diffusion) expansion of the electron-hole plasma is most probable at low (liquid-helium) temperatures.[106–108] Very simple models of sound generation by the electronic mechanism with fast hydrodynamic motion of the electron-hole pairs have been proposed in Refs. 109–111. A characteristic feature of the excitation of deformation waves in this regime is the predominant localization of the sound sources at the leading edge of the expanding electron-hole plasma.[109,110] The profile of the optoacoustic pulses is largely determined by the time dependence of the velocity of the plasma front.[109] The complexity of the detailed analysis of the process is due to both the quasisynchronous character of the generation of coherent acoustic waves at near-sonic velocities of the front[110,111] and the stimulated character of the radiation of phonons when they move at supersonic velocities.[112] In both cases the reverse (slowing) influence of the excited acoustic waves on the expansion of the electron-hole plasma is substantial.[110,112]

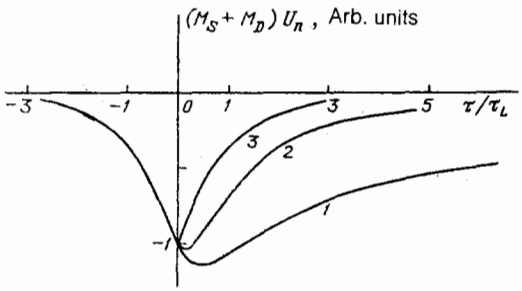

FIG. 20. Deformation profiles in optoacoustic pulses for supersonic diffusion of electron-hole pairs and different values of the surface recombination velocity ($M_S = S/c_a$). $M_S = 0$, $M_D \gg 1$ (*1*), $M_S = M_D \gg 1$ (*2*), $M_S \gg M_D \gg 1$ (*3*).

In closing this section we point out that ultrashort sound pulses generated by the concentration-deformation mechanism have been observed in a-As_2Te_3.[33] In addition, there are indications that the assumption used in writing Eq. (3.21), namely that the carriers instantaneously relax to the bottom of the energy bands can break down in optoacoustic experiments. According to unpublished data of Grahn *et al.*,[113] the velocity of spatial expansion of a photogenerated electron-hole plasma depends on the excess of the photon energy $h\nu_L$ over the width of the band gap E_g of the crystal.

3.3. Laser initiation of piezogeneration of sound

Breakdown of the neutrality of the electron-hole plasma ($n^e \neq n^h$), which is possible at high frequencies and was not taken into account in Sec. 3.2, plays a fundamental role in optical excitation of piezoelectric crystals, since it results in an additional mechanism of exciting sound. The electric field E that arises upon spatial separation of electrons and holes (owing to their differing mobilities) deforms the crystal as a result of the piezoelectric effect:[76,77,96,114]

$$G = G_T + G_n + G_E, \quad G_E = -eE, \tag{3.31}$$

where e is the effective piezoelectric modulus in the action of an electric field along the z axis.

In very simple models describing the internal macroscopic electric field E, one neglects the direct piezoelectric effect, while assuming the electromechanical coupling to be weak. If we do not take account of the piezoelectric fields that arise in the deformation of the crystal, the equation for E is

$$\varepsilon\varepsilon_0 \frac{\partial E}{\partial z} = Q_V, \quad \underset{z=0}{E = 0}, \quad \underset{z\to\infty}{E = 0}, \tag{3.32}$$

where ε_0 is the dielectric permittivity of the vacuum, and ε is the relative dielectric permittivity.

It is assumed in the problem Eq. (3.32) that the surface $z = 0$ is uncharged, which agrees with the subsequent neglect of the surface recombination of electrons and holes. In this discussion we shall also neglect the bulk recombination of electrons and holes, which when necessary can be taken into account by analogy with Sec. 3.2.[96,114,115] Then the following formula is valid for the volume charge density Q_V in (3.32):

$$Q_V = -Q_0(n^e - n^h), \tag{3.33}$$

where Q_0 is the magnitude of the electron charge. To describe the diffusion separation of electrons and holes, we shall use an equation linearized in the small deviations of the concentrations of free charge carriers from their equilibrium values n_0^m ($n^m \equiv n_0^m + n^m$, $|n^m| \ll n_0^m$):

$$\frac{\partial n^m}{\partial t} = D_m \frac{\partial^2 n^m}{\partial z^2} \pm \mu_m n_0^m \frac{\partial E}{\partial z} + \frac{W}{h\nu_L}. \tag{3.34}$$

The conditions for the absence of fluxes of carriers through the surface of the crystal are

$$j^{m} = -D_{m}\frac{\partial n^{m}}{\partial z} \mp \mu_{m} n_{0}^{m} E \underset{z=0}{=} 0. \tag{3.35}$$

By using Eqs. (3.32) and (3.33), we can conveniently represent (3.34) in the form

$$\frac{\partial n^{m}}{\partial t} = D_{m}\frac{\partial^{2} n^{m}}{\partial z^{2}} \mp \omega_{Mn}(n^{e} - n^{h}) + \frac{W}{h\nu_{L}}. \tag{3.36}$$

We can see already from Eqs. (3.36) and (3.21) that the frequencies of Maxwellian relaxation of charges ($\omega_{Mm} \equiv \mu_{m} n_{0}^{m} Q_{0}/\varepsilon\varepsilon_{0}$) can play a role in the piezoelectric generation of sound analogous to that of the recombination frequency ω_{R} in the electronic mechanism. However, in contrast to recombination, which prevents an increase in the density of the electron-hole plasma, Maxwellian relaxation prevents the breakdown of electric neutrality (deviation of n^{e} from n^{h}).

In the limting case of a quasineutral electron-hole plasma ($n^{e} \simeq n^{h} \equiv n$), the electric field E that arises in the system must bring about the joint motion of electrons and holes ($j^{e} \approx j^{h}$). The conditions $n^{e} = n^{h}$ and $j^{e} = j^{h}$ allow one to find this field (called the Dember field) directly by using Eq. (3.35)

$$E \equiv E_{D} = -\frac{D_{e} - D_{h}}{\mu_{e} + \mu_{h}}\frac{1}{n_{0}}\frac{\partial n}{\partial z}. \tag{3.37}$$

However, the density n of the quasineutral plasma in Eq. (3.37) is determined by the equation of ambipolar diffusion, Eq. (3.21). The generation of shear acoustic waves by the Dember field has been analyzed in Ref. 116, and the generation of longitudinal waves in Ref. 77.

Before we proceed to the results of investigations of the piezoelectric excitation of sound, we note that a description of the dynamics of the carriers based on Eqs. (3.32)–(3.35) may also necessary at high frequencies in analyzing the concentration-deformation mechanism (Sec. 3.2). The influence of the spatial separation of electrons and holes can be especially important when the deformation potentials d^{e} and d^{h} have opposite signs.

The general solution of the problem of Eqs. (3.31)–(3.35) was derived[42] by using the integral transforms of Eq. (3.3). The description of the spectrum $\tilde{U}_{E}(\omega)$ in (3.5) of a deformation pulse excited by the inverse piezoelectric effect is considerably simplified when mobile holes are generated only in the photoexcitation of the crystal ($n_{0}^{h} \equiv 0$). For the conventional model of optical sources of Eq. (3.9), the spectrum of the optoacoustic pulse (for $n_{0}^{h} = 0$) can be written

$$\tilde{U}_{E}(\omega) = \frac{(1-R)eQ_{0}(D_{e} - D_{h})}{h\nu_{L}\varepsilon\varepsilon_{0}\rho c_{a}^{4}}\frac{\omega_{\alpha}^{2}}{\omega^{2} + \omega_{\alpha}^{2}} \times \frac{(-i\omega)\omega_{De}\omega_{Dh}}{[\omega^{2} + \omega_{De}(\omega_{Me} - i\omega)](\omega_{Dh} + i\omega)} I_{i}\tilde{f}(\omega). \tag{3.38}$$

In Eq. (3.38) we have separated out the characteristic frequencies $\omega_{Dm} = c_{a}^{2}/D_{m}$, the physical meaning of which is similar to that of the frequency ω_{D} (Sec. 3.2). The proportionality of the optoacoustic signal (3.38) to the difference of the diffusion coefficients of electrons and holes indicates the diffusion nature of the separation of charges.

Because of Eq. (3.38) one can separate out in the spectral transfer function of piezooptoacoustic conversion a factor that fully characterizes the influence of the depth of penetration of radiation on the efficiency of conversion of light into sound:

$$K_{E}^{1}(\omega) \equiv K_{D}^{1}(\omega)K_{\alpha}(\omega), \quad K_{\alpha}(\omega) = \omega_{\alpha}^{2}/(\omega^{2} + \omega_{\alpha}^{2}), \tag{3.39}$$

$$K_{D}^{1}(\omega) = \frac{(-i\omega)\omega_{De}\omega_{Dh}}{(\omega^{2} - i\omega\omega_{De} + \omega_{Deb}^{2})(\omega_{Dh} + i\omega)}. \tag{3.40}$$

The spectral transfer function $K_{D}^{1}(\omega)$ does not depend on ω_{α}, but only on the other characteristic "internal" frequencies: ω_{De}, ω_{Dh}, and $\omega_{Deb} = \sqrt{\omega_{Me}\omega_{De}}$, the Debye frequency of screening of the electron charge. The possibility of factoring $K_{E}^{1}(\omega)$ of (3.39) indicates that, just as in thermoelastic sound generation (Sec. 3.1), the finite depth of penetration of light necessarily limits the efficiency of conversion of frequencies in the region $\omega > \omega_{\alpha}$.

This limit does not affect optoacoustic conversion [$K_{E}^{1}(\omega) \approx K_{D}^{1}(\omega)$] if ω_{α} substantially exceeds the other characteristic frequencies: $\omega_{\alpha} \gg \omega_{DM}, \omega_{Deb}$. If $\omega_{Deb} \ll \omega_{Dm}$, the spectral transfer function $K_{D}^{1}(\omega)$ actually corresponds to piezoelectric insulators:

$$K_{D}^{1}(\omega) \approx \frac{\omega_{De}\omega_{Dh}}{(\omega_{De} + i\omega)(\omega_{Dh} + i\omega)}, \tag{3.41}$$

since it does not depend on the equilibrium concentration of charge carriers. According to (3.41), to expand the spectral band of efficiently converted frequencies, we must first of all decrease the mobility of the electrons (when $\mu_{e} > \mu_{h}$), and then that of holes.

In the limiting situation $\omega_{Dm} \ll \omega_{L}$ the inverse Fourier transform of (3.4) with the aid of expression (3.38) and (3.41) enables one to obtain the following description of the profile of the deformation pulse:

$$U_{E}(\tau) = \frac{K_{E}^{0}}{2} I_{i}\tau_{L}\frac{\omega_{De}\omega_{Dh}}{\omega_{De} - \omega_{Dh}}\left(1 - \frac{\tau}{|\tau|}\right)\exp(\omega\tau)\Big|_{\omega_{Dh}}^{\omega_{De}}.$$

In the case of immobile holes ($D_{h} \to 0$, $\omega_{Dh} \to \infty$), the latter relation defines the optoacoustic signal ($U_{E}(\tau) \sim [1 - (\tau/|\tau|)]\exp(\omega_{De}\tau)$) with the characteristic duration $\tau_{a} = \omega_{De}^{-1} = D_{e}/c_{L}^{2} \equiv \tau_{D}$. Physically, the time τ_{D} is the time elapsed after the laser δ-exposure when the acoustic wave propagating from the boundary passes the front of the cloud of diffusing electrons ($c_{L}\tau_{D} = \sqrt{D_{e}\tau_{D}}$). This time $\tau_{D} > 0$ exists because for short times the diffusion of free electrons occurs at supersonic velocities. Actually, in times of the order of τ_{D} the velocity of sound and the characteristic decreasing velocity of carrier diffusion also become equal. Thus, in this regime the characteristic acoustic pulse length equals the time of detachment of the acoustic wave from the diffusion wave. Physically this is explained by the fact that, after the deformation pulse overtakes the front of moving electrons, it ceases to strengthen.

However, more importantly an increase in the Debye screening frequency ω_{Deb} with increased n_{0}^{e} due to doping of the semiconductors can shorten the acoustic pulses. When $\omega_{Deb} > \min(\omega_{Dm})$, the maximum efficiency of optoacoustic conversion is realized at the frequency $\omega \sim \omega_{Deb}$. However, we note that the $K_{D}^{1}(\omega)$ in Eq. (3.40) with increasing ω_{Deb}

occurs because of the decrease in the efficiency of conversion of the low frequencies. Actually, screening of the low-frequency ($\omega < \omega_{Deb}$) electric fields occurs with increasing ω_{Deb}.

In the limiting case $\omega_{Dm} \ll \omega_{Deb} \ll \omega_L$, we can use the following expression for $K_E^1(\omega)$ to calculate the profiles of deformation pulses:

$$K_E^1(\omega) \approx K_D^1(\omega) \approx -\frac{\omega_{De}\omega_{Dh}}{\omega^2 + \omega_{Deb}^2}. \tag{3.42}$$

Using Eqs. (3.4), (3.38), and (3.42), we find

$$U_E(\tau) = -\frac{K_E^0}{2} I_i \frac{\omega_{De}\omega_{Dh}}{\omega_{Deb}} \tau_L \exp(-\omega_{Deb}|\tau|).$$

Consequently, the duration of the acoustic pulse in this regime is governed by the time of Debye screening ($\tau_a = 2\omega_{Deb}^{-1}$).[42] If, however, the light absorption cannot be considered to take place at the very surface then $K_E^1(\omega)$ is given by Eq. (3.39). In particular, where $\omega_{Dm} \ll (\omega_{Deb}, \omega_\alpha) \ll \omega_L$, we find by using Eqs. (3.4), (3.39), and (3.42) that

$$U_E(\tau) = -\frac{K_E^0}{2} I_i \frac{\omega_{De}\omega_{Dh}}{\omega_{Deb}} \tau_L \frac{\omega_\alpha}{\omega_{Deb}^2 - \omega_\alpha^2} \times [\omega_{Deb}\exp(-\omega_\alpha|\tau|) - \omega_\alpha \exp(-\omega_{Deb}|\tau|)].$$

Thus the profile of the optoacoustic pulse depends to a large extent on the ratio of the frequencies ω_{Deb} and ω_α. An important fact is ω_{Deb} is large enough even at a relatively low degree of doping of the semiconductors. In the case of nondegenerate statistics of the non-equilibrium carriers we have

$$\omega_{Deb} = \sqrt{\omega_{Me}\omega_{De}} = Q_0 c_a (n_0^e/\varepsilon\varepsilon_0 k_B T)^{1/2},$$

and at room temperature $\omega_{Deb} > 10^{11}$ s^{-1} for $n_0^e > 10^{16}$ cm^{-3}.

Just as for the concentration-deformation mechanism, in the surface absorption of light a substantial role can be played in piezoelectric generation by surface recombination of carriers. To take account of its influence, one modifies the boundary conditions (3.35) and (3.32):

$$-D_m \frac{\partial n^m}{\partial z} \mp \mu_m n_0^m E \underset{z=0}{=} -S_m n^m,$$

$$\varepsilon\varepsilon_0 E \underset{z=0}{=} -Q_0 \int_{-\infty}^{t} (S_e n^e - S_h n^h) dt'.$$

The last relation describes the connection between the surface charge and the velocities S_m of surface recombination of electrons and holes. The spectral transfer function modified in the case of piezodielectrics ($n_0^m \equiv 0$) and surface absorption ($\alpha \to \infty$) depends on the characteristic frequencies ω_{Dm} and $\omega_{Sm} = S_m^2/D_m$. If the surface recombination is weak, $\omega_{Sm} \ll \omega_{Dm}$, then $K_D^1(\omega)$ acquires the form (3.41), according to which the efficiency of optoacoustic conversion declines sharply ($|K_D^1| \sim \omega^{-2}$) at frequencies $\omega > \omega_{Dm}$. Analysis shows that strong surface recombination retards the decrease in efficiency with frequency. For example, in the limiting regime $\omega_{Dm} \ll \omega_{Sm}$ the following form of $K_D^1(\omega)$ is valid:

$$K_D^1(\omega) = -\frac{\omega_{De}\omega_{Dh}}{(\omega_{De} - \omega_{Dh})} \frac{\omega_{Se}^{1/2} - \omega_{Sh}^{1/2}}{\sqrt{-i\omega}(\omega_{Se}^{1/2} + \sqrt{-i\omega})(\omega_{Sh}^{1/2} + \sqrt{-i\omega})}. \tag{3.43}$$

Additional analysis shows that in this case the spectrum of the acoustic pulse replicates the spectrum of the electric field and the charge on the surface: $\tilde{U}_E(\omega) \sim \tilde{E}(\omega, z=0)$. As we see from (3.43), the surface becomes charged if $\omega_{Se} \neq \omega_{Sh}$. In the opposite case, even with strong surface recombination, excitation of sound in the bulk of the crystal dominates. The dependence of the profile of the optoacoustic pulse on the ratio of ω_{Se} to ω_{Sh} is seen most distinctly when $\omega_{Dm} \ll \omega_{Sh} \ll \omega_\alpha$, ω_L:

$$U_E(\tau) \sim \left(1 + \frac{\tau}{|\tau|}\right) \exp(\omega\tau)\operatorname{erfc}\sqrt{\omega\tau}\Big|_{\omega=\omega_{Se}}^{\omega=\omega_{Sh}}.$$

In the limit when holes are not trapped by the surface ($S_h = 0, \omega_{Sh} = 0$), the last formula describes a unipolar deformation pulse ($U_E(\tau) \sim -[1 + (\tau/|\tau|)]\exp(\omega_{Se}\tau)$ $\operatorname{erfc}\sqrt{\omega_{Se}\tau}$) having the characteristic duration

$$\tau_a \sim \omega_{Se}^{-1} = D_e/S_e^2 \equiv \tau_S.$$

In its physical meaning the time τ_S is the time of surface recombination of condution electrons photogenerated near the surface by instantaneous laser exposure. In the time τ_S the diffusion spreading of the cloud of non-equilibrium electrons reduces their characteristic diffusion velocity $v_{De} \sim \sqrt{D_e/t}$ such that it becomes equal to the velocity of surface recombination S: $\sqrt{D/\tau_S} = S$. Therefore, at times greater than τ_S, diffusion of carriers cannot prevent their capture by the surface. Thus, under these conditions the characteristic duration τ_a of the acoustic pulse is of the order of the time τ_S of capture of mobile non-equilibrium electrons by the surface of the piezoelectric crystal. Physically, this is explained by the fact that charge redistribution near the surface is complete in a time of the order of τ_S.

We can point out another possibility of shortening the duration of an optoacoustic pulse. Here, an external electric field E_0 can hinder the diffusion of charged particles from the surface and the consequent expansion of the region of sound generation. More exactly, one can create conditions under which the motion of fast carriers from the surface is retarded and that of slow particles accelerated, while on the whole the expansion of the cloud of photogenerated carriers occurs more slowly. For example, in a piezodielectric ($n_0^e = n_0^h = 0$) with immobile (heavy) holes ($\mu_h = 0$), the field E_0 must be oriented along the positive direction of the z axis ($E_0 > 0$) so as to force the mobile electrons toward the surface of the crystal. Although, when $E_0 \neq 0$, the specimen is deformed even in the stationary state, nonetheless, in the linear approximation for acoustic waves, Eq. (3.1) is valid, as before. Also Eqs. (3.32) and (3.33) maintain their form, but for increases of the electric field E.

The equations of motion of the carriers linearized by using the condition $|E| \ll E_0$ are

$$\frac{\partial n^m}{\partial t} = D_m \frac{\partial^2 n^m}{\partial z^2} \pm \mu_m E_0 \frac{\partial n^m}{\partial z} + \frac{W}{h\nu_L},$$

$$-D_m \frac{\partial n^m}{\partial z} \mp \mu_m E_0 n^m \underset{z=0}{=} 0. \tag{3.44}$$

On the basis of the model Eqs. (3.31)–(3.33) and (3.44), a description was obtained in Ref. 42 for the spectrum of the deformation pulse in the case $\mu_h = D_h = 0$. In particular, in the case of surface absorption of light the spectral transfer function of piezooptoacoustic conversion is

$$K^1_{E_0}(\alpha \to \infty) \approx \frac{(-i\omega)M\omega_{De}}{\omega^2 + [(M\omega_{De}/2) + \sqrt{(M\omega_{De}/2)^2 - i\omega\omega_{De}}]^2}. \tag{3.45}$$

Here we have introduced the notation $M = v_{de}/c_a = \mu_e E_0/c_a$ for the Mach number of drift motion of the electrons in the *dc* electric field E_0 (v_{de} is the drift velocity). At subsonic drift velocities of the carriers ($M^2 \ll 1$), Eq. (3.45) qualitatively differs from (3.41) only because of the additional assumption that the holes are fixed ($\omega_{Dh} = \infty$). However, now one can shift the region of efficient optoacoustic conversion toward high frequencies, not only by increasing ω_{De}, but also by increasing the field E_0 (i.e., increasing M). However, we note that the characteristic frequency $M\omega_{De}$, in contrast to ω_{De}, does not depend on the mobility of the carriers. When $M \gg 1$ the maximum efficiency is obtained when $\omega \sim M\omega_{De} \gg \omega_{De}$:

$$K^1_{E_0} \approx \frac{(-i\omega)M\omega_{De}}{\omega^2 + M^2\omega^2_{De}}. \tag{3.46}$$

Comparison of expressions (3.46) and (3.13) shows that the profile of the acoustic pulse in the case $\omega_{De} \ll M\omega_{De} \ll \omega_\alpha, \omega_L$ resembles (3.14), while its duration is $\tau_a \sim 2(M\omega_{De})^{-1}$. We note that the pulse is shortened because, as the field increases, it can arrest diffusion at ever higher frequencies.

If the absorption cannot be considered to be of surface type, then in the most interesting case of strong electric fields ($M^2 \gg 1$) the following relation holds:

$$K^1_{E_0} \approx \frac{(-i\omega)M\omega_{De}(\omega^2_\alpha - M^2\omega^2_{De})}{(\omega^2 + M^2\omega^2_{De})(\omega^2 + \omega^2_\alpha)}. \tag{3.47}$$

When $\omega_\alpha \gg M\omega_{De}$, this goes over to (3.46). When $\omega_\alpha = M\omega_{De}$, sound is not excited via the piezoelectric mechanism, since charge separation does not occur. This is a consequence of the fact that, with an exponential depth distribution of the optical sources of non-equilibrium charge carriers, the velocity of possible diffusion ($v_{De} \sim n_e^{-1} D_e (\partial n_e/\partial z) \sim \alpha D_e$) does not depend on z and can be compensated by drift in a homogeneous electric field at every point of space ($\alpha D_e = v_{de}$ when $\omega_\alpha = M\omega_{De}$).

For $M\omega_{De} > \omega_\alpha$, the electrons are predominantly displaced toward the boundary; i.e., charge separation occurs not as a result of diffusion of electrons, but as a result of their drift. The direction of the internal electric field E, and also, as is implied by expression (3.47), the polarity of the acoustic pulse, are inverted. The profile of the acoustic pulse in the case $\omega_{De} \ll (M\omega_{De}, \omega_\alpha) \ll \omega_L$ can be written as[42]

$$U_{E_0} = \frac{K^0_{E_0}}{2} M\omega_{De} I_1 \tau_L \left(\frac{\tau}{|\tau|}\right) [\exp(-\omega_\alpha|\tau|) - \exp(-M\omega_{De}|\tau|)].$$

In the last description $K^0_{E_0} \sim M^{-1}$,[42] and therefore, when $M\omega_{De} \gg \omega_\alpha$, the increase in amplitude of the wave with increasing E_0 is completely saturated.

Reference 42 reported a numerical comparison of the efficiencies of the different mechanisms in the generation of acoustic pulses of duration $\tau_a \approx 25$ ps in GaAs. The z axis lies along the piezoactive ⟨111⟩ direction in the crystal. To generate such deformation pulses, first, one must use laser pulses of duration $\tau_L \lesssim 25$ ps, and second, the upper bound of the spectrum of efficiently converted frequencies must lie in the region $\omega \gtrsim \omega_a \sim 4\tau_a^{-1} = 1.6 \times 10^{11}$ s^{-1}.

In the thermoelastic mechanism of optoacoustic conversion, the relationship $\omega_\alpha \gtrsim \omega_a$ must be realized to fulfill the second condition. This allows us to estimate: $\alpha \gtrsim 3 \times 10^5$ cm^{-1}. Consequently, the light absorption must be of interband type (with $h\nu_L \gtrsim 2E_g$). An estimate of the parameter m_χ for $\alpha \approx 3 \times 10^5$ cm^{-1} yields: $m_\chi \approx 0.1 \ll 1$. Consequently heat conduction does not influence the process of sound generation, while the listed conditions ($\tau_L \lesssim 25$ ps, $\alpha \approx 3 \times 10^5$ cm^{-1}) are sufficient.

Under the same conditions of absorption we have $m_D \gg 1$, and hence the diffusion of the electron-hole plasma broadens the acoustic pulse excited by the electronic mechanism. To generate pulses of duration $\tau_a \approx 25$ ps we must also have $\omega_R \lesssim \omega_a$ ($\tau_R \lesssim 6$ ps). Such nonlinear recombination times of the electron-holes plasma are obtained in GaAs at concentrations of free carriers $n \gtrsim 10^{19}$ cm^{-3} (Ref. 117). Upon attaining $\omega_R \sim \omega_a$ (e.g., by increasing the intensity of photoexcitation), the ratio of the efficiencies of the electronic and thermoeleastic mechanisms is determined by virtue of Eqs. (3.10) and (3.24), by the dimensionless parameter:

$$\left|\frac{\tilde{U}_n(\omega \sim \omega_\alpha \sim \omega_R \sim \omega_a)}{\tilde{U}_T(\omega \sim \omega_\alpha \sim \omega_a)}\right| \approx \frac{|d|\rho c}{(h\nu_L - E_g)K\beta}_{GaAs} \approx 10.$$

Thus the electronic mechanism proves to be an order of magnitude more efficient than the thermoelastic mechanism.

To generate acoustic pulses of duration $\tau_a \sim 25$ ps under the conditions being discussed via the inverse piezoelectric effect, one must increase the frequency of Debye screening to $\omega_{Deb} \gtrsim \omega_a \approx 1.6 \times 10^{11}$ s^{-1}, which can be done at very moderate concentrations of free electrons $n^e \gtrsim 10^{16}$ cm^{-3}, achieved by doping the GaAs. Under optimal conditions the efficiency of piezooptoacoustic conversion is an order of magnitude higher, in view of relations (3.24) and (3.38), than that due to the electronic mechanism:

$$\left|\frac{\tilde{U}_E(\omega \sim \omega_{Deb} \sim \omega_\alpha \sim \omega_a)}{\tilde{U}_n(\omega \sim \omega_R \sim \omega_\alpha \sim \omega_a)}\right| \sim \frac{|e|Q_0 c_a^3 (D_e - D_h)}{\varepsilon\varepsilon_0 D_e D_h |d| \omega_a^2}_{GaAs} \approx 10.$$

Finally, if an electric field $E_0 \gtrsim 10^4$ V/cm is present in the region of light absorption, then drift separation of photogenerated electrons and holes is realized. Here the relative efficiency of the piezo- and electronic mechanisms can be estimated as follows:

$$\left|\frac{\tilde{U}_{E_0}(\omega \sim \omega_\alpha \sim \omega_a \ll M\omega_{De})}{\tilde{U}_n(\omega \sim \omega_\alpha \sim \omega_a \gg \omega_D)}\right| \approx \frac{|e|Q_0 c_a}{\varepsilon\varepsilon_0 |d| \omega_a}_{GaAs} \approx 6 \cdot 10^2.$$

This estimate points up the promise of using such optoelectric schemes for efficient generation of deformation waves. We note that the increase in efficiency of laser piezogeneration of surface waves in CdS when a supplementary external electric field is applied has been observed experimentally.[118]

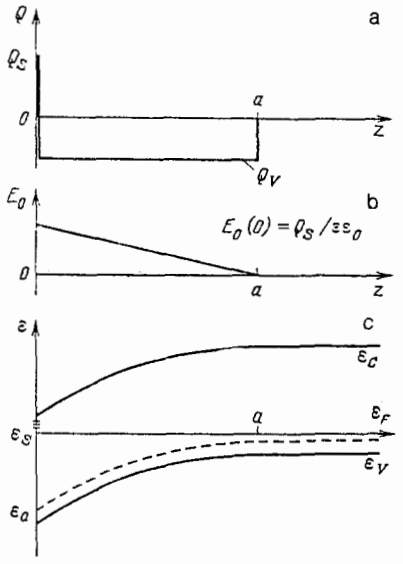

FIG. 21. Distribution of charge density (a), electric field (b), and the spatial structure of the energy levels of a p-type semiconductor (c) near the surface $z = 0$. $\mathscr{E}_S$—hole-trapping levels, $\mathscr{E}_a$—acceptor impurity level, $\mathscr{E}_{F,c,v}$—Fermi level, conduction band, and valence band, Q_S—surface charge density.

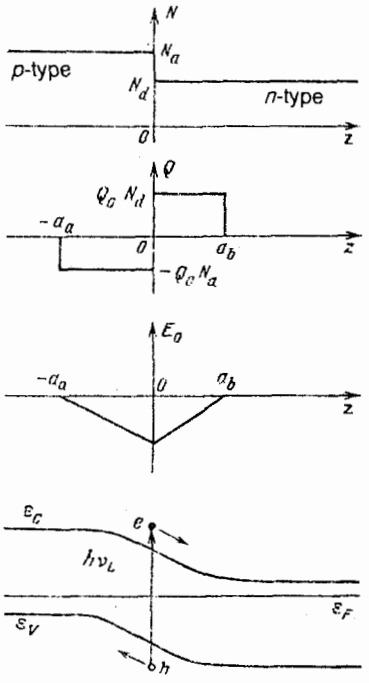

FIG. 22. Simple model of a p–n junction (N_{ad}—concentration of acceptor and donor dopants).

We note especially that laser generation of high-frequency acoustic waves in the presence of dc electric fields is not at all an exotic case difficult to implement in practice. Actually, electric fields are always present near the surfaces of crystals, and in particular, surfaces of semiconductors. For example, if hole-trapping centers (e.g., Tamm surface states) exist on the surface of a p-type semiconductor, then the surface becomes charged positively. A layer of space charge is formed near the surface, in which the electric field $E_0(z)$ is directed away from the surface and can hinder the diffusion expansion of the photogenerated electrons (Fig. 21). In other words, the energy bands of the semiconductor are bent at the surface so that the minimum electron energy occurs near the surface. Therefore the photogenerated electrons drift toward the surface. If the photoexcitation of carriers occurs uniformly in space in the region of this built-in field ($\alpha^{-1} \gg a$, where a is the thickness of the depletion layer), then it is the electric field, rather than diffusion, that causes the charge separation.

An analogous situation can be found in the interband absorption of light in the region of a p–n junction (Fig. 22). The authors of Ref. 119 were the first to analyze the piezogeneration of sound by the separation of photogenerated charge carriers by the internal field of a p–n junction.[119] To give an idea of the magnitude of the possible effects, we point out that typically the width of the space-charge layer does not exceed a few micrometers, while the electric fields, which depend on the doping level, can attain values $E_0 \sim 10^4$–10^5 V/cm.[120] In intense brief laser exposures, such large fields can be nearly fully compensated by the internal electric fields E created by the separation of the non-equilibrium electrons and holes. Naturally, here the amplitudes of the excited deformation waves can be of the order of magnitude of the stationary deformations existing in the region of the p–n junction ($U \sim 10^{-6}$–10^{-4}).

A logical extension of the analysis of optoacoustic conversion at a single p–n junction is the study of laser generation of sound in heterostructures. Of course, it is not expedient to use heterostructures for exciting single ultrashort deformation pulses. However, generation of a sequence (train) of acoustic pulses can occur in a heterostructure by using a single laser pulse when the spatial modulation of the sound sources is determined by the periodicity of the heterostructure. Under certain conditions the acoustic signal actually can be quasiharmonic with a frequency determined by the time of passage of sound through the spatial period of the structure.

The idea of obtaining a quasiperiodic signal by using a single laser pulse was formulated in Ref. 82 for thermoelastic generation of sound in a medium having periodic modulation of the light absorption coefficent. In the case of heterostructures the spatial modulation of optical sources of nonequilibrium carriers can be unimportant. However, the modulation of sound sources can be predominantly associated with the motion of non-equilibrium carriers in the periodic potential of a heterostructure (Fig. 23). The most important circumstance is that at present it is technologically possible to grow heterostructures with a period of the order of the interatomic distances in the semiconductor (doped superlattices[120]). Thus the possibility has been opened for generation of coherent sound at the extreme attainable frequencies. We note that periodic electric fields can be associated with a space-charge lattice persisting after exposure of photorefractive crystals to crossed light fields. Sound generation by repeated exposure of a crystal to modulated laser radiation has been studied in Ref. 122. However, the maximum attainable frequencies of generated sound in this scheme are substantially lower than those attainable in superlattices.

Along with the generation mechanisms that we have already discussed (thermoelastic, deformational, and piezoelectric), the possiblity of direct action of the electric field of

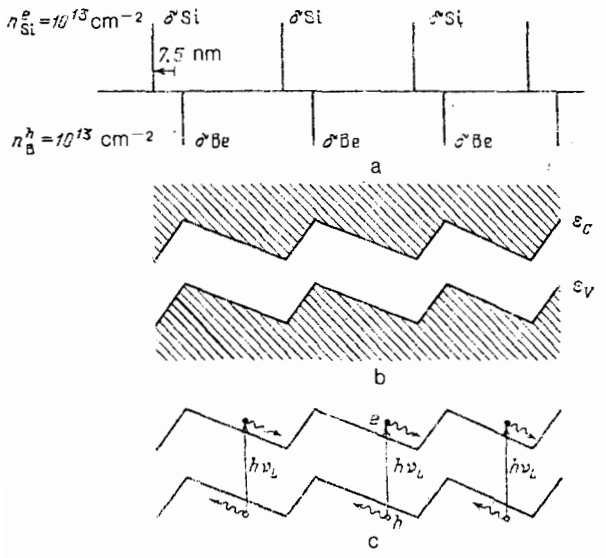

FIG. 23. Diagrams of the doping profile (a), saw-tooth band structure (b), and excitation of free carriers and charge transport (c) in a δ-doped specimen of GaAs.[121]

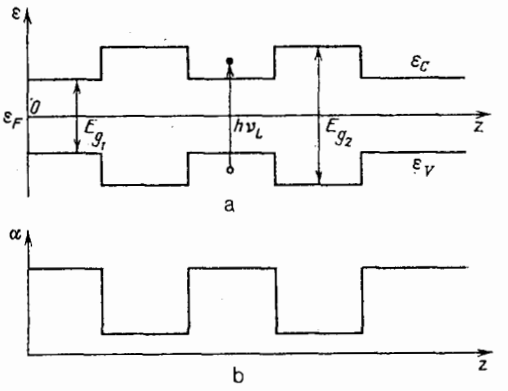

FIG. 24. Profiles of the energy bands of a semiconductor composite structure—sequences of single potential wells (a) and the coefficient of interband absorption of light of photon energy $E_{g1} < h\nu_L < E_{g2}$ (b).

the light wave on the layers of bound space charge has been discussed in the context of doped superlattices.[123-125] In this case the sources of sound in the wave equation (3.1) have the form

$$\frac{\partial G}{\partial z} = -Q_V(z)E_L(z, t), \tag{3.48}$$

where $Q_V(z)$ is the space charge density and $E_L(z,t)$ is the electric field of the electromagnetic wave. Since in this mechanism sound is generated at the frequency of the optical radiation, the generation of even terahertz sound requires one to use lasers in the far infrared range.[125]

Periodic electric fields just as large ($E_0 \sim 10^4$–10^5 V/cm) as in doped superlattices[126] (including δ-doped ones[121]) can be obtained also in composite superlattices in which the local quasielectric field arises in a region of smooth transition between semiconductors having different band gaps.[127] According to the results of Ref. 127, in regions with a space-dependent band gap (in regions of semiconductor composition variable in z), one can obtain highly supersonic ($M \gg 1$) velocities of drift motion of carriers for picosecond times. In particular, such a drift separation of charges can occur in each of the periods of the superlattice having a saw-tooth band gap profile E_g (Ref. 127).

When the superlattice consists of a sequence of isolated (rectangular) quantum wells,[120] the modulation of the acoustic sources when $E_{g1} < h\nu_L < E_{g2}$ is predominantly associated with the modulation of the light absorption coefficient (in the last inequality the $E_{g1,2}$ are the band gaps of the constituents of the two-component superlattice) (Fig. 24). A situation of this type has been analyzed in Ref. 128 in a study of the generation of Rayleigh surface waves in a normally sectioned superlattice. An important aspect is that the confinement of the photogenerated carriers inside the potential wells prevents the diffusion spreading of the concentration lattice and thus produces an enhanced efficiency of optoacoustic conversion at high frequencies.

Quasiharmonic optoacoustic signals was first experimentally observed in such compositional superlattices (a-Ge:H/a-Si:H).[113,129] They had frequencies determined by the spatial periodicity of the structure. We note that surface-localized high-frequency vibrations ($\nu_a \gtrsim 200$ GHz) were detected in addition to the propagating modes when the first surface layer was acoustically softer (a-Si:H). The picosecond technique of excitation and probing was used also for diagnostics of multilayer metal films.[130] Thermoelastic and/or deformational sound generation was achieved in these experiments.[113,129,130] However, experiments in structures based on piezoelectric conductor crystals have been performed only at considerably lower frequencies.[131,132]

Having touched upon the problems of laser generation of quasimonochromatic acoustic waves, we must not fail to mention the attempts at direct conversion (without frequency change) of optical radiation into sound at the boundary of transparent piezoelectric crystals.[133,134] According to Eqs. (3.31) and (3.1), the piezoelectric sources in this situation are δ-localized at the surface of the crystal [$\partial G_E/\partial z \sim \partial e/\partial z \sim \delta(z)$]. Reference 133 reported the use of this method of generalizing coherent acoustic shear waves in quartz at liquid-helim temperatures at frequencies of 0.891 and 2.53 THz. For this purpose modulated laser radiation in the far infrared region was used (wavelengths of 337 and 118 μm). However, subsequently several groups of investigators could not repeat the experiment of Ref. 133, even by using a more refined technique of data accumulation and processing.[134] Besides the strong scattering of terahertz acoustic waves during propagation, according to Ref. 134, the reason for this failure might be the nonideality of the surface of the piezoelectric crystal. A simple calculation explaining this assumption was performed[77] for longitudinal sound and an electromagnetic wave polarized along the direction of propagation z. The damage to the surface layer of the crystal was modeled by a gradual (rather than jumpwise) onset of piezoelectric properties with distance away from the boundary $z = 0$:

$$e = e_0[1 - \exp(-z/L)],$$

where L denotes the depth of the damaged layer. For such a situation the mathematical formalism of Eqs. (3.31) and (3.5) leads to the following description of the deformation spectrum in the traveling wave:

$$\tilde{U}(\omega) \approx \frac{e_0 E_0}{\rho c_a^2} \frac{1}{1 + (L\omega/c_a)^2} \tilde{f}(\omega - 2\pi\nu_L), \tag{3.49}$$

where E_0 is the amplitude of the sinusoidal electric field. The solution (3.49) is valid near the frequency $\omega \approx 2\pi\nu_L$. In writing it, we have taken account of the great difference in velocities of light and sound.

The factor $\tilde{f}(\omega - 2\pi\nu_L)$ in expression (3.49) describes the spectrum of excited acoustic waves in the case of an ideal boundary ($L = 0$). For $2\pi\nu_L \gg \tau_L^{-1}$, the sound is quasimonochromatic ($\omega \approx 2\pi\nu_L$). However, according to (3.49), nonideality of the boundary strongly suppresses the high-frequency acoustic waves in the region $\omega \gtrsim c_a/L$. Physically, this comes from the nonsynchronous excitation of sound at different points of the surbsurface layer and its quenching by interference. As L increases, the efficiency of sound generation declines. For efficient generation of acoustic waves with terahertz frequencies, the thickness of the damaged layer must not exceed the acoustic wavelength ($\sim$1–10 nm).

In closing the discussion of sound generation by laser initiation of the inverse piezoelectric effect, we only point out that there are other possibilities for the appearance of electric fields in the action of light on matter. In particular, the pyroeffect[135,136] and the Glass effect.[115,137]

3.4. Electrostrictive sound generation

It is impossible to generate acoustic waves by the surface piezoelectric effect or by direct action on bound charges as described in (3.48) by means of lasers in the visible range, since acoustic waves of such high frequencies ($\omega \sim 10^{15}\ s^{-1}$) do not propagate in crystals. Moreover, the piezoelectric effect is absent in all crystals having a center of symmetry. In such situations the components of acoustic sources quadratic in the magnitude of the electric field E can play a substantial role:

$$G_S = \frac{1}{8\pi} a_{ijkl} E_k E_l; \tag{3.50}$$

Here a_{ijkl} is the photoelastic or electrostriction tensor. Such an electrostriction mechanism of sound generation in Eq. (3.50) can be highly effective in the field of crossed light beams if the optical dispersion of the medium allows synchronous generation of sound at the difference frequency $\omega = 2\pi(\nu_{L1} - \nu_{L2})$, where $\nu_{L1,2}$ are the frequencies of the laser radiation.[138–140]

In the case of optical waves counterpropagating collinearly with the z axis, we can represent the sound sources in the form

$$G_S \approx \frac{1}{8\pi} Y E_{L1} E_{L2} \cos(\omega t - k_a z + \Delta k z - \varphi_0), \tag{3.51}$$

where we assume that a special choice of the geometry of the problem and of a crystal having a certain symmetry enables us to describe the generation by using a single electrostrictive constant Y.[141] In expression (3.51) we have omitted the harmonic sources having the frequecies $2\nu_{L1}$, $2\nu_{L2}$, and $\nu_{L1} + \nu_{L2}$, which do not lie in the spectral range of acoustic waves; k_a is the wave number of the sound, and φ_0 is a constant phase shift. The symbol Δk is used for the difference of wave numbers of the acoustic and light waves:

$$\Delta k = k_a - k_{L1} - k_{L2} = (\omega/c_a) - 2\pi[(\nu_{L1}/c_{L1}) + (\nu_{L2}/c_{L2})].$$

A description of the acoustic waves excited by the harmonic sources of (3.51) can also be obtained by using the solution (3.5).[77] In particular, the following expression is valid for the amplitude of deformation in a sound wave propagating from a rigid boundary at $z = 0$:

$$|U|_{max} = \frac{|Y| E_{L1} E_{L2}}{16\pi\rho c_a^3} \left| \frac{\sin(z\Delta k/2)}{\Delta k/2} \right|. \tag{3.52}$$

According to (3.52), when the conditions of wave synchronization are fulfilled ($\Delta k = 0$), the acoustic signal increases linearly with z, and to describe its limit, it may be necessary to take account of the abosrption of sound (including nonlinear absorption[141]). Nonlinear acoustic effects can play an important role also in stimulated Brillouin scattering.[141] We point out that a rather simple mathematical description of the nonlinear limitation of the acoustic signal excited by moving harmonic sources has been developed in Refs. 142–143.

Under conditions of synchronization, with account taken of the inequality $\omega \ll 2\pi\nu_{L1,2}$, the following relation holds between the frequencies of the acoustic and optical waves:

$$\nu_a \approx 2\frac{c_a}{c_L}\nu_L,$$

where c_L is the velocity of light in the cyrstal. For typical values of the parameters, the frequencies of acoustic waves when optical pumping is used do not exceed tens of GHz.[140] To proceed into the region of higher frequencies of sound, one must create sources of coherent short-wavelength radiation (far ultraviolet and x-ray ranges).

Biharmonic pumping can also be used to generate optical phonons (Raman scattering[144]). From the standpoint of generating high-frequency acoustic waves, it is important that the optical phonons decay effectively into acoustic phonons with wave vectors near the edge of the Brillouin zone (Fig. 25). Acoustic transverse phonons of terahertz frequencies have been experimentally observed in the decay of optical phonons, those created in Raman scattering,[145] and those emitted as the nonequilibrium carriers photogenerated by interband light absorption in semiconductors relax to the bottom of the energybands.[146,147] It was shown in Ref. 148 that a more effective method of obtaining nonequilibrium phonons, as compared with the method of biharmonic pumping,[145] is to use direct resonance excitation of dipole-active lattice vibrations. Here there is particular interest in the possibility of obtaining stimulated decay of optical phonons,[149] which might lead to generation of coherent acoustic

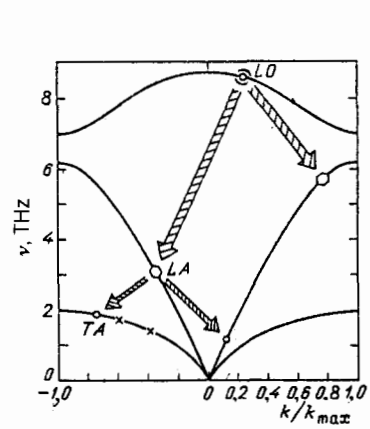

FIG. 25. Diagram of the decay of an optical phonon in GaAs satisfying the laws of conservation of energy and momentum in three-phonon processes.[146,147]

waves. We note that, according to Ref. 150, the increase in the occupation numbers of the optical modes can be compensated to such a degree by the broadening of the region of creation of acoustic phonons that the critical occupation numbers of the acoustic modes are not reached. Nonlinear effects in the generation of optical and acoustic phonons by resonance infrared radiation have also been discussed in Ref. 148.

In closing this section we point out that the interest in processess of light-induced non-steady-state interactions of optical and acoustic modes of crystal vibration has not waned up to the present.[17,151,152] Broad prospects are opened up also by the recently achieved generation in semiconductors,[153] metals,[154] and superconductors[155] of coherent monochromatic optical phonons by the surface absorption of femtosecond laser pulses. In the opinion of the authors of Ref. 153, the excitation of an optical mode in GaAs results from the action of non-steady-state electric fields created by the spatial separation of photogenerated electrons and holes [see, e.g., Eqs. (3.32)–(3.35) and (3.44)]. The changes in the electric fields can be particularly large as a result of the screening of the built-in electric field near the surface of the crystal by nonequilibrium photogenerated carriers (Fig. 21). We note that radiation by the surface of photoexcited semiconductors of femtosecond electromagnetic video-frequency pulses has already been detected experimentally.[156,157] This is evidence favoring the mechanism proposed in Ref. 153 for the generation of monochromatic optical phonons, and also favoring the possibility of generation of ultrashort longitudinal acoustic pulses via the inverse piezoelectric[42] (Sec. 3.13).

3.5. Sound generated by light pressure and in the processes of melting and ablation of targets

In general, the flux of optical radiation incident on the phase boundary of two media is split into reflected and refracted components. Here the momentum vector of the photon changes. The pressure exerted in this process on the specimen by the electromagnetic field is called the light pressure. The resultant pressure on the surface is determined by the law of conservation of momentum:

$$P_L = \left(\frac{I_R + I_i}{c_L} - \frac{I_T}{c_{Ln}}\right) f(t) = \left[(1+R)\frac{I_i}{c_L} - (1-R)\frac{I_i}{c_{Ln}}\right] f(t), \tag{3.53}$$

where $I_{R,T}$ are the intensities of the reflected and transmitted radiation, and c_L and c_{Ln} are the velocities of light in the medium from which the exposure is exerted and in the specimen. Equation (3.53) corresponds to the case of normal incidence of the radiation.

The case most promising from the standpoint of detecting acoustic pulses caused by light pressure of ultrashort laser pulses is that of optical irradiation of the phase boundary between two transparent media. Here, in the absence of absorption of the energy of the radiation, the observation of pulses of light pressure is not impeded by the thermoelastic, deformation, or the piezoelectric mechanisms of sound generation. The strictive excitation of acoustic waves in (3.50) occurring at the boundary (in a region of sharp change in the photoelasticity tensor) is one of the components of light pressure. Also, the total action of the electromagnetic field on the cyrstal in the case of transparent media is also localized in the region of the spatial gradient of the dielectric permittivity.[158] Vassilev[159] has studied the dependence of the light pressure on the angle of incidence of ultrashort laser pulses.

For light incident on the surface of a highly reflective medium ($1 - R \ll 1$) it is possible in principle that under certain conditions, light pressure can dominate over the other mechanisms of sound generation. In this situation the optical action also occurs at the surface, since when $R \approx 1$ we can neglect the momentum carried away by the refracted wave:

$$P_L \approx \frac{2I_i}{c_L} \approx 1\ \text{Gbar} \cdot \frac{I_i}{1{,}5 \cdot 10^{18}\ \text{W/cm}^2}. \tag{3.54}$$

Analysis shows that, when P_L predominates, besides increasing the light reflection coefficient R, one must create nonoptimal conditions for producing the other mechanisms of optoacoustic conversion. For this purpose, in the experiment of Ref. 160 an aluminum coating was used (which provided large values of $R \approx 0.975$ (for $\lambda_L \sim 10.6\,\mu$m) and of the heat conductivity $\chi \sim 1$ cm^2/s), in addition to long laser pulses ($\tau_L \sim 1\,\mu$s). In this case the region of light absorption is thermally thin for thermoelastic generation of sound, while the heat conductivity at the characteristic frequencies $\omega \sim \omega_L \sim \tau_L^{-1}$ is considerably subsonic. At these frequencies the use of the transfer function (3.16) and Eq. (3.54), give us the estimate

$$\frac{P_L}{P_T} \sim \frac{2}{1-R}\frac{c_a}{c_L}\frac{c}{\beta c_a^2}\frac{c_a\tau_L}{\sqrt{\chi\tau_L}} \sim \tau_L^{1/2}. \tag{3.55}$$

For typical metals the dimensionless parameter $c/\beta c_a^2$ is of the order of unity (in Al: $c/\beta c_a^2 \approx 1.5$). By our estimates in Ref. 160 we obtained the condition $P_L \sim P_T$, where P_T is the pressure in the acoustic wave excited by the thermoelastic wave. At the same time, even in experiments with nanosecond laser pulses,[161] the thermoelastic generation of sound dominates.

With ultrashort laser pulses, supersonic electronic heat conduction is obtained in metals ($|K_T^I| \approx \sqrt{\omega_\chi/|\omega|}$). Here, instead of (3.55), the following estimate is valid:

$$\frac{P_L}{P_T} \sim \frac{2}{1-R}\frac{c_a}{c_L}\frac{c}{\beta c_a^2}\frac{\sqrt{\chi\tau_L}}{c_a\tau_L} \lesssim \frac{2}{1-R}\frac{v_F}{c_L}\frac{c}{\beta c_a^2}, \tag{3.56}$$

where we have taken into account that the velocity of diffusion heat conduction cannot exceed the Fermi velocity v_F of the degenerate electrons. For typical values $v_F \sim 2 \times 10^8$ cm/s, we must have $R \gtrsim 0.98$ to satisfy $P_L \sim P_T$.

In constrast to the situations (3.55) and (3.56) discussed above, the action of electromagnetic radiation on specimens of poor reflectivity ($R \ll 1$) is distributed over the region of absorption of photons passing through the phase boundary of the media. As the preceding analysis shows, the light pressure in this case will be strongly masked by the action of the other mechansims of sound generation (in particular, the thermoelastic mechanism). Thus, the hopes of observing light pressure in a pulsed mode can hinge only on restricting the efficiency of the other mechanisms of optoacoustic converion via nonlinear effects. The point is that, e.g., the pressure P_{SH} in the shock wave excited during

steady state laser ablation of the target (due to recoil pressure) should increase sublinearly with increasing light intensity:[162]

$$P_{SH} \sim I_i^{2/3}. \quad (3.57)$$

A very simple explanation of this relation stems from the idea that the energy flux proportional to I_i carried away by the shock wave depends on the cube of the velocity v_{SH} of the shock wave ($I_i \sim \rho v_{SH}^3$), whereas the pressure is proportional to the square of the velocity ($P_{SH} \sim \rho v_{SH}^2$). In strong shock waves an extreme compression of the material is attained, so that the density ρ is independent of I_i and the relation (3.57) is satisfied. A detailed calculation of the hydrodynamics of the expansion of the material indicates also that the pressure at the ablation front (and, corresponding, the pressure in the shock wave) depends on the photon energy, the mean mass number A of the nuclei of the target, and the mean charge per ion $\langle z \rangle$:[162]

$$P_{SH} \sim 1\ \text{Gbar} \cdot \left(\frac{A}{2\langle z \rangle}\right)^{1/3} \left(\frac{h\nu_L}{1\ \text{eV}}\right)^{2/3} \left(\frac{I_i}{10^{17}\ \text{W/cm}^2}\right)^{2/3}. \quad (3.58)$$

At the same time the light pressure (3.54) increases linearly with the intensity of the irradiation. Taking into account that $A/2\langle z \rangle \gtrsim 1$, we can formally estimate for that for a photon energy $h\nu_L \gtrsim 1$ eV the light pressure must predominate over recoil pressure when $I_i \gtrsim 3 \times 10^{21}$ W/cm^2. However, such high intensities of optical exposure are not yet attainable under laboratory conditions. The reasons for such a pessimistic estimate are the strong absorption of light due to the formation of a plasma layer having the critical concentration of electrons as a result of steady-state ablation. In the next section of the review it will be shown that in the process of subpicosecond laser pulses the plasma being formed cannot expand. The nonlinear increase in the reflection coefficient in the hot plasma of solid-like density can lead to predominance of light pressure, even at light intensities $I_i \gtrsim 3 \times 10^{16}$ W/cm^2.

However, before we take up this question, we shall present a number of references to studies on other destructive mechanisms of laser generation of sound. As far as we know, acoustic waves excited in the evaporation and boiling of materials have so far been observed experimentally only in liquids (see, e.g., Refs. 163–165). Changes in the pulsed optoacoustic signals caused by the melting of the surface of semiconductors have been detected in Refs. 166–168. References 169–172 were devoted to the theoretical study of this effect. Reference 109 discussed how the possibility of the motion of the melt-crystal bondary with near-sonic velocities would affect the profile of acoustic pulses. Since picosecond experiments of this type have not yet been performed, we shall not take up this question in greater detail. We only point out that the changes in the optoacoustic signal during melting the surface involve both the change in the density of the material upon melting and simply the change in the thermoelastic contribution due to the change in the reflection (and absorption) coefficients of light (e.g., because of formation of a highly reflective phase near the surface). Of course, under fine control of the parameter of the optical exposure, melting and recrystallization may not cause irreversible changes in the surface; here the mechanism of sound generation is nondestructive. Excitation of ultrasound in the laser thermo initiation of a metal-semiconductor phase transition has been observed in the experiment of Ref. 173. The relative changes in the density of matter in this phase transition, just as in melting, can reach tens of percent.

4. NEW DEVELOPMENTS IN THE OPTOACOUSTIC TECHNIQUE OF GENERATING SHOCK WAVES. EXCITATION OF STRONG SHOCK PULSES WITH HIGH-POWER ULTRASHORT LASER PULSES

The interest in the possibility of laser excitation of ultrashort shock pulses involves the fact that one can use this technique not only for diagnostics of materials, but also, as is especially important, for active intervention in physical processes and initiation of physical processes in picosecond times. We have already mentioned the influence of shock waves on the processes of spontaneous and stimulated emission of light,[56–61] which is actively used for their optical measurement. Among other manifestations of the strong compression of matter in a shock wave, we list the possibility of ionization by pressure,[174] generation of metastable metallic phases in semiconductors (at $P \gtrsim 150$ kbar in Si[175]), and amorphization.[176] One observes the formation of point defects in regions of large deformation gradients,[177] whereas the large pressures in a shock wave and the associated adiabatic heating can facilitate the annealing of defects; shock waves can initiate both melting[178] and crystallization.[179] The development of laser generators of ultrashort shock pulses would enable the study of such processes with picosecond time resolution.

The problem of shortening the duration of shock pulses has not been posed in the experimental studies known to the present authors. However, as is implied by the results of Refs. 51 and 66, the duration of compression pulses at distances no greater than several tens of micrometers does not exceed a few nanoseconds. It is also important that the duration of the front of the shock wave measured experimentally (with a pressure $P_{SH} \sim 30$ Mbar) proves to be in the picosecond range ($\lesssim 10$ ps[63]). Thus, for generating ultrashort shock pulses, additional fast decay of the pressure behind the front of the shock wave is of fundamental significance.

In this section of the review we discuss a number of features of the excitation of shock pulses in the interaction of femtosecond laser fields with matter. It is shown in Sec. 4.1 that the shaping of the shock pulse occurs during nonsteady-state electronic heat conduction in the laser plasma, which has a solid-like density. The characteristic duration of shock pulses is of the order of the time of detachment of the shock wave from the front of the thermal wave, and is determined by the rate of energy transfer from the electrons to the ions as they collide. Therefore, to shorten shock pulses it is advantageous to use targets made of heavy elements, which allow multiple ionization of the atoms with the least energy expenditure, which leads to an increased frequency of electron-ion collisions.

Section 4.2 discusses how the characteristics of the excited shock pulses depend on the fact that the motion of the plasma can be insignificant during the ultrashort optical irradiation. In this situation a layer having the critical plasma concentration (which strongly absorbs the laser radiation) cannot form. Nonetheless, the increase in the light reflection coefficient with increasing intensity can lead to a decrease in

the efficiency of conversion of the laser pulses into shock pulses.

The decrease in the efficiency of ultrafast conversion of laser energy into the thermokinetic pressure of the electron gas with increasing intensity of the optical irradition is especially severe for targets made of light elements, when even with total ionization the charge of the ions does not exceed several units. It is shown in Sec. 4.3. that in such a situation, owing to the sharp decline in the rate of electron-ion collisions with increasing electron temperature, it is possible to indent the surface of the target under the action of light pressure, which increases proportionally to the intensity of the laser light.

4.1. Shaping of shock pulses in the process of retardation of the fast nonlinear electronic heat conduction

In this section we shall discuss the following feature of laser ablation of targets that is specific to ultrashort laser pulses of duration $\tau_L \lesssim 10$ ps: for certain parameters of the radiation, the laser plasma does not expand during the light absorption. The ablation of the target material itself occurs at late times, after the ultrashort laser pulse has ceased. However, at times $t \lesssim \tau_L$, a hot plasma with solid-like density is formed. Measurements of the light reflection coefficient[11] favor such a scheme of non-steady-state ablation under subpicosecond exposures, as do the results of the theoretical estimates presented below.[180]

The absorption of laser radiation initiates the heating of the target, plasma formation, ablation of material, and formation of shock waves. For ultrashort laser pulses, these processes can be separated in time, since at short times $0 \lesssim t \lesssim \tau_L$ during the process and after completion of the laser pulse, the heat wave propagates into the interior of the material with a velocity greater than the velocity of the shock wave (or the velocity of expansion of the target material). That this statement is valid for times shorter than the time of elastic electron-ion collisions ν_{ei}^{-1} follows from a comparison of the thermal velocity of the electrons $v_e \sim (T_e/m_e)^{1/2}$, which determines the ballistic heat conduction, and the characteristic velocity of hydrodynamic perturbations in the two-temperature plasma $v_S \sim (\langle z\rangle T_e/m_i)^{1/2}$ ($m_{e,i}$ are the masses of the electrons and ions). At times exceeding the reciprocal of the rate of electron-ion collisions

$$t \gtrsim \nu_{ei}^{-1}, \tag{4.1}$$

the heat conduction is of diffusion type. Here the velocity of propagation of heat falls off with time faster than does the velocity of propagation of the shock wave. Therefore there is a characteristic time τ_0 when the shock wave overtakes the front of the heat wave

$$z_T(\tau_0) \sim z_{SH}(\tau_0). \tag{4.2}$$

Here z_T is the depth of penetration of heat (the coordinate of the front of the heat wave), while z_{SH} is the coordinate of the shock wave front. At the time τ_0 the formation of the shock pulse is completed.[178] We note that the same hypothesis was used in analyzing the possibilities of laser modeling of a high-velocity shock.[181] The process of gradual "growing" of the shock wave out of the thermal wave and its detachment with separation from the irradiated surface has been observed in Ref. 66.

Since the velocity of expansion into empty space by the mass of the material in which the shock pulse is formed coincides in order of magnitude with the velocity of sound,[178] heat conduction at times $t \lesssim \tau_0$ can be considered to occur in a stationary medium. In the second stage ($t \gtrsim \tau_0$) the issue is the propagation of the shock pulse. Here it is important that one can neglect heat conduction, since it is slow in comparison with the hydrodynamic motion (the shock wave breaks away from the heat wave). The parameters of the excited shock pulse are determined by the conditions at the instant of detachment $t \sim \tau_0$: the condition of detachment (4.2) and the condition of equality of the characteristic pressure in the shock pulse to the pressure P_e of the electron gas:

$$P_e(\tau_0) \sim \langle z\rangle n_i T_e \sim \rho_0 v_{SH}^2 \sim P_{SH}(\tau_0). \tag{4.3}$$

where $\langle z\rangle n_i \sim n_e$, $n_{e,i}$ is the concentration of electrons and ions in the plasma.

Under the conditions of interest to us of high-power laser exposures, the principal role in energy transport is played by fast nonlinear electronic heat conduction.[178,181] An important factor that determines the rate of energy transport by electrons is the frequency of elastic electron-ion collisions:

$$\nu_{ei} \sim \frac{1}{3}\frac{Q_0^4 n_i \langle z\rangle^2}{m_e^{1/2} T_e^{3/2}}\Lambda \sim \frac{Q_0^4 n_i \langle z\rangle^2}{m_e^{1/2} T_e^{3/2}}, \tag{4.4}$$

where Λ is the Coulomb logarithm. It determines the diffusion coefficient of the electrons:

$$D_e \sim v_e^2 \nu_{ei}^{-1} \sim T_e^{5/2}/Q_0^4 n_i m_e^{1/2} \langle z\rangle^2. \tag{4.5}$$

Correspondingly, for the characteristic energy density W_V at the time

$$t \gtrsim \tau_L \tag{4.6}$$

the estimate

$$W_V \sim W_S^a/z_T(t) \sim W_S^a/(D_e t)^{1/2} \tag{4.7}$$

is valid; here W_S^a is the surface density of absorbed laser energy, and we asume that the heat conduction length exceeds the depth l_p of penetration of light:

$$z_T(t) \sim (D_e t)^{1/2} \gtrsim l_p \equiv \alpha^{-1}. \tag{4.8}$$

In our order-of-magnitude estimates, assuming that $\langle z\rangle \gtrsim 1$, we shall neglect the thermal energy of the ions and the energy spent in ionization as compared with the thermal energy of the electrons

$$W_V \sim \frac{3}{2}(n_e T_e + n_i T_i) + n_i \sum_{\langle z\rangle'=1}^{\langle z\rangle} i(\langle z\rangle') \sim \frac{3}{2}\langle z\rangle n_i T_e. \tag{4.9}$$

Here $i(\langle z\rangle)$ is the energy necessary to remove an electron from an ion having the charge $\langle z\rangle - 1$. In this approximation in describing the process of heat conduction we no longer need to take into account the energy exchange between electrons and ions.[182]

Expressions (4.5), (4.7), and (4.9) give the following description of the process of electronic heat conduction:

$$T_e \sim m_e^{1/9} Q_0^{8/9} n_i^{-2/9} t^{-2/9} (W_S^a)^{4/9}, \tag{4.10}$$

$$z_T \sim m_e^{-1/9} Q_0^{-8/9} n_i^{-7/9} \langle z\rangle^{-1} t^{2/9} (W_S^a)^{5/9}. \quad (4.11)$$

We shall assume that ionization ceases when the kinetic energy of a free electron becomes insufficient to eject the next bound electron:

$$i(\langle z\rangle) \sim T_e. \quad (4.12)$$

We note that the approximate relation (4.12) is also implied by the Saha equations[178] for $T_e \gtrsim i(2)$.[178] We shall model the $i(\langle z\rangle)$ relationship with the power law

$$i(\langle z\rangle) \sim i_0 \langle z\rangle^{1/\Delta}, \quad (4.13)$$

where physically, the quantity i_0 is of the order of magnitude of the ionization energy of the neutral atom, and Δ is a free parameter. In the model of hydrogen-like ions we have $\Delta = 1/2$. Relations (4.12) and (4.13) determine the dependence of the average ion charge on the electron temperature:

$$\langle z\rangle \sim (T_e/i_0)^{\Delta}. \quad (4.14)$$

To determine the instant τ_0 of detachment of the shock wave from the heat wave, the characteristic spatial dimension of the region of generation $z_0 \sim z_T(\tau_0) \sim v_{SH}(\tau_0)\tau_0$, and the pressure at the front of the shock pulse at the instant of detachment $P_0 \sim P_{SH}(\tau_0)$, it suffices to substitute (4.10), (4.11), and (4.13) into the system of equations (4.2) and (4.3) and solve it:

$$\tau_0 \sim (W_S^a)^{(1-2\Delta)/(2-\Delta)} \sim 50 n_i^{-2} \rho_0 i_0^{3/2} \text{ ps}, \quad (4.15)$$

$$z_0 \sim (W_S^a)^{(4-5\Delta)/3(2-\Delta)} \sim n_i^{-4/3} \rho_0^{1/3} i_0 (W_S^a)^{1/3} \ \mu\text{m}, \quad (4.16)$$

$$P_0 \sim (W_S^a)^{2(1+\Delta)/3(2-\Delta)} \sim 10 \rho_0^{-1/3} n_i^{4/3} i_0^{-1} (W_S^a)^{2/3} \text{ Mbar}. \quad (4.17)$$

The first parts of expressions (4.15)–(4.17) show only how the physical quantities vary with increasing absorbed laser energy; for arbitrary values of the parameter Δ. The second parts give numerical estimates, but only in the special case $\Delta = 1/2$. Here and below the physical parameters are normalized to the following characteristic values: n_i—to 5×10^{22} cm^{-3}, ρ—to 3 g/cm^3, i_0—to 10 eV, and W_S^a—to 100 J/cm^2. The system of estimates (4.15)–(4.17) is valid for

$$\tau_L \lesssim \tau_0. \quad (4.18)$$

We note that the analysis of Eqs. (4.2) and (4.3) also reveals the relation between the characteristic time τ_0 and the frequency ν_{ei}:

$$\tau_0 \sim \frac{1}{\langle z\rangle} \frac{m_i}{m_e} \nu_{el}^{-1} \sim \tau_{ei}.$$

Thus, the detachment time of the shock pulse from the heat wave is of the order of the time τ_{ei} of energy transfer from the electron to the ion subsystem in elastic collisions.[181]

According to (4.15), the condition $\Delta = 1/2$ chosen for numerical estimates is critical in the sense that τ_0 does not depend on the energy W_S^a of the absorbed radiation. For this case let us indicate the limits of applicability of the system of estimates (4.15)–(4.17) for parameter values coinciding with the characteristic values.[180] First of all, the duration of the laser action, in view of (4.15) and (4.18), must not exceed tens of picoseconds ($\tau_L \lesssim 50$ ps). The lower bound on the magnitude of the energy of the laser exposure W_S^a is imposed by our assumption that the electronic component of the plasma is nondegenerate: $W_S^a \gtrsim 1$ J/cm^2. For such energy densities the inequality (4.8) is satisfied not only for times $t \sim \tau_0$ in (4.16), but also for times $t \sim \tau_L \gtrsim 100$ fs. Therefore we can use expression (4.10) to estimate the maximum electron temperature, so as to determine the upper bound of attainable energies W_S^a by using the inequality

$$T_e(\tau_L) \gtrsim W_V^{osc}. \quad (4.19)$$

Here W_V^{osc} is the average of the kinetic energy of over a period oscillation of the electrons in the laser field:

$$W_V^{osc} \sim \frac{Q_0^2}{4\pi m_e \nu_L^2} \frac{I_L}{c_L} \sim 75 (h\nu_L)^{-2} \tau_L^{-1} W_S^a \text{ eV}. \quad (4.20)$$

In the numerical estimate $h\nu_L$ is normalized to 1 eV, and τ_L to 100 fs. When (4.19) fails, the estimate (4.4) becomes incorrect, the process of heat conduction changes, and the generation of the shock pulse can predominantly involve not the action of the thermokinetic forces (the pressure of the randomly moving electrons), but the action of the nonlinear force of the electromagnetic field.[183] In view of relations (4.10) and (4.20), the inequality (4.19) is satisfied when $W_S^a \lesssim 10^5$ J/cm^2 for femtosecond pulses ($\tau_L \sim 100$ fs) of short-wavelength radiation ($h\nu_L \sim 4$ eV).

In view of Eq. (4.17), for attainable values of the surface density of absorbed energy $W_S^a \sim 1$–10^5 J/cm^2 (for $\tau_L \sim 100$ fs the corresponding range of light intensities is $I_a \sim 10^{13}$–10^{18} W/cm^2), shock pulses are obtained with pressures $P_0 \sim 0.5$ Mbar–1 Gbar. In the terminology used in Ref. 178, in solids shock waves with pressures $P_{SH} > 10$ Mbar are termed strong. For a qualitative description of strong shock pulses, one can treat the solid as being an electron-nuclear gas with an adiabatic index $\gamma = 5/3$ (Ref. 178) and use the existence of self-similar solutions of the gas-dynamic equations describing the shock wave formed in the expansion of the gas after the instantaneous energy release at the surface. Thus, one can use the relations (4.15)–(4.17), which were derived by using the conditions (4.2) and (4.3), for joining the self-similar solutions of the problems of nonlinear heat conduction and gas dynamics.

If r_0 is the characteristic dimension of the region of focusing of the light, then we can use the one-dimensional equations of gas dynamics to describe the shock pulse at distances less than r_0^2/z_0. The only length scale in the motion initiated by the instantaneous laser action δ-localized at the boundary with the vacuum is the variable coordinate of the shock pulse front:

$$z_{SH} = Bt^{\beta}, \quad (4.21)$$

where β is the self-similarity index, and the parameter B is determined by the characteristics of the exposure. Since the characteristic pressure at the front of the shock pulse can be represented in the following form from dimensional considerations

$$P_{SH} \sim \rho(\dot{z}_{SH})^2 \sim \rho B^2 \beta^2 t^{2\beta-2}, \quad (4.22)$$

we can determine the value of the parameter B from the conditions (4.15)–(4.17), which actually are the initial conditions for (4.21) and (4.22): $z_{SH}(\tau_0) \sim z_0$, $P_{SH}(\tau_0) \sim P_0$.

The value of the self-similarity index $\beta \approx 0.61$ was obtained for $\gamma = 5/3$ by numerical solution of the equations of hydrodynamics describing the self-similar motion.[178] Consequently, we can write the dependence of the coordinate of the front and the pressure at the front of the shock pulse at the pont of observation on the absorbed energy density (in the case $\Delta = 1/2$) in the form[180]

$$z_{SH} \sim (W_S^a)^{1/3}, \quad P_{SH} \sim (W_S^a)^{1.1}. \tag{4.23}$$

The superlinear increase in the pressure is due to the approach of the region of formation of the shock front to the measuring region.

In Ref. 180 an approximate relationship between the duration τ_{SH} of the shock pulse at half-height and the time of arrival t_{SH} of the front of the shock pulse at the point of observation was obtained by a linear approximation to the profile of the shock pulse (in the case of interest to us with $\gamma = 5/3, \beta = 0.61$):

$$\tau_{SH} \approx 0.2 t_{SH}. \tag{4.24}$$

In view of relation (4.24), the duration of a strong shock pulse increases proportionally to its time of propagation. Since the emergence of the hydrodynamic motion into a self-similar regime occurs in a time considerably exceeding the time of generation of the shock pulse,[178] then estimate (4.24) is correct when $t_{SH} \gg \tau_0$. Consequently, in any case the duration of the shock pulse τ_{SH} exceeds its generation time τ_0. Thus, the propagation of a shock pulse under conditions of expansion of matter into the vacuum cannot shorten its duration (which, of course, was of the order of τ_0 at the instant of cessation of generation).

Thus a simple shortening of the duration of the laser exposure does not suffice for a corresponding shortening of the shock pulse. When $\tau_L < \tau_0$ the duration of the shock pulse in the model being discussed does not depend on τ_L, but is determined by the characteristics of the target employed. According to relations (4.15), for targets with $0 \leqslant \Delta \leqslant (1.2)$, τ_{SH} exceeds tens of picoseconds. In this case the width τ_f of the front of the shock wave does not affect the total duration of the shock pulse, since, according to the results of Ref. 63, $\tau_f < 10$ ps. To obtain shorter shock pulses, targets are required with $\Delta > 1/2$, in which the time of electron-ion energy exchange $\tau_{ei} \sim \tau_0$ is shortened when the absorbed energy density is decreased. Thus, for generating ultrashort shock pulses, targets are preferable in which the energy used in multiple ionization of the atoms is minimal. We note that these conclusions are correlated with the results of Ref. 182, in which it was proposed to add heavy elements to a target made of light atoms for fast cooling of the electrons and retardation of the process of heat conduction.

4.2. Influence of the laser-energy dependence of the absorptivity of the target on the efficiency of conversion of laser pulses into shock pulses

The surface density of absorbed laser energy W_S^a figures in all the relations presented above. Therefore, to use them in practice, we must also relate W_S^a with the surface energy density of the incident radiation, W_S^i. In a number of cases one can obtain the relation optimal from the standpoint of ablation-induced generation of strong shock pulses: $W_S^a \approx W_S^i$. First, according to the results of Ref. 184, to increase the energy contribution into the target one can apply a periodic system of submicrometer grooves on the absorbing surface. Second, the absorption of the greater part of the incident radiation can occur upon exposure to rather long laser pulses $\tau_L \gtrsim 10$–100 ps.[62–64,66] In this situation a plasma layer can be formed with the critical concentration in which strong light absorption can occur, even within the time of the optical exposure.

However, if we do not take special measures,[184] then the dependence of the light reflection coefficient on the parameters of the plasma for laser exposures $\tau_L \lesssim 10$ ps can lead to a substantial decrease in the absorptivity of the target. For example, in Ref. 11 in exposures to silicon of radiation with the parameters $\tau_L \sim 160$ fs, $h\nu_L \sim 4$ eV, an approximately threefold diminution was observed in the absorptivity $1 - R$ for a change of W_S^i for 1 J/cm^2 to 10^2 J/cm^2. The results of Ref. 19 also indicate a tendency to increase in the reflection coefficient of aluminum for radiation with $h\nu_L \sim 4$ eV, $\tau_L \sim 400$ fs at $W_S^i \gtrsim 50$ J/cm^2.

The dependence of the light reflection coefficient on W_S^a leads to a nonlinearity in the relation between the absorbed W_S^a and the incident W_S^i radiation energies

$$W_S^a = (1 - R(W_S^a))W_S^i. \tag{4.25}$$

In the Drude model of the dielectric permittivity of a plasma, the Fresnel reflection coefficient depends on the relative magnitude of three frequencies: ν_{ei} (4.4), $2\pi\nu_L$, and the electron plasma frequency ω_p. By using relation (4.14) and the previous normalizations of the physical quantitites, we can estimate:

$$\nu_{ei} \approx 1{,}5 \cdot 10^{15} n_i i_0^{-2\Delta} T_e^{2\Delta-(3/2)}\ \text{s}^{-1}, \tag{4.26}$$

$$2\pi\nu_L \approx 1{,}5 \cdot 10^{15} h\nu_L\ \text{c}^{-1}, \tag{4.27}$$

$$\omega_p \equiv \left(\frac{4\pi n_e Q_0^2}{m_e}\right)^{1/2} \approx 1{,}5 \cdot 10^{16} n_i^{1/2} i_0^{-\Delta/2} T_e^{\Delta/2}\ \text{c}^{-1}. \tag{4.28}$$

In the case

$$\nu_{ei} < 2\pi\nu_L < \omega_p \tag{4.29}$$

the Drude model gives the following expressions for the absorption coefficient of the light α and the absorptivity

$$\alpha \approx 2\frac{\omega_p}{c_L} \sim 10^6 n_i^{1/2} i_0^{-\Delta/2} T_e^{\Delta/2}\ \text{см}^{-1}, \tag{4.30}$$

$$1 - R \approx 2\frac{\nu_{ei}}{\omega_p} \sim 0.2 n_i^{1/2} i_0^{-3\Delta/2} T_e^{-3(1-\Delta)/2}. \tag{4.31}$$

According to (4.31) the reflection coefficient varies during optical exposure because of the variation of the electron temperature. For estimates we shall use as a characteristic value of $1 - R$ the value obtained for $t \sim \tau_L$. Then Eqs. (4.10), (4.25), and (4.31) allow us to determine the relation between W_S^a and W_S^i,

$$W_S^a \sim (W_S^i)^{3/(5-2\Delta)}.$$

The exponents in the first parts of the description (4.15)–(4.17) are altered correspondingly. Of course, the numerical estimate of τ_0 for the special case $\Delta = 1/2$ in (4.15) is not

altered. However, the other parameters of the shock pulse take on the following values:

$$z_0 \sim 0{,}5\rho^{1/3}n_i^{-7/6}i_0^{3/16}\tau_L^{1/24}(W_S^i)^{1/4}\ \mu\text{m},$$
$$P_0 \sim 1{,}5\rho^{-1/3}n_i^{5/3}i_0^{-11/8}\tau_L^{1/12}(W_S^i)^{1/2}\ \text{Mbar}. \tag{4.32}$$

Just as in the general case, an increase in R leads to retardation of the rate of increase of the parameters of the shock pulse with the laser power, as compared with (4.16) and (4.17). The relation $P_{SH} = P_{SH}(W_S^i)$ at the point of observation [like (4.23)] can become sublinear.

As before, the region of applicability of the obtained results is bounded from below by the condition of nondegeneracy of the plasma: $W_S^i \gtrsim 1$ J/cm^2. When this condition is fulfilled, the source of heating of the electrons is a surface source $z_T(\tau_L) > \alpha^{-1}$ in (4.30), while the estimate of the frequencies (4.26)–(4.28) indicates the fulfillment of the condition (4.29) for radiation with $h\nu_L \sim 4$ eV. The upper bound on the attainable energy densities imposes a condition of applicability of the Fresnel formulas for the light relfection coefficient. They can be used if the width δ of the transition region from the vacuum to a plasma of solid-like density does not exceed the penetration depth of the radiation $l_p \sim \alpha^{-1}$. The maximum velocity v_a of outflow of gas into the vacuum, as is known,[178] is of the order of the velocity of the ion-sonic wave

$$v_a = \frac{2}{\gamma - 1}v_S = \frac{2}{\gamma - 1}\left(\frac{\langle z\rangle T_e}{m_i}\right)^{1/2}.$$

Here an estimate of the fulfillment of the inequality $\delta \sim v_a\tau_L < \alpha^{-1}$ leads to the condition $W_S^i \lesssim 10^3$ J/cm^2. Additional tests show that, with this restriction on the energy density of the laser pulse, condition (4.19) is satisfied, while the distance traveled by an electron in a period of oscillations in the light field is smaller than the depth of penetration of the light. The latter condition together with (4.19) indicates that in this case for the near ultraviolet ($h\nu_L \approx 4$ eV) a certain type of skin effect is realized in the infrared region of the spectrum.[185]

Thus, for laser pulses of length $\tau_L \sim 100$ ps with $\Delta = 1/2$, the set of estimates (4.32) is applicable at intensities of optical exposure in the range $W_S^i \sim 10^{13}$–10^{16} W/cm^2. According to (4.32), generation of shock pulses with pressures $P_{SH} \sim 0.1$–5 Mbar can occur under such conditions. Further increase in intensity in this case must give rise to a smooth transition to the regime (4.15)–(4.17) discussed earlier, with an increase in the absorptivity of the plasma as it expands.

4.3. The possibility of predominance of light pressure over thermokinetic pressure of electrons

Comparison of the pressures in (4.17) and (4.32) with the light pressure of (3.53) shows that, under real experimental conditions in the regime $\Delta = (1/2)$, P_L is negligibly small. This is because the transition to the regime of strong absorption of light occurs before P_L of (3.54) and P_0 of (4.32) can become equal when the light intensity is increased. The scenario of development of events with increasing W_S^i can be different in the case of targets made of light elements, when the mean charge of the ions $\langle z\rangle$ is constant (independent of T_e), because of the total ionization of the atoms, even at moderate intensities. The transition to this regime occurs by the formal replacement in (4.12) and the subsequent estimates $i_0 \to (A/2)^{-1/\Delta}$, $\Delta \to 0$. Here we have $\langle z\rangle \sim A/2 \sim$ const. Here a situation can occur in which the expansion of the plasma opposes the light pressure.

Comparison of the thermokinetic pressure of the electrons

$$P_e \sim \langle z\rangle n_i T_e \sim 3{,}2n_i\langle z\rangle^{7/5}\tau_L^{-2/15}(I_S^A)^{4/15}\ \text{Mbar}$$

with the light pressure (3.54), for which the following estimate holds under conditions of strong reflection ($R \approx 1$):

$$P_L \approx 0{,}3\tau_L^{-1}W_S^i\ \text{Mbar},$$

shows that the latter predominates when

$$W_S^i \gtrsim (W_S^i)_{cr}^{(1)} \sim 2{,}5\cdot 10^3 n_i^{15/11}\langle z\rangle^{21/11}\tau_L^{13/11}\ \text{J/cm}^2. \tag{4.33}$$

At energies exceeding a critical energy $(W_S^i)_{cr}^{(1)}$, the surface of the target is indented. Beginning at $P_L - P_e \gtrsim 10$ Mbar, for a qualitative description of the motion of the surface we can use the assumption of the extreme compression of the near-surface electron-nuclear gas ($n_i^{SH} \approx 4n_i$, for $\gamma = 5/3$). Then the front of the shock wave penetrates into the target with a velocity $v_{SH} = 2(P_L/3\rho)^{1/2}$, while the surface moves with a velocity $v_0 \sim (3/4)v_{SH}$.

The extreme compression must be taken into account in determining the upper bound of the energy contributions at which the stated regime can be realized. The upper bound on W_S^i can also be associated with the applicability of the Fresnel formulas for calculating the reflection of light. In the present case the broadening of the δ-jump in the dielectric permittivity at the vacuum-plasma boundary can involve the spatial separation of the electrons and ions under the action of the light pressure

$$\delta \sim \frac{1}{\langle z\rangle Q_0(4n_i)}\left(\frac{P_L}{2\pi}\right)^{1/2}.$$

The inequality $\delta \leqslant l_p$ is satisfied when

$$W_S^i \lesssim (W_S^i)_{cr}^{(2)} \sim 4{,}7\cdot 10^6 n_i\langle z\rangle\tau_L\ \text{J/cm}^2. \tag{4.34}$$

Finally, when the relation

$$W_S^i \lesssim (W_S^i)_{cr}^{(3)} \sim 2{,}3\cdot 10^3 n_i^{-15/4}\langle z\rangle^{-21/4}\tau_L^{1/2}(h\nu_L)^{15/2}\ \text{J/cm}^2. \tag{4.35}$$

breaks down, then all of the estimates must be modified with account taken of the fact that the skin effect in the infrared region of the spectrum will not occur, but rather the anomalous skin effect (since the inequality $l_p < v_e/2\pi\nu_L$ breaks down).

Comparison of the ciritical energies $(W_S^i)_{cr}$, (4.34) and (4.35), clearly indicates the advantage of using short-wavelength lasers to expand the possible range of radiation energies in which the surface is indented. When $h\nu_L \sim 4$ eV, $\tau_L \sim 100$ fs, $n_i \sim 5\times 10^{22}$ cm^3, the proposed system of estimates indicates that the condition $P_L > P_e$ can be satisfied in the energy range $W_S^i \sim 2\times 10^4$–2.5×10^5 J/cm^2. Indentation of the surface can occur also at higher energies of irradiation. However, to describe this situation, one must use formulas for α and $(1 - R)$ differing from (4.30) and (4.31) (the

anomalous skin effect[185,186]). Even in this energy range (the corresponding intensities of laser exposure are $I_i \sim 2\times 10^{17}$–2.5×10^{18} W/cm^2), the light pressure on the surface approaches gigabar values ($P_L \sim 60$–800 Mbar). At such pressures, in the course of the characteristic time of laser exposure ($\tau_L \sim 100$ fs), the entire region of light penetration is involved in shock compression.

The estimates show that, in the regime being studied $P_L > P_e$ [$I_i \sim (2$–$25)\times 10^{17}$ W/cm^2], the characteristic frequencies of electron-ion collisions ($\nu_{ei} \sim 10^{14}$–3.5×10^{13} s^{-1}) already lie at the boundary of the condition (4.1) for diffusion heat conduction (for $\tau_L \sim 100$ fs). The flux of heat introduced into the target $W_S^a \sim (1-R)W_S^i \sim (3\times 10^{15}$–$1.5\times 10^{16})$ W/cm^2 is close to that at which one expects a limitation on heat conduction under conditions of ablation of the targets by long pulses.[162,186] Finally, as additional estimates show, when $P_L > P_e$ condition (4.19) can begin to break down. Correspondingly, the characteristic vibrational velocities of an electron begin to exceed the thermal velocities. Then plasma instability can develop,[186,187] accompanied by an increase in the effective rate of electron scattering. This process can impede the compression of the plasma by light pressure and completely rule out the possibility of indentation of the surface. On the whole we can state that the experimental studies of optoacoustic conversion under the action of high-power femtosecond short-wavelength radiation can yield very interesting information on the physics of ultrafast electron-ion energy exhange in a plasma. The further development of theoretical models should first of all include taking account of the possible nonequilibrium character of the ionized composition of the plasma,[182] analysis of the possibility of development of plasma instabilities in the presence of the high-power electromagnetic wave (e.g., at $T_e \sim W_V^{osc}$ [Ref. 186]), and should also include the description of the structure of ultrashort shock pulses of durations $\tau_{SH} \lesssim 1$–10 ps.

5. THE PHYSICAL FACTORS AFFECTING THE DURATION OF RECORDED OPTOACOUSTIC PULSES

The entire preceding treatment has been devoted to discussing the topic mentioned in the heading of this section. Here we shall take up in addition the transformation of the spectra (and profiles) of ultrashort acoustic pulses during their propagation (outside the region of generation). We shall analyze separately the possibilities of controlling the duration of sound pulses through nonlinear changes in the acoustic sources as the intensity of laser exposure is increased.

5.1. The role of dissipation, diffraction, acoustic nonlinearity, and dispersion in the propagation of acoustic pulses

Since in Sec. 3 we were interested only in the various physical mechanisms of optoacoustic conversion, for compactness of presentation we neglected the absorption, dispersion, and non-one-dimensionality of the acoustic waves and the nonlinear effects in the crystal-lattice dynamics. Methods are known that enable one to take account of these factors, even at the stage of excitation of the acoustic pulses.[77] However, as the estimates show for most of the experimental efforts of optoacoustic conversion, there is no need of this. The characteristic distances at which the role of these accumulating effects is appreciable (i.e., the absorption length L_{ABS}, the diffraction length L_{DIF}, the dispersion length L_{DIS}, and the nonlinear length L_{NL}) usually substantially exceed the size of the region of sound generation, L_G. Of course, this directly involves the fact that in applied studies the investigators mainly try to achieve nondestructive excitation of plane waves. To do this, in particular, by controlling the focusing of the radiation, one tries to obtain localization of the sound sources in a volume in the form of a disk.

When the inequalities $L_{ABS}, L_{DIF}, L_{DIS}, L_{NL} \gg L_G$ are satisfied, the problem of determining the optoacoustic signal in the detection region (at distances $z \gg L_G$) can be solved in two stages. In the first stage the valid solutions are those derived by using the method of spectral transfer functions, Sec. 3. These solutions (3.4)–(3.6), which are valid outside the region of generation ($z > L_G$), are fixed as boundary conditions for the problem of the evolution of a quasiplanar acoustic pulse of finite amplitude in a dissipative medium. In the second stage, to describe this evolution, while neglecting anisotropy of the crystal, we can use the equation

$$\frac{\partial}{\partial\tau}\left[\frac{\partial U}{\partial z}+\frac{\varepsilon}{c_a}U\frac{\partial U}{\partial\tau}-\frac{b}{2\rho c_a^3}\frac{\partial^2 U}{\partial\tau^2}-\frac{a^2}{24c_a^3}\frac{\partial^3 U}{\partial\tau^3}\right]=\frac{c_a}{2}\Delta_\perp U. \qquad (5.1)$$

Here ε is the nonlinear acoustic parameter of the medium,[77,188,189] b is the dissipative parameter,[188,189] a is the crystal-lattice parameter,[40,190] and $\Delta_\perp$ is the Laplace operator in coordinates normal to the z axis. When dispersion is neglected ($a = 0$), Eq. (5.1) in the nonlinear theory of sound beams[189,191] is commonly called the Khokhlov–Zabolotskaya equation. In the absence of sound absorption ($b = 0$), Eq. (5.1) is also known as the Kadomtsev–Petviashvili equation. The solution of the quasilinear evolution equation (5.1) is possible in the general case only by numerical methods.[191] However, analytical results can be obtained in the case in which the spatial scales of the physcial effects being analyzed strongly differ.

If the smallest characteristic length is the dissipation length ($L_G \ll L_{ABS} \ll L_{DIF}, L_{DIS}, L_{NL}$), then (5.1) reduces to the linear parabolic equation

$$\frac{\partial U}{\partial z}-\frac{b}{2\rho c_a^3}\frac{\partial^2 U}{\partial\tau^2}=0.$$

Naturally this can be solved, e.g., by using the integral transformations (3.3). Representation of the solution in the spectral form

$$\tilde{U}(\omega, z) = \tilde{U}(\omega, z=0)\exp(-b\omega^2 z/2\rho c_a^3) \qquad (5.2)$$

shows that each spectral component falls off exponentially with distance. Physically the quantity $\alpha_a = b\omega^2/2\rho c_a^3$ is the absorption coefficient of sound of frequency ω. Absorption that increases quadratically with the frequency ($\alpha_a \sim \omega^2$) is ordinarily called high-frequency absorption, since, according to Eq. (5.2), it leads to narrowing of the spectrum of broad-band acoustic signals. In crystals other forms of dependences $\alpha_a = \alpha_a(\omega)$ (differing from $\sim\omega^2$) can also be realized, especially at low temperatures, or in the terahertz frequency region.[76]

Using the description (3.11) of the generated deformation wave $\tilde{U}(\omega, z=0)$ and the solution (5.2), we can conveniently write the profile of the signal in the detection region as

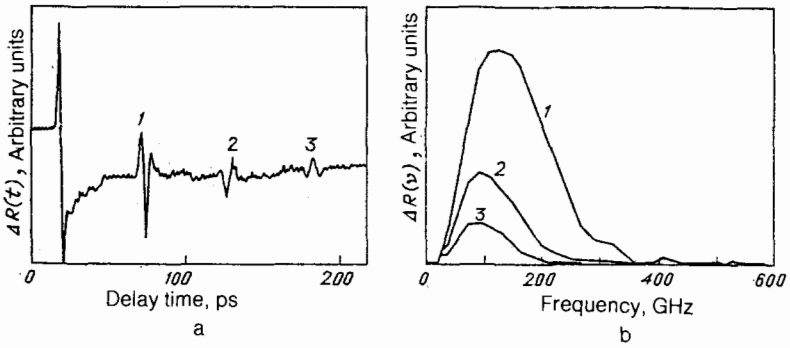

FIG. 26. Profiles (a) and spectra (b) of variations of the light reflection coefficient initiated by multiple reflections of ultrashort optoacoustic deformation pulses in an a-SiO_2 film.[193]

$$U(z, t) = (2\pi)^{-1} \int_{-\infty}^{\infty} K(\omega) I_i \tilde{f}(\omega) \exp(-\alpha_a(\omega) z - i\omega\tau) d\omega. \tag{5.3}$$

According to (5.3) we can speak of the spectral transfer function of optoacoustic conversion + propagation:

$$K(\omega, z) = K(\omega) \exp(-\alpha_a(\omega) z). \tag{5.4}$$

This depends on the distance z between the regions of generation and detection of the sound. It is clear that the narrowing of the spectrum of efficiently transferred frequencies with increasing z in Eq. (5.4) must lead to increased duration of the acoustic pulses.

Distortions of the profiles of ultrashort deformation pulses caused by absorption of acoustic waves during propagation have been observed in many experiments (e.g., Refs. 31, 33, 35, 36, 192). By measuring such changes in the spectrum of the optoacoustic signal, one can not only estimate,[33,35,36] but also measure very exactly the absorption coefficients of hypersound at room temperature. In a recent experiment[193] picosecond optoacoustic technology (see Sec. 2) was used to determine the dependence of the absorption of longitudinal sound waves at temperatures 80–300 K in the frequency band $\nu_a \sim 180$–250 GHz in a-SiO_2. Figure 26a shows the profiles of the variation of the reflection of light caused in an aluminum optoacoustic converter by the three first echo pulses arriving after re-reflection in a 1480-Å glass layer. The corresponding spectra of the echo signals are shown in Fig. 26b. The results of the measurements of the temperature-dependence of the absorption of hypersound at two frequencies ($\nu_\alpha = 201$ and 238 GHz) are shown in Fig. 27. We note that, according to the measurements[193] at room temperature, the variation in absorption in the frequency band $\nu_a \sim 30$–200 GHZ agrees within the limits of experimental error with the ω^2 law. The experiment[193] clearly demonstrates the unique potentialities of broad-band optoacoustic spectroscopy of thin films.

In view of the expression (5.3) for the dissipation length L_{ABS}, we should consider the reciprocal absorption coefficient of sound at the largest of the characteristic frequencies of the generated signal $U(\tau, z = 0)$:

$$L_{ABS} \sim 2\rho c_a^3 / b\omega_{max}^2$$

When $z \gtrsim \alpha_a^{-1}(\omega_{max})$ the exponential factor in (5.3) begins to influence the spectrum of the optoacoustic signal. At the same time, at distances exceeding the absorption length of sound having the smallest of the characteristic frequencies ($z \gtrsim \alpha_a^{-1}(\omega_{min})$), almost all the information on the details of the spectrum $\tilde{U}(\omega, z = 0)$ is lost. The profile of the deformation pulses at such distances is associated with a differential or integral transformation with the Gaussian profile

$$U(\tau, z) \sim \exp(-\rho c_a^3 \tau^2 / 2bz) \tag{5.5}$$

depending on the behavior of the spectrum $\tilde{U}(\omega, z = 0)$ at low frequencies ($\omega \to 0$). If $\tilde{U}(\omega = 0, z = 0) \neq 0$ (e.g., in the case (3.42), the profile of the optoacoustic pulse is Gaussian (5.5). If $\tilde{U}(\omega \to 0, z = 0) \sim \omega$ [e.g., in the case that (3.13) holds], the profile of the optoacoustic pulse resembles the first derivative of (5.5), etc. In all cases at distances $z > \alpha_a^{-1}(\omega_{min})$, in view of (5.5), the characteristic acoustic pulse length increases with the distance traveled as $z^{1/2}$.

If the smallest characteristic length is the diffraction length ($L_G \ll L_{DIF} \ll L_{ABS}, L_{DIS}, L_{NL}$), then Eq. (5.1) corresponds to the quasioptical approximation in diffraction theory:[189,190]

$$\frac{\partial}{\partial z} \frac{\partial U}{\partial \tau} = \frac{c_a}{2} \Delta_\perp U. \tag{5.6}$$

For a Gaussian transverse distribution of sound in the beam in the generation region we have:

$$U(\tau, z = 0, r) = U(\tau, z = 0) \exp(-r^2 / r_0^2),$$

where r is the transverse coordinate, r_0 is the radius of the beam, and we can represent the spectrum of the optoacoustic signal that satisfies Eq. (5.6) on the axis $r = 0$ in the form[77,189]

$$\tilde{U}(\omega, z, r = 0) = \frac{\tilde{U}(\omega, z = 0, r = 0)}{1 + (2ic_a z / r_0^2 \omega)}. \tag{5.7}$$

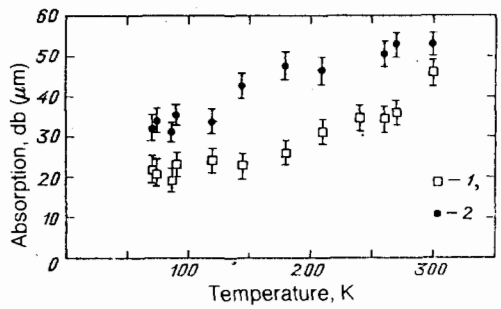

FIG. 27. Temperature dependence of the absorption of sound at the frequencies 201 GHz (*1*) and 238 GHz (*2*) measured by broadband optoacoustic spectroscopy.[193]

According to (5.7), diffraction, in contrast to high-frequency absorption (5.2), primarily influences the low-frequency components of the spectrum of the optoacoustic signal. The decrease in the low-frequency components begins appreciably to influence the spectrum (and the profile) of the deformation pulse starting at distances z of the order of

$$L_{\text{DIF}} \sim r_0^2\omega_{\min}/2c_a.$$

By virtue of Eq. (5.7), the following approximate relation is valid at distances $z \gg r_0^2\omega_{\max}/2c_a$

$$\tilde{U}(\omega, z, r=0) \approx \frac{r_0^2}{2c_a z}(-i\omega)\tilde{U}(\omega, z=0, r=0).$$

According to this result, the profile of the diffracted optoacoustic signal on the axis resembles the time derivative of the profile of the generated deformation pulse.

Naturally, the diffraction "cutoff" of the low-frequency region of the spectrum of the optoacoustic signal can shorten the duration of optoacoustic pulses as they propagate. In the following very graphic example, there are two characteristic frequencies strongly differing in amplitude in the spectrum of the original optoacoustic signal. We assume that the thermoelastic optoacoustic conversion occur in a thermally thick region of light absorption. Using Eqs. (3.11) and (3.13), we can write the deformation spectrum $\tilde{U}(\omega)$ outside the region of generation as

$$\tilde{U}(\omega) \sim \frac{-i\omega}{\omega^2+\omega_\alpha^2}\exp\left[-\left(\frac{\omega}{\omega_L}\right)^2\right]. \tag{5.8}$$

In the case $\omega_L \gg \omega_\alpha$ ($4\tau_L^{-1} \gg \alpha c_a$) the profile of a deformation pulse having the spectrum (5.8) has the characteristic bipolar form of (3.14), but the transition from the compression phase to the rarefraction phase does not occur instantaneously, but after a finite time $\sim\tau_L$ (see Fig. 18). Here the characteristic acoustic pulse length is $\tau_a(z=0) \sim (\alpha c_a)^{-1}$. Because of Eq. (5.7) the spectrum of the optoacoustic pulse at distances $z \gg r_0^2\omega_L/2c_a$ resembles the spectrum of the intensity envelope of the laser pulse

$$\tilde{U}(\omega, z) \sim -\frac{r_0^2}{2c_a z}\exp\left[-\left(\frac{\omega}{\omega_L}\right)^2\right]. \tag{5.9}$$

Consequently the diffraction of the waves can shorten the optoacoustic deformation pulse length down to $\tau_a \equiv \tau_L$. Many experimental studies of the dependence of the profiles of the diffracted optoacoustic signals on the distance traveled by the pulses and/or on the focusing conditions have been carried out, mainly in liquids (e.g., Refs. 83, 194, and 195). Figure 28 shows the profiles of optoacoustic pulses excited thermoelastically near the free surface of n-C_7H_{16} at different distances from the boundary.[83] The oscillations on the tail of the experimental profiles involve excitation of the natural vibrations of the receiving transducer. In this experiment the condition $\tau_L \ll (\alpha c_a)^{-1}$ was attained. Comparison of Figs. 28a and 28b graphically demonstrates the diffraction-induced decrease in the characteristic duration of optoacoustic pressure pulses in going from the near to the far wave field.

If the smallest characteristic length is the nonlinearity length ($L_G \ll L_{NL} \ll L_{DIF}, L_{DIS}, L_{ABS}$), then (5.1) reduces

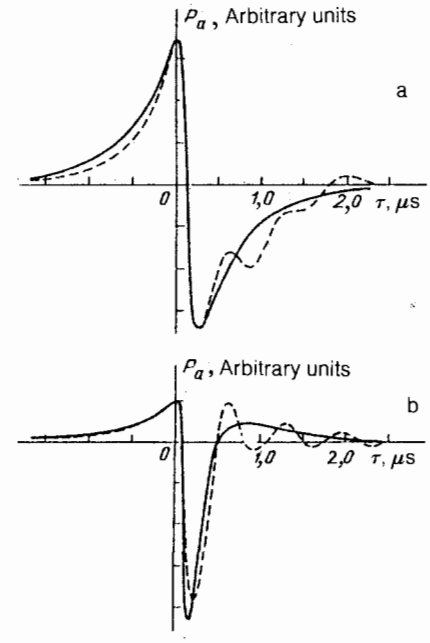

FIG. 28. Profiles of optoacoustic pressure pulses in n-heptane at distances $z = 3.4$ mm (a) and $z = 158$ mm (b) from the free surface ($z = 0$). Solid profiles—theory, dashed profiles—experiment.[83]

to the simple wave equation

$$\frac{\partial U}{\partial z} + \frac{\varepsilon}{c_a}U\frac{\partial U}{\partial \tau} = 0,$$

whose solution

$$U = U_0(\tau - \varepsilon\frac{U}{c_a}z) \tag{5.10}$$

implicity describes the relation between the deformation profile U at the distance z and its profile $U_0 \equiv U(\tau, z=0)$ at the boundary of the region of optoacoustic conversion. According to the principles of nonlinear acoustics,[77,188,189] the deformation pulses described by Eq. (5.10) are distorted because the velocity of propagation of acoustic waves depends on their amplitude. For example, in the case $\varepsilon > 0$ the compression regions ($U < 0$) propagate with velocities exceeding the velocity c_a of sound waves of infinitesimally small amplitude. Figure 29 shows the results of calculation by Eq. (5.10) of the transformation of the profile of an optoacoustic signal that at the exit from the generation region was a unipolar pulse having the characteristic pulse length $\tau_a(z=0) \sim \tau_0$. Analysis shows that the profile (the loops in

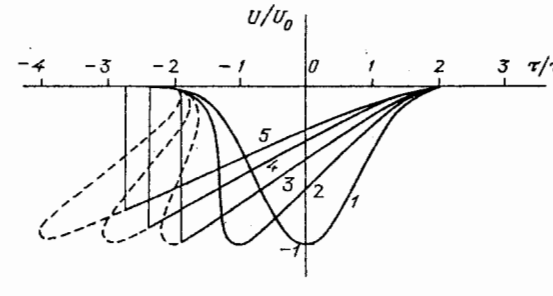

FIG. 29. Nonlinear transformation of the profile of an optoacoustic compression pulse during propagation in an ideal medium. The parameter of the profiles is the dimensionless distance to the sound-generation region: $z/L_{NL} = 0$ (*1*), 1 (*2*), 2 (*3*), 3 (*4*), and 4 (*5*).

Fig. 29) becomes non-single-valued at distances z of the order of

$$L_{\rm NL} \sim \frac{c_a}{\varepsilon}\left(\frac{dU}{d\tau}\right)^{-1} \sim \frac{c_a\tau_0}{\varepsilon U_0},$$

where U_0 is the characteristic value of the deformation in the acoustic pulse. In the case of signals of complex form we have $L_{\rm NL} \sim c_a/\varepsilon U_0\omega_{\max}$. Physically, the multivalued behavior indicates the formation of a shock front whose location is determined by the law of conservation of momentum in the acoustic pulse (the law of equal areas[189]). At distances $z \gg c_a/\varepsilon U_0\omega_{\min}$ an isolated unipolar pulse of arbitrary form inevitably becomes triangular (see Fig. 29), with its duration increasing with the traveled distance as $z^{1/2}$, while the amplitude falls off as $z^{-1/2}$.[190] The asymptotic form of multipolar signals depends strongly both on the sequential order of the compression and rarefaction phases an on the magnitude of the total momentum of the optoacoustic pulse. Thus, for example, the duration of a symmetric bipolar signal beginning with a rarefraction phase (in the case $\varepsilon > 0$) is not altered by the nonlinear effects.

Nonlinear distortions of the profiles of optoacoustic pulses resembling that depicted in Fig. 29 have been experimentally detected in liquids,[35,83,196,197] and in solids.[198] In liquids[35,192] and solids (e.g., Ref. 199), the propagation of light-induced weak shock waves with velocities exceeding that of linear sound has also been observed.

In the region of localization of a front formed in the propagation of an acoustic pulse (see Fig. 29), the role of the terms containing the higher derivatives in Eq. (5.1) becomes more important. In other words, to describe the structure of the front, we must take account of the absorption and/or dispersion of the acoustic waves. If $L_{\rm ABS} < L_{\rm DIS}$, then the evolution of the profiles of deformation pulses occurs in agreement with the Burgers equation:

$$\frac{\partial U}{\partial z} + \frac{\varepsilon}{c_a}U\frac{\partial U}{\partial \tau} - \frac{b}{2\rho c_a^3}\frac{\partial^2 U}{\partial \tau^2} = 0. \tag{5.11}$$

The width of the front is locally stabilized if the generation of harmonics by the acoustic nonlinearity is compensated by their dissipation in the high-frequency region of the spectrum. Here the second and third terms of Eq. (5.11) are comparable in magnitude in the region of the front. This condition allows us to estimate the dependence of the width of the front τ_f on the amplitude $U_0(z)$ of the shock wave: $\tau_f \sim b(2\rho c_a^2 \varepsilon U_0(z))^{-1}$. We recall that the width of the front of a shock wave in water was observed in the experiment of Ref. 50 to be reduced to $\tau_f \lesssim 20$ ps. However, we emphasize that, in the given case, as is implied by analyzing the solution (5.10) (see also Sec. 4), the enrichment of the spectrum of the deformation pulse in high-frequency components does not reduce its length during nonlinear propagation.

The condition $L_{\rm DIS} < L_{\rm ABS}$, even for ultrashort deformation pulses, can be obtained only at cryogenic temperatures (by using the maximum possible suppression of the interaction of coherent acoustic waves with the thermal vibrations of the crystal lattice). The evolution of the profile of the optoacoustic pulse is the described by the Korteweg–de Vries equation

$$\frac{\partial U}{\partial z} + \frac{\varepsilon}{c_a}U\frac{\partial U}{\partial \tau} - \frac{a^2}{24c_a^3}\frac{\partial^3 U}{\partial \tau^3} = 0. \tag{5.12}$$

Under the condition $L_{\rm NL} < L_{\rm DIS}$ this describes the detachment of solitons from the steepining front of the deformation pulse. The duration τ_s of the solitons being formed can be estimated from the order-of-magnitude equality of the nonlinear and dispersion terms in Eq. (5.12)

$$\tau_s \sim \frac{a}{2c_a}\left(\frac{1}{6\varepsilon U_0(z)}\right)^{1/2}, \tag{5.13}$$

where $U_0(z)$ is the amplitude of deformation at the front of the shock wave at the instant of detachment of the soliton. According to (5.13)[40,200] solitons with close to the least possible durations in discrete structures ($\tau_s \sim 2a/c_a$) can be obtained at levels of deformation $U_0(z) \gtrsim 10^{-2}\varepsilon^{-1}$ actually attainable in laser experiments (the range of typical values of the acoustic nonlinearity parameter is $1 \lesssim \varepsilon \lesssim 20$).

To conclude this section, we shall discuss another factor that influences the duration of optoacoustic pulses, one that does not accumulate during propagation. Also, this factor does not involve the transformation of sound sources distributed in the volume (Sec. 5.2). The subject is the affect on the form of the profiles of optoacoustic pulses caused by roughness of the surface being irradiated. The features of the thermoelastic optoacoustic conversion in the case of agitation of the surface of a liquid has been analyzed in detail in a series of theoretical studies.[201–206] In the case of laser generation of ultrashort sound pulses it seems natural first of all to estimate the effect of coarse-scale roughness of the surface on this process. According to the results obtained in Ref. 203, when the roughness scale (the characteristic radius of curvature of the roughness[205]) is large in comparison with the spatial extent of the acoustic pulses, and we can neglect the repeated reflections and scattering of the emitted sound by neighboring regions of the random surface, the following representation for the spectrum of the mean field in the pulse is valid:

$$\langle \tilde{P}_a \rangle = \tilde{P}_a(\omega, \sigma = 0)\varphi(\omega). \tag{5.14}$$

Here $\tilde{P}_a(\omega,\sigma = 0)$ is the spectrum of the optoacoustic pulse in the absence of roughness of the boundary, while $\varphi(\omega) \equiv \langle \exp(i\omega\xi/c_a) \rangle$ is the characteristic function of the random quantity ξ, which is the vertical displacement of the surface $z = 0$ at the point having the coordinates x and y ($\xi = \xi(x,y)$). If the displacements obey a normal distribution law with the rms height of the roughness σ, then averaging over the realizations of the random surface $\xi = \xi(x,y)$ yields

$$\varphi(\omega) = \exp\left(-\frac{\sigma^2\omega^2}{2c_a^2}\right). \tag{5.15}$$

According to Eqs. (5.14) and (5.15), roughness of the surface creates an additional high-frequency cutoff of the spectrum of efficiently transferred frequencies at $\omega \lesssim \omega_\sigma \equiv \sqrt{2}c_a/\sigma$. In particular, if the frequency ω_σ is smaller than all the characteristic frequencies of the spectrum $\tilde{P}_a(\omega,\sigma = 0)$, then the duration of the sound pulses is completely determined by the characteristic time $\tau_\sigma = \omega_\sigma^{-1}$.[204] Analogously, under certain conditions ultrashort optoacoustic deformation pulses must contain information on the roughness of the irradiated surface of the solid. An increase in the duration of the front of an optoacoustic

pulse, caused by the use of a carbon coating instead of an aluminm one as the absorber, has been observed experimentally.[51] In the opinion of the authors, this was due specifically to the inhomogeneity of the carbon coating.

5.2. Control of the duration of acoustic pulses via nonlinear processes in the optoacoustic converter

Pulsed optical action can cause a substantial change in those physical parameters of the medium that depend on the temperature and the concentration of photogenerated electron-hole pairs. In such cases the optoacoustic phenomena depend to a great extent on the intensity of the incident laser radiation, although nonlinear acoustic effects in the excitation zone, as before, show up only weakly. In this section we shall discuss some of the similar regimes of generation of deformation pulses by volume sources nonlinear in intensity of the optical field.

According to the results of Refs. 77, 83, 207, and 208, in the case of dielectric media (poor heat conductors) the so-called "thermal nonlinearity" can be important in the thermoelastic mechanism of optoacoustic conversion (3.6). This phenomenonon involves the nonlinear coupling of the thermoelastic sources of sound of (3.6)–(3.7) with the intensity of the optical radiation through the temperature dependence of the bulk expansion coefficient ($\beta = \beta(T)$). In a theoretical analysis in the first approximation, it is sufficient to use a linear approximation for the $\beta(T)$ dependence:[207,208]

$$\beta(T) = \beta(T_0) + (d\beta/dT)_{T_0}(T - T_0), \qquad (5.16)$$

where T_0 is the initial temperature of the medium. It is clear that most favorable condition for observing the thermal nonlinearity in optoacoustic conversion is $\beta(T_0) = 0$. In particular, this is satisfied in water at the temperature $T_0 = 4$ °C. When the initial condition $\beta(T_0) = 0$ is fulfilled, the amplitude of the optoacoustic pulse increases quadratically with the absorbed energy. However, in the general situation, Eq. (5.16), the efficiency of optoacoustic conversion can increase with increasing intensity of the optical exposure. In the present context it is important that, in when $\beta(T_0)$ and $(\partial\beta/\partial T)_{T_0}$ have the same sign, the thermal nonlinearity can shorten the acoustic pulses, since the more strongly heated near-surface regions excite sound more efficiently. That is, as the intensity of the exposure increases, the characteristic depth of localization of the thermoelastic sound sources, (3.6) decreases when the duration of the excited optoacoustic pulses is determined by the time of passage of sound through the generation region, this leads to a decrease in the duration of the optoacoustic pulses. When we take account of the finite duration of the laser exposure, the thermal nonlinearity leads to an increase in the efficiency of sound excitation towards the end of the optical pulse and to an asymmetry of the acoustic signal.[207] Besides the studies already cited,[83,207] Ref. 209 was devoted to studying the thermal nonlinearity.

The depth of penetration of light can influence most directly the size of the near-surface region of localization of the sound sources. According to the linear theory (Sec. 3), in many physical situations the transfer functions of optoacoustic converion can be factored in such a way that one can explicitly separate out the factor $\omega\omega_\alpha/(\omega^2 + \omega_\alpha^2)$, which depends on the absorption coefficient for laser radiation $\alpha = \omega_\alpha/c_a$ [see, e.g., (3.13), (3.26), and (3.39)–(3.40)]. When the separated factor is decisive in the spectral transfer function of optoacoutic conversion, an increase in the light absorption coefficient leads both to an upward frequency shift of the region of most efficient sound generation ($\omega \sim \omega_\alpha$), and to a change in the width $\Delta\omega$ of the spectral band of efficient generation ($\Delta\omega \sim \omega_\alpha$).

In the case of the thermoelastic mechanism of the optoacoustic effect, the influence of the depth of penetration of light on the spectrum (and profile) of the optoacoustic pulses is strongest if the region of light absorption is thermally and acoustically thick [relation (3.13)]. In the case of nonlinear light absorption, the variation in $l_p = \alpha^{-1}$ with increasing intensity of optical exposure produces a proportional variation in the optoacoustic pulse length. It has been shown[210] that a nonlinear increase in the absorption coefficient ($\partial\alpha/\partial I_a > 0$) under such conditions shortens the deformation pulse length. We note also that the profiles of the optoacoustic pulses contain more detailed information on the spatial distribution of the radiation in the medium. An example[210] is the case of simultaneous light absorption in interband two-photon processes ($\alpha_{TPA} \sim I_a$) and by free carriers [$\alpha_{FCA} \sim n \sim (I_a)$] under conditions of fast nonlinear recombination of an electron-hole plasma ($\tau_R(n) \ll \tau_L$). The superlinear increase in the amplitude of the acoustic pulses and the shortening of their leading edges has been observed experimentally[211] in the irradiation of GaSb by yttrium-erbium-aluminum garnet radiation ($\lambda_L \approx 2.92$ μm, $\tau_L \approx 65$ ns). In this study the nonlinear light absorption showed up on a background of strong linear absorption ($\alpha_0 \approx 25$ cm^{-1}). According to the estimates of the authors the increase in concentration of carriers created by two-photon absorption ($2h\nu_L \approx 0.85$ eV $< E_g \approx 0.72$ eV) was saturated as a result of Auger recombination:

$$k_2 I_a^2/2h\nu_L \sim n/\tau_R(n) \sim \gamma n^3, \quad I_a \equiv (1 - R) I_i:$$

Here k_2 is the coefficient of two-photon absorption, and γ is the coefficient of radiationless Auger recombination. Here the following expression for the light absortion coefficient is valid:

$$\alpha(I_a) = \alpha_0 + \alpha_{FCA} + \alpha_{TPA} \approx \alpha_0 + \sigma(k_2/2h\nu_L\gamma)^{1/3} I_a^{2/3} + k_2 I_a.$$

Here σ is the light-absorption cross section of free carriers. It was found that the superlinear increase in temperature in the near-surface region of the GaSb crystal predominantly involved the absorption of energy by the additional non-equilibrium free carriers.

In the case $\tau_R(n) \gtrsim \tau_L$, nonlinear light absorption affects both the temperature and the concentration-deformation generation of sound. In Ref. 212 an analytic description was obtained for the profiles of optoacoustic pulses in the case in which non-equilibrium carriers were generated by single-photon interband absorption, while the distribution of radiation intensity in space was determined by absorption by free carriers ($\alpha_{FCA} \gg \alpha$):

$$\frac{\partial n}{\partial t} \approx \frac{\alpha I_a}{h\nu_L} \exp\left(-\sigma \int_0^z n(t, z')dz'\right) f(t) - \gamma n^3,$$

$$\frac{\partial T}{\partial t} \approx \sigma n \frac{I_a}{\rho c} \exp\left(-\sigma \int_0^z n(t, z')dz'\right) f(t) + \frac{E_g}{\rho c}\gamma n^3.$$

An analysis[77,212] showed that the duration of the acoustic pulses excited thermoelastically during the optical exposure decreases with increasing intensity of optical exposure faster than the duration of pulses excited by the concentration mechanism. This comes about because as the energy of the laser exposure increases, not only doe the dimensions of the region of heat release diminish, but in addition there is an increase in the fraction of the absorbed optical energy that is directly spent in heating the crystal lattice.

We note also that optoacoustic conversion has been studied under non-steady-state saturation of the light absorption.[213] It was found that the motion of the bleaching front can lead to the same transformation of the profile of an optoacoustic pulse (the leading edge is steeper than the trailing edge) as do the nonlinear acoustic effects during sound propagation. According to Ref. 213 we should expect a substantial symmetry of optoacoustic pulses at supersonic velocities of motion of the bleaching front. In addition, in the case of moving acoustic sources, we should expect excitation of the shortest pulses at near-sonic velocities.[109,111,214,215] The physical reason for the possible shortening of the optoacoustic pulses is the spatial grouping of the acoustic waves excited at different points of the trajectory of the sources. If the velocity of the sound sources depends on the intensity of the optical exposure (e.g., in the model of an expanding layer of a photogenerated electron-hole liquid[109]), the possibility arises of optimizing the conditions of optoacoustic conversion.

From the results [Eq. (3.26)] of the linea theory, the spectral transfer function of concentrational optoacoustic conversion depends on the recombination time of the photogenerated carriers, even when their motion can be neglected (for a diffusionally thick absorption region). Under intense enough laser pulses characteristic concentrations n of nonequilibrum carriers can be attained that are so high that their lifetimes will be determined by nonlinear recombination. That is, the lifetimes will depend on the concentration, and hence, on the light intensity. Here the process of optoacoustic conversion begins to depend strongly on the intensity of laser exposure. Since the lifetime of the carriers decreases with increasing n ($\tau_R \sim n^{-2}$ for Auger recombination, $\tau_R \sim n^{-1}$ for radiative two-particle recombination), the characteristic frquency $\omega_R = \tau_R^{-1}$ increases with increasing I_a. Here we can qualitatively write the transformation of the optoacoustic signal (3.11), (3.26)as

$$\tilde{U}_n(\omega) \sim \frac{(-i\omega)^2\omega_\alpha}{(\omega_R - i\omega)(\omega^2 + \omega_\alpha^2)}\tilde{f}(\omega) \qquad (5.17)$$

in the case $\omega_\alpha \ll \tau_L$. If the inequality $\omega_R \ll \omega_\alpha \ll \omega_L$ is satisfied for the starting intensities of the optical exposure, then the spectrum of the acoustic pulse is controlled by the factor $(-i\omega)\omega_\alpha/(\omega^2 + \omega_\alpha^2)$, while its profile resembles that described by Eq. (3.14). However, if fast bulk recombination $\omega_\alpha \ll \omega_L \ll \omega_R$ can be achieved by increasing the intensity I_a, then the spectrum of Eq. (5.12) can be approximated as follows:

$$\tilde{U}_n(\omega) \sim -\frac{\omega_\alpha}{\omega_R}\tilde{f}(\omega).$$

Consequently the profile of the optoacoustic pulse excited by the deformation mechanism comes to resemble the profile of the laser pulse.[39] Here the duration of the deformation pulse was shortened to $\tau_a \sim \tau_L \ll \omega_\alpha^{-1} = l_p/c_a$. The possibilities of shortening optoacoustic pulses excited by the concentration mechanism to durations less than the time of passage of sound through the region of light absorption were first discovered in a theoretical study.[39] In thermoelastic optoacoustic conversion a similar phenomenon is not possible, since a mechanism of fast turnoff of the acoustic sources that does not depend on the scale of the region of their spatial localization (like the recombination of carriers in the concentration mechanism is lacking). In this regard we also note that the scheme discussed in Ref. 41 for excitation via the concentration mechanism of optoacoustic pulses of duration $\tau_a \gtrsim 0.1$ ps under the condition $\tau_R = \infty$ actually presupposes the δ-localization of the sound sources (at the bondary of the region of photogeneration of carriers).

A shortening of the duration of the concentration component of an optoacoustic pulse by increasing the intensity of optical exposure in silicon was observed experimentally in Ref. 85 (Fig. 30). By increasing the surface density of laser radiation ($\lambda_L \approx 1.06$ μm, $\tau_L \approx 20$ ns) in the range $W_s^i \sim 0.01$–0.6 J/cm^2, the duration of the compression phase in the acoustic wave was shortened from $\tau_a \sim (\alpha c_a)^{-1} \sim 100$ ns to $\tau_a \sim \tau_L \sim 20$ ns (see Fig. 30). According to the theoretical calculations,[85,86,212] the time of Auger recombination of carriers becomes comparable to τ_L for $W_s^i \sim 0.25$ J/cm^2.

The linear theory predicts [see, e.g., formula (3.29)], that a decrease in the recombination time can also lead to expansion of the region of efficient optoacoustic conversion toward high frequencies when the region of light absorption is diffusionally thin. The physical reason for this is the decrease in the size of the region of localization of the carriers, which under these conditions can be determined by the diffusion length $l_D \sim \sqrt{D\tau_R}$. In particular, when the inequalities $\sqrt{\omega_D\omega_R} \ll \omega_L \ll \omega_R$, are satisfied, then one can write spectrum of the concentration component of the optoacoustic pulse, using (3.29) and (3.11), in the form

$$\tilde{U}_n \sim \frac{(-i\omega)^2(\omega_D\omega_R)^{1/2}}{(\omega^2 + \omega_D\omega_R)\omega_R}. \qquad (5.18)$$

Consequently, the characteristic duration of the acoustic

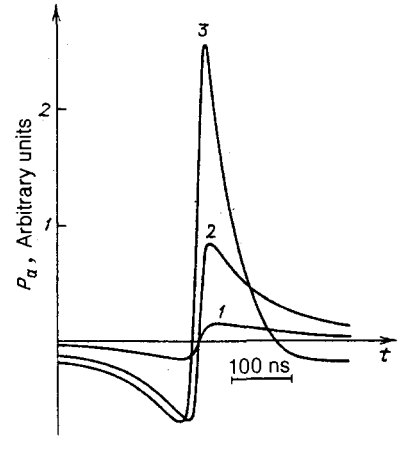

FIG. 30. Profiles of optoacoustic pressure pulses recorded[85] at different energies of laser exposure in silicon W_s^i (J/cm^2) = 0.014 (*1*), 0.14 (*2*), and 0.53 (*3*).

pulse is determined by the time of passage of sound through the region of localization of the electron-hole plasma:

$$\tau_a \sim (\omega_D\omega_R)^{-1/2} \sim (D\tau_R)^{1/2}/c_a \sim l_D/c_a.$$

However, we note that the inequality that determines the applicability of (5.18) necessarily requires supersonic velocities of diffusion v_D of the electron-hole plasma:

$$v_D \sim l_D/\tau_R \sim (\omega_R/\sqrt{\omega_R\omega_D})c_a \gg c_a.$$

If the recombination of the carriers occurs nonradiatively, then, according to Eqs. (3.21) and (3.22), the length l_D determines the characteristic dimension of the region of recombination heating. The thermoelastic component of the optoacoustic pulse initiated by recombination heating:

$$\tilde{U}_T \sim (-i\omega)\sqrt{\omega_D\omega_R}/(\omega^2 + \omega_D\omega_R)$$

can predominate over the concentration component (5.18). An analytic calculation was performed[100] to determine the profiles of optoacoustic pulses under similar conditions. The nonlinearity of the recombination processes ($\tau_R \sim n^{-2}$) and the dependence of the diffusion coefficient of the degenerate electron-hole plasma on the concentration ($D \sim n^{5/3}$), characteristic of predominant scattering of carriers by ionized inclusions, were taken into account exactly.[216] It was established that in this special low-temperature region the depth of penetration of the plasma depends extremely weakly on the intensity of optical pulse ($l_D \sim I_a^{-1/17}$), since the decrease in the lifetime of electron-hole pairs is almost completely compensated by their increased diffusion coefficient.

Other nonlinear effects also can hinder the diffusion expansion of the region of localization of photogenerated carriers. For example, the results of optoacoustic experiments[217] in germanium indicate the confinement of a nondegenerate photoexcited electron-hole plasma in the near-surface potential well that arises from the local photoinduced thermal alteration of the bandgap of the semiconductor ($\partial E_g/\partial T < 0$).

The possibility of altering the durations of optoacoustic pulses excited via the piezoeletric effect is additionally determined by the dependence of the frequencies of Maxwell relaxation and Debye screening on the concentration of nonequilibrium carriers ($\omega_{Mm} \sim n^m$, $\omega_{Deb} \sim \sqrt{n^m}$) at sufficiently intense laser exposures ($n^m \gg n_0^m$, $m = e,h$). In agreement with (3.42) the linear theory predicts, in particular, that under certain conditions the characteristic optoacoustic pulses length can be of the order of the Debye screening time $\tau_a \sim \tau_{Deb} \sim \omega_{Deb}^{-1}$.[42] A nonlinear model describing this situation can be studied analytically in the case in which the instantaneous optical exposure leads to the photoexcitation of electrons into the conduction band from the surface energy levels. If we consider the holes to be immobile, then after such a surface photogeneration they create a dc field inside the crystal:

$$E_h = \frac{Q_s^h}{2\varepsilon\varepsilon_0} = \frac{Q_0 W_s^a}{2\varepsilon\varepsilon_0\hbar\omega_L} \sim W_s^a. \tag{5.19}$$

Since $f(t) \sim \delta(t)$, the motion of the electrons is described by the homogeneous equation

$$\frac{\partial n^e}{\partial t} = \mu_e\frac{\partial}{\partial z}(En^e) + D_e\frac{\partial^2 n^e}{\partial z^2}. \tag{5.20}$$

Since holes are absent in the bulk of the crystal ($n^h \equiv 0$), then, according to Eqs. (3.32) and (3.33) the changes in the electric field E in the crystal involve only the distribution of the electron concentration:

$$\frac{\partial E}{\partial z} = -\frac{Q_0}{\varepsilon\varepsilon_0}n^e. \tag{5.21}$$

By using Eqs. (5.20) and (5.21) and the condition that the field falls off into the interior of the crystal [$E(z\to\infty) = 0$], we can derive a closed equation for the electric field:

$$\frac{\partial E}{\partial t} - \mu_e E\frac{\partial E}{\partial z} - D_e\frac{\partial^2 E}{\partial z^2} = 0. \tag{5.22}$$

The Burgers equation (5.22) allows an analytic solution that satisfies the initial condition $E(z>0, t=0) = 0$ and the boundary conditions $E(z=0+0, t) = 2E_h$ $E(z\to\infty, t) = 0$. The characteristic value of the electric field E, of course, is of the order of E_h in (5.19). Here the characteristic depth of penetration of the field can be estimated from the order-of-magnitude equality of the nonlinear and diffusion terms in Eq. (5.22):

$$z_s = D_e/\mu_e E_h \sim (W_s^a)^{-1}. \tag{5.23}$$

At such distances the retardation of the diffusional motion of the electrons completely compensates their drift in the electric field. Correspondingly, a steady-state field distribution

$$E(t=\infty, z) = E_h\left(1 + \frac{z}{|z|}\right)\left(1 + \frac{z}{z_s}\right)^{-1} \tag{5.24}$$

is established in a characteristic time

$$t_\infty \sim z_s/v_{dr}^e \sim z_s/\mu_e E_h \sim (W_s^a)^{-2}. \tag{5.25}$$

We can easily see that t_∞ is of the order of the Maxwellian relaxation time of the conduction electrons having the characteristic concentration $n_s^e \sim Q_s^h/Q_0 z_s$. We note that, according to (5.25), the time of establishment of the distribution, Eq. (5.24), is inversely proportional to the square of the absorbed laser energy. If the characteristic velocities of the electrons $v_{dr}^e \sim v_D^e \sim \mu_e E_h \sim W_s^a$ are supersonic, then from the standpoint of generating acoustic waves the electric-field distribution of (5.24) is established instantaneously. Yet the excitation of optoacoustic pulses actually occurs in a steady-state field. The profile of the deformation pulse has the form

$$U(\tau) \sim \frac{eE_h}{\rho c_a^2}\frac{\tau}{|\tau|}\left(1 + \frac{c_a|\tau|}{z_s}\right)^{-1}. \tag{5.26}$$

The characteristic duration of the acoustic pulse is determined by the time of passage of sound through the region $0 \leqslant z \leqslant z_s$ of the space-charge distribution: $\tau_a \sim z_s/c_a \sim \tau_{Deb}(n_s^e)$. Because of relations (5.19) and (5.23), when one increases the intensity of the optical pulse the amplitude of the optoacoustic pulse (5.26) increases linearly ($|U| \sim W_S^a$), while the pulse length is reduced [$\tau_a \sim (W_S^a)^{-1}$]. We emphasize that in this model the concentration of electrons increases, both directly because of their increased rate of photogeneration, and owing to the decreased size of their region of localization.

In closing this section we note that the results of a num-

ber of experiments show that radiative transport of energy can play an important role under certain conditions in the space-time dynamics of the photogenerated electron-hole plasma in semiconductors,[218,219] and in the heating of targets during ablation.[69,220] However, we know of no studies of the influence of this effect on the process of optoacoustic conversion.

Finally, we note that nonlinear changes in optoacoustic pulses can involve the nonlinear dependence of the boundary conditions on the intensity of laser exposure in the equations determining the bulk sources of acoustic waves. For example, according to Eqs. (3.30) and (3.43), the spectral transfer functions depend on the surface recombination velocities, which, like the bulk recombination, can be nonlinear.[221] Yet in the process of thermoelastic optoacoustic conversion in metals, a temperature dependence of the absorptive power $(1-R)$ of the target surface can be manifested.[222]

Of undoubted interest is a possible broadening of the duration τ_a of optoacoustic pulses as compared with the duration τ_L of the laser pulses (to eliminate this discrepancy is the fundamental problem in the design of optoacoustic generators of ultrashort deformation pulses) can be useful in designing optoacoustic devices to measure the duration of ps–fs optical pulses. Thus, it has been shown[223] that in the two-photon absorption of light in the field of two colliding laser pulses one of the characteristic measures of the region of localization of the sound sources can prove to be of the order of the spatial extent of the laser pulses in the medium: $L_G \sim c_L \tau_L$. Here the duration of the excited deformation pulses can be proportional to τ_L $[\tau_a \sim L_G/c_a \sim (c_L/c_a)\tau_L]$, but enlarged on the scale $(c_L/c_a) \sim 10^3$–10^4. In this way measurement of the duration of fs-ps laser pulses can be reduced to measuring the duration of ps–ns deformation pulses.[223]

6. CONCLUSIONS

We hope that this review contains enough information to gain an idea of the potentialities of the so-called "picosecond hyperacoustics"[224] in the field of both applied and fundamental studies. The effectiveness and reliability of the experimental methods that use optoacoustic generation and acoustooptical reception of ultrashort deformation pulses is indicated by the vigorous growth in recent years in the number of publications in this field. Besides the studies already cited, we refer also to the experimental studies, Refs. 225 and 226.

The material presented in this review allows one to conclude with full justification that pico- and femtosecond lasers have stimulated the rise of a new field of physical and applied acoustics–picosecond hyperacoustics. Although the experimental technique of picosecond hyperacoustics is still in the process of development, and in constrast to pico- and femtosecond laser technology, the limiting parameters of ultrashort acoustic pulses are still to be realized, it is reasonable to give a brief sketch of the new possibilities opening up in spectroscopy, diagnostics of non-equilibrium states, in nonlinear acoustics, and in the physics of the action of high-power acoustic pulses on matter.

1) Picosecond acoustic technology is the basis of time-dependent acoustic spectroscopy—it is also called acoustic echo spectroscopy.

Acoustic echo spectroscopy has already been applied successfully for diagnostics of thin films and multilayer structures. For a film thickness $d \sim 10^3$ Å, multiple acoustic echo yields direct information on the velocity and the acoustic absorption of the material of the film at hypersonic frequencies; numerous examples were presented in Sec. 2. As was already stated, for $\tau_a \sim 100$ fs the methods of time-dependent acoustic spectroscopy can be used in principle for probing crystal lattices: $c_a \tau_a \sim 5$ Å.

2) Generators of ultrashort acoustic pulses whose spectra extend to frequencies of 10^{11}–10^{13} Hz (100 GHz–10 THz) can be effective sources in schemes of broad-band, "frequency" acoustic spectroscopy.[3)]

We note, incidentally, that to extend the acoustic spectrum, laser generation of ultrashort acoustic simple pulses is far from being the only possibility.

Substantial progress in hyperacoustics can involve the development of methods of obtaining sequences of ultrashort laser pulses with high ($\nu_r \sim 0.1$–10 THz), adjustable repetition rates. Amplitude modulation of light at frequencies $\nu_r \approx 0.3$ THz[228] and $\nu_r \approx 5$ THz[229] has been obtained by exploiting the development of instability during propagation of laser radiation in optical fibers. By using such optical systems, one can excite and record sound directly at the frequency of repetition of the laser pulses or its harmonics.

3) We note also that in essence an efficient spectroscopic diagnostic method involves the actual process of optoacoustic sound generation (see, e.g., Refs. 35, 85, and 217)–going to pico- and femtosecond pulses allow one, as was shown in Sec. 3, to extract information on the dynamics and relaxation of carriers in semiconductors.

4) Essentially new data can be obtained by picosecond hyperacoustics in the diagnostics of short-lived non-equilibrium states in solids. In this sense the "cold" melting of semiconductors[8,9] and the dense femtosecond laser plasma (see Refs. 11–13 and the material of Sec. 4) briefly mentioned in the Introduction are especially interesting.

In the former case we refer to completely unusual states of the crystal lattice that persist for times not exceeding ~ 200 fs, in which, according to optical data,[8,9] long-range order breaks down, but short-range order persists.

The properties and origin of this state are not well understood; we can naturally expect that acoustic probing in

TABLE I. Record achievements in the generation of electromagnetic and acoustic pulses at the end of 1990.

	Pulses				
Parameter	Microwave	Optical	Incoherent x rays	Acoustic	Parameter
Duration	1 ps	600 fs	500 fs	5 ps	Duration
Power:					Pressure:
at present	10^3 W	10^{15} W		3×10^7 bar	at present
projected	10^9 W	10^{17} W		3×10^9 bar	projected
					Pressure gradient:
Intensity:					
at present	10^6 W/cm^2	10^{19} W/cm^2	10^8 W/cm^2	10^5 bar/Å	at present
projected	10^{10} W/cm^2	10^{20} W/cm^2	10^{11} W/cm^2	10^7 bar/Å	projected

the subpicosecond time scale would enable one to clarify the situation substantially.

An extremely interesting physical object with unusual acoustic properties is the dense femtosecond laser plasma.

Even at an electron temperature $T_e \sim 300$ eV, the expansion velocity of the plasma does not exceed 5×10^7 cm/s. Hence the cluster of laser plasma near the surface of the target, which has a characteristic thickness of 500–600 Å, practically maintains its dimension for times of 200–300 fs; thus a condensed medium arises with a density of 10^{22}–10^{23} cm^{-3} and a temperature reaching 10^6 K.

5) Among the new problems of nonlinear acoustics initiated by the advances in generating ultrashort deformation pulses and short shock pulses are the generation of acoustic solitons and the initiation of fast phase transitions by the high dynamic pressure (see also the material of Secs. 2 and 4).

In summarizing the material presented in this review, we can state that the methods of generating short high-power radiation bursts that were first developed in laser physics exert an ever-growing influence on physical and applied acoutics. Table I characterizes the record attainments in generating short high-power pulses obtained by the end of 1990 in the optical range, in the microwave range, in soft x rays, and acoustics. Unique possibilities are opened up in time-domain spectroscopy of the behavior of matter under conditions of extremely strong agents. The technology of high-power pico- and femtosecond pulses allows one to apply extreme agents for extremely short times.

The combination of optical, x-ray, and acoustic methods in studying fast processes, in exciting strongly non-equilibrium states of matter, and in studying fast phase transitions in nonlinear dynamics will undoubtedly bring many new, unexpected results. We can expect with full justification that the contribution of the acoustic methods to the development of this highly promising and broad branch of physics will be ever more important.

[1)]We are discussing the qualitative diagram characterizing the orders of magnitude in avalanche and tunneling ionization of a nonresonance medium; for more details see Ref. 16.

[2)]Extensive information on the physical and technical aspects of optoacoustic studies in the megahertz frequency range is contained in the review articles, Refs. 24–26.

[3)]The terms time-domain and frequency-domain spectroscopy came from contemporary optical spectroscopy; actually the question is that of "Fourier-conjugate" methods of extracting information on the parameters of matter.

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