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On The Generation of Terahertz Pulses Using Thermoelastic Effect

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Abstract. Time-resolved terahertz spectroscopy can be proposed as a method of non-destructive testing of various parts, for example, aircraft composite materials. The theoretical background and the possibility of exciting pulses of electromagnetic waves in the range of terahertz oscillations using picosecond acoustic pulses excited by laser pulses based on the thermoelastic effect are studied.

Keywords: terahertz spectroscopy, terahertz oscillations, laser pulses, thermoelastic effect

1. Introduction

High-energy THz pulses are attractive for such applications as nonlinear interaction with nanostructured materials [1], nonlinear spectroscopy [2, 3], molecular alignment [4], and others [5, 6]. There are quite a few methods of generating THz radiation today [7-14]. They include the most popular method of optical rectification of femtosecond laser pulses with titled pulse fronts in lithium niobate [7-9], photoconductive antennas [10], emission of THz waves from the surface of material due to ultrafast charge transfer [11], THz pulse emission caused by the acceleration of electron bunches in free-electron lasers [12], and others [13, 14]. Experiments using subpicosecond lasers have demonstrated the generation and detection of acoustic and shock waves in materials with terahertz frequencies [15-17]. It has been shown that these pulses propagating in piezoelectrics produce THz radiation [17]. However, the method proposed in [17] for generation of high-frequency ultrasonic waves involves ablation, i.e. vaporization of material from the surface of the optoacoustic generator based on an aluminum film and the generation of ultrashort powerful ultrasonic pulses with uncontrolled shape. Moreover, aluminum is not the best choice for devices capable of generating powerful ultrasonic pulses and consequent THz electromagnetic radiation in piezoelectric material.

This paper analyzes the possibility of generating terahertz electromagnetic pulses through the use of picosecond acoustic pulses excited via the laser-induced thermoelastic effect.
2. Theoretical framework.

The thermooptical sound excitation mechanism is responsible for the generation of extremely short acoustic pulses whose duration is limited by the Debye frequency. The shortest acoustic pulse duration obtained so far is about 10 ps. Even in the thermoelastic regime [19-22] when no damage is produced on the surface of an absorbing material, the amplitude of acoustic (pressure) pulses can be tens and hundreds of MPa, being limited by laser-heating-induced phase transitions of the material. The propagation of acoustic pulses in a piezoelectric is accompanied by charge transport, which can possibly incite short (picosecond) electromagnetic pulses with a frequency of up to a few THz (terahertz band). This paper analyzes this possibility.

The thermo-optical excitation of sound involves the heating of a medium by a short laser pulse and subsequent thermal expansion of the heated layer [20, 22]. The duration of the acoustic pulse in this case is determined by the laser pulse duration and the time sound takes to travel through the heat-generation layer with a characteristic depth of $c^\beta$, where $c$ is the optical absorption coefficient. The duration of the excited acoustic signal is limited by this coefficient. Therefore, it is preferable to use metals or semiconductors as generators provided that their energy gap width is less than the laser photon energy and $c$ reaches $10^9$ m$^{-1}$.

Absorption of the optical photon energy causes the excitation of the electron subsystem of a metal; its energy is thermalized and temperature becomes stabilized after a few electron-phonon collisions with duration $\tau_{e-p} \approx 10^{-11} - 10^{-12}$ s have occurred [19, 20], which usually takes several picoseconds. The processes occurring at the initial stage of laser heating are usually described by the kinetic model for the electron and phonon subsystems of the medium [19]. Within quite a long time interval ($t \geq 3 + 5\tau_{e-p}$), the temperature of the electron subsystem of the medium becomes equal to that of the phonon system and local thermodynamic equilibrium is reached. At these timescales, the processes can be described by the traditional Fourier thermal diffusion model.

In view of the foregoing, we assume that if a laser pulse is several picoseconds in duration, the electron-ion subsystem in metals reaches thermodynamic equilibrium and the temperature of electrons becomes practically equal to that of the lattice. Given that the pulse has energy $100 \mu$J and duration $\tau_0 = 2nc$ and taking into account that the coefficient of reflection from metal surface can reach 90%, the absorbed energy surface density is of the order of $w_0 = 0.5 J/m^2$ at optical beam width $a = 2mm$. In this case, we have the thermoelastic regime, when the optical beam is absorbed in the near-surface zone of material and there are no phase transitions. In this regime, the temperature $T$ of the heated region increases by a few tens of degrees. Thus, silver with the light absorption coefficient $\alpha = 10^8$ m$^{-1}$ is heated by $\Delta T = -\frac{\alpha w_0}{\rho_0 c_p} \approx 20K$ (here $\rho = 10.6 \times 10^3$ kg/m$^3$ is the density of silver and $c_p = 250$ J/(kg × K) is the isobaric specific heat). The subsequent expansion of the heated region leads to the generation of elastic waves. The spectrum and time profile of the resulting pulses of elastic waves can be determined using the traditional model of thermooptical sound excitation [21, 22]. In this case, the wave equation written for scalar potential $\varphi(\vec{r}, t)$ of the field of oscillation velocities of particles $\vec{V}(\vec{r}, t) = \nabla \varphi(\vec{r}, t)$ is as follows [21]:

$$c_0^2 \frac{\partial^2 \varphi(\vec{r}, t)}{\partial t^2} - \Delta \varphi = -\frac{\alpha \beta}{\rho_0 c_p} \left( 1 - 4 \frac{c_i^2}{3c_0^2} \right) I_0 e^{-\alpha z} H(x, y) g(t)$$

(1)
Here \( c_0 \) and \( c_t \) are the longitudinal and shear wave velocities; \( \rho_0 \) is the density, \( C_r \) is the isobaric specific heat, \( I_0 \) is the peak intensity of temporal envelope \( g(t) \) of laser pulse, \( \beta \) is the coefficient of volumetric expansion, and \( H(x,y) \) describes the intensity distribution of the optical beam over the cross section. The spatial extent of the excited acoustic pulses \( c_0 \tau_0 \sim 20 \text{ nm} \) is small as compared to the laser beam spot size; therefore it is sufficient to use the plane-wave model and regard the problem as a one-dimensional one. So, it can be assumed that \( H(x,y) = 1 \) and \( \Delta \varphi = \partial^2 \varphi / \partial z^2 \) (the acoustic beam propagates along the \( z \) axis); only one component of the oscillation velocity of particles is nonzero: \( V_z = \partial \varphi / \partial z \). The temporal profile and spectral range of the acoustic signals can be determined from equation (1) using the transfer function method [23], in accordance with which the spectrum of the excited pulse is equal to the product of the Fourier transform of the temporal envelope of the intensity of the laser pulse \( g(\omega) \) and the transfer function \( K(\omega) \) of the layer in which optoacoustic conversion takes place. Transfer function \( K(\omega) \) is determined using the following characteristics: the light absorption coefficient of the medium and ultrasound velocity therein:

\[
V_z(t=t-z/c_0) = \frac{\beta w_0}{2\rho_0 C_v} \int_{-\infty}^{t} K(\omega)g(\omega)e^{-i\omega t} d\omega \tag{2}
\]

For the free boundary of the absorbing medium, the transfer function is \( K(\omega) = -i\alpha \omega (\alpha^2 c_t^2 + \omega^2)^{-1} \) and \( g(\omega) = \exp(-\omega^2 \tau_0^2 / 4) \) for a Gaussian beam. Figure 1 shows the amplitude spectra and temporal profiles of ultrasonic signals for the absorption coefficient \( \alpha = 10^4 \text{ m}^{-1} \), laser pulse duration \( \tau_0 = 2 \text{ ns} \), and longitudinal wave velocities \( c_0 = 4,3 \cdot 10^8 \text{ m/s} \) (curve 1) and \( c_0 = 6,0 \cdot 10^7 \text{ m/s} \) (curve 2). In these cases, parameter \( A = \alpha c_0 \tau_0 \) is 0.8 and 1.2, respectively, and the generation is most effective [24]. The amplitude spectrum of the signals extends to 2 THz.

To justify the geometry of the problem, let its spatial scales be estimated. The main parameters of metals that can be used as optoacoustic generators are given in Table 1. Here \( \lambda \) is the coefficient of thermal conductivity, \( \chi = \lambda / (\rho_0 C_r) \) is the thermal diffusion coefficient, and \( T_m \) is the melting temperature.

Table 1. The main parameters of metals that can be used as optoacoustic generators

| Material  | \( \rho_0 \), \( 10^3 \text{ kg/m}^3 \) | \( C_r \), J/(kg-K) | \( \lambda \), W/(m-K) | \( \chi \), \( 10^{-6} \text{ m}^2/\text{s} \) | \( c_0, 10^3 \text{ m/s} \) | \( \beta \), \( 10^{-8} \text{ K}^{-1} \) | \( T_m \), °C |
|-----------|-----------------|------------------|-------------------|----------------|--------------|----------------|----------|
| Nickel    | 8.9             | 440              | 52                | 12             | 5.63         | 13.7           | 1455     |
| Chrome    | 7.2             | 462              | 97                | 30             | 6.84         | 8.4            | 1880     |
| Silver    | 10.6            | 250              | 425               | 142            | 3.60         | 20.1           | 960      |
| Aluminum  | 2.7             | 903              | 236               | 84             | 6.20         | 24.2           | 600      |
The depth of light penetration is $L_\alpha = \alpha^{-1} = 10 \text{ nm}$; the penetration depth of laser radiation is $3L_\alpha = 30 \text{ nm}$. The thermal diffusion length during laser exposure $L_T = \sqrt{\chi \tau_0}$ varies from 5 nm (for nickel) to 20 nm (for silver). This means that a metal film no more than 20 nm thick is heated uniformly during the action of laser pulse. The spatial extent of the acoustic pulse $L_\alpha = c_0 \tau_0$ varies from 8 to 15 nm.

![Figure 1. Amplitude spectra and temporal profiles of ultrasonic signals](image)

Thus, the characteristic spatial scales of the problem are no more than 30 nm. The following geometry is proposed: a metal film 60-100 nm thick is deposited on a lithium niobate substrate whose surface roughness is no worse than $\lambda/30$; this film absorbs optical radiation. The subsequent thermoelastic expansion of the heated zone of the metal film causes the generation of a short powerful ultrasonic pulse. This pulse propagates into lithium niobate where a broadband electromagnetic wave pulse is produced due to the piezoelectric effect. Pressure amplitude $p_0$ of the ultrasonic pulse is $p_0 = \frac{\alpha c_2 \beta w_0}{2C_p}$ and the magnitude of the electric field strength is $E_0 = d p_0 / \varepsilon_0$, where
$d = 6 \cdot 10^{-12} \text{C/N}$ is the piezoelectric coefficient for lithium niobate and $\varepsilon_0 = 8.85 \cdot 10^{-12} \text{C/m \cdot V}$.

Estimates of $p_0$, $E_0$, and temperature rise due to laser pulse absorption by metal films are given in Table 2.

| Material   | $\Delta T, K$ | $p_0, \text{MPa}$ | $E_0, 10^5 \text{V/m}$ |
|------------|---------------|-------------------|------------------------|
| Nickel     | 21            | 25                | 14                     |
| Chrome     | 15            | 21                | 12                     |
| Silver     | 20            | 26                | 15                     |
| Aluminum   | 21            | 21                | 12                     |

As follows from Table 2, the amplitude of ultrashort ultrasonic pulses generated in metals in the thermoelastic regime is about 20 MPa, the absorbed energy density being 0.5 J/m². The near-surface region is heated by no more than 25 °C. The subsequent propagation of the resulting pulse in a piezoelectric of the lithium niobate type results in the generation of an electromagnetic wave pulse with an electric field strength of about 107 V/m and a frequency of 0.1 to 2.5 THz. If the surface energy density will increase by an order of magnitude, the pressure amplitude and electric field strength will increase by an order of magnitude as well. In this case, local heating of a metal film will reach 200-250 °C so that thermal nonlinearity will come into play; this means that the temperature dependence of the thermal expansion coefficient should be taken into account. This effect will cause the spectral range of the excited THz pulses to be extended up to 4 THz.

3. Conclusions

This paper shows, that the amplitude of ultrashort ultrasonic pulses generated in metals in the thermoelastic regime is about 20 MPa, the absorbed energy density being 0.5 J/m². The near-surface region is heated by no more than 25 °C.

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