An experimental demonstration of the effect of mechanical stresses on the laser generation of acoustic vibrations in various materials

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Abstract. The process of generating an acoustic signal during laser heating of metals and dielectrics is analyzed in the framework of the theory of thermoelasticity. A comparison of the experimental optoacoustic signal and the theoretical distribution of mechanical stresses for plates with a hole of aluminum alloy and silicon nitride is performed. It was found that the standard theory of thermoelasticity can qualitatively describe the dependence of the optoacoustic signal on the stress near the hole in the silicon nitride plate, but is not enough to correctly describe the dependence of the signal on the stress near the hole in the metal plate. It is noted that in order to achieve agreement between the obtained experimental and theoretical results for metals, in addition to the thermal effect of laser radiation on the lattice, it is necessary to take into account the additional effect of the electron system.

1. Introduction

Laser optoacoustic methods have been successfully used to obtain information on the mechanical, thermoelastic and thermophysical properties of bulk materials, solid-state structures, and thin films. At present, theoretical methods have been developed in detail for calculating optoacoustic signals in a gas medium contacting with the object under study, elastic vibrations and waves excited in solid objects and structures. The theoretical conclusions have been confirmed by results of numerous experiments performed on solid objects and liquids.

Of particular interest is the use of laser optoacoustic microscopy methods for assessing residual stresses in various materials. It has been experimentally shown that internal and external mechanical stresses in regions with a complex rheological structure significantly affect the characteristics of optoacoustic signals [1-4]. Comparison of experimental data with existing models of thermoelastic generation of acoustic vibrations showed that the usual approach, taking into account only thermal expansion, does not allow us to correctly describe the dependence of the optoacoustic signal on internal stresses. Moreover, it was shown by various authors that the laser-induced excitation of acoustic waves in conductors and dielectrics has a different character [5-7].

This paper presents experimental results of optoacoustic investigations performed on stressed metals and dielectrics with known distribution of stress around small holes. The stress distribution corresponds approximately to the solution of the Kirsch problem [8]. To explain the experimental results, we proposed to use the thermoelastic equation of motion for a real crystal lattice. It is assumed that the lattice has defects such as vacancies, interstitial atoms, voids, grain boundaries, cracks, etc. The time and energy dynamics of these defects provides a significant contribution to the elastic...
behavior of the lattice. It is important that this approach takes into account the dynamics of metal electron subsystem.

2. Methods
In this paper, we studied the relationship between optoacoustic signals and mechanical stress in various materials using an example of a model problem for which the stress distribution is well known. The object of the study was rectangular plates of metal and ceramic with a hole. The stress distribution near the hole under uniaxial loading is known and is determined by solving the Kirsch problem. Using methods of laser scanning optoacoustic microscopy, images of the regions of the studied samples around small holes were obtained under an external uniaxial load. To adequately describe the behavior of optoacoustic signals depending on the load, it was proposed to take into account quasi-equilibrium excitations of defective states in such objects with their subsequent gradual relaxation to the initial states. In the framework of this work, it is assumed that additional generation of defects in the material occurs due to temperature modulation upon absorption of laser radiation.

Experimental studies were performed on metal plates with a hole with a diameter much smaller than the size of the sample. The signal from the plate was recorded by scanning optoacoustic microscopy with a piezoelectric signal detection. The experimental setup is shown in figure 1. The heat waves were excited by intensity-modulated radiation from a solid-state laser at a wavelength of 532 nm. The average radiation power on the surface of the object was 40 mW. Temporary modulation of the exciting laser radiation was carried out by an acousto-optic modulator at the resonant frequencies of piezoelectric transducers. Then the periodically modulated laser beam was focused on the surface of the sample into a spot with a diameter of 15 μm. The piezoelectric transducer was slightly pressed to the rear side of the sample through coupling gel. The sample was located in the region of maximum sensitivity of the piezoelectric sensor in its central region. To eliminate the possible influence of the vibration modes of piezoelectric sensors, we used a round piezoelectric sensor with a working resonant frequency of 101 kHz and a rectangular piezoelectric sensor with a resonant frequency of 141 kHz. The piezoelectric signal was measured by a high-frequency lock-in amplifier with a special preamplifier. A compressive load was applied to the sample using a screw mechanism mounted on a two-axis automated stage. The construction moved in two directions to make an optoacoustic image.
3. Experimental results

The article presents the results of two experiments on the study of stressed objects. Optoacoustic images of the region around the hole in the silicon nitride sample are shown in figure 2. Figure 2a shows the region in the unstressed sample. The signal distribution is almost uniform. In the image of the uniaxially loaded sample at -57 MPa (figure 2b), the areas with an increased and decreased signal are located diametrically around the hole. It is generally accepted that compressive stresses have negative values. The location of these areas is close to the distribution of the compression and extension areas around the hole. Namely, the dark areas in the figure correspond to compression, and light areas with an increased signal correspond to stretching. This behavior of the optoacoustic signal qualitatively corresponds to the theoretical prediction based on thermodynamic representations of the dependence of thermal expansion coefficient on deformations. However, quantitatively, the obtained effect of stress on the signal is much larger than a few percent predicted by the theory. The maximum signal magnitude is 2.2 for the region where theoretically the tensile stress is expected to be +57 MPa, provided that the signal magnitude far from the hole at -57 MPa is 1. The minimum magnitude is 0.31 for the region with compressive stress near -170 MPa.

Figure 3 demonstrates results of the similar experiment with the aluminum plate. The initial distribution of the optoacoustic signal is generally uniform, which corresponds to the absence of strong internal stresses in the initial state of the plate. The image obtained at a compressive stress of -57 MPa (figure 3b) shows also diametrically located light and dark regions around the hole. The fundamental difference from the previous case is that an increase in the signal relates to compressive...
stress and, correspondingly, a signal decrease relates to tensile stress. This fact contradicts the classical thermoelastic theory of the formation of an optoacoustic signal for stressed objects.

Thus, the experiments performed on metal and dielectrics show that to describe the thermoelastic generation of ultrasonic vibrations in various materials, it is not enough to use thermodynamic approaches that describe the well-known dependence of the coefficient of thermal expansion on deformation.

4. Discussion
To describe the laser generation of acoustic vibrations, a pair of equations of heat conduction and motion is usually considered. For solids, we can restrict ourselves to thermoelastic coupling only in the equation of motion. In works devoted to stress analysis by measuring thermal emission, the dependence of thermoelastic parameter on stresses is theoretically considered basing on classical thermodynamics. The stress dependence was explained by the temperature dependence of the elastic properties of a material [9]. This dependence for the coefficient of thermoelastic coupling \( k_t \) is determined by the following expression

\[
\frac{\partial k_t}{\partial \sigma} = \frac{K \alpha_T(\sigma)}{\rho} = \frac{\partial E}{\partial T},
\]

where \( \rho \) is the density of the medium, \( K \) is the bulk modulus, \( \alpha_T \) is the volume thermal expansion coefficient, \( E \) is Young’s modulus, and \( \partial E / \partial T \) is negative for the vast majority of materials. That is why the optoacoustic signal should increase slightly in the presence of tensile stress, which is not consistent with the experimental data.

The application of IR methods with cyclic mechanical loading for stress analysis demonstrates good quantitative confirmation of this dependence of the volume thermal expansion coefficient for various materials [9, 10]. Our experiments with ceramic samples demonstrate that optoacoustic methods based on thermoelastic expansion allow the detection of stresses that are qualitatively predicted by classical thermodynamics. As for metals, the experiment results are fundamentally different. Classical thermodynamics fails because there are dynamic processes in metals with excitation of defects that it does not take into account and which do not appear in ceramics. Such a difference, apparently, is associated with a difference in chemical bonds in ceramics and metals. Ceramics are characterized by strong covalent bonds, while metals are characterized by weaker metal bonds.
As was shown in [3,11] using optothermal methods, constant mechanical stresses do not noticeably affect the temperature fluctuations caused by modulated laser radiation. To describe the above experimental results, it is necessary to introduce additional sources of vibrations in the equation of motion.

It is assumed that the lattice has various types of defects. To adequately describe the behavior of optoacoustic signals depending on the load, we propose to take into account quasi-equilibrium excitations of defective states in such objects with their subsequent gradual relaxation to the initial states. In the framework of this work, it is also assumed that additional generation of defects in the material occurs due to temperature modulation upon absorbing laser radiation.

These transitions are accompanied by corresponding changes in the free energy [12] and the volume of the crystal lattice. In accordance with [13], the presence of defects in a material causes a change in its free energy proportionally to the concentration of defects, a dilatation parameter, and a comprehensive compression modulus.

To calculate the displacement of the surface of an object in accordance with the proposed model, a variable component of the free energy gradient caused by the presence of defects should be added to the equations of motion. In the case of a metal sample, an additional term describing the inhomogeneous change in the pressure of the electron gas due to thermally excited electrons from defects in the metal structure should be added. As a result, the equation of motion will look as follows

\[
\frac{\partial^2 u}{\partial t^2} = c_l^2 \Delta u + (c_l^2 - c_t^2) \text{grad}(\text{div} u) - \frac{K\alpha_T}{\rho} \text{grad} T + \frac{K}{\rho} \sum_d \Omega_d \text{grad} n_d + \frac{2E_F}{3\rho} \text{grad} n_e, \tag{1}
\]

where \( u \) is the displacement vector, \( c_l \) and \( c_t \) are the longitudinal and transverse speeds of sound, \( \rho \) is the density of the medium, \( K \) is the bulk modulus, \( \alpha_T \) is the volume expansion coefficient, \( \Omega_d \) is a dilatation parameter for a defect of type \( d \), \( E_F \) is Fermi energy, \( n_d \) and \( n_e \) are concentrations of lattice defects and defect electrons correspondingly. The dilatation parameter describes the change in the volume because of formation of one defect. Summation is made for all defects of any type. \( \Omega_d \) can be either greater or less than 0, for example, for interstitial atoms and vacancies, respectively. The third term in the right-hand side of this equation describes the deformation source due to the thermal expansion in inhomogeneous temperature field, the fourth term describes stresses introduced into the medium by the lattice defects, and the last term describes pressure of thermally excited electrons from defects.

The dynamics of the concentration of defects can be described using the particle balance equation with a source function in the form of the Arrhenius law:

\[
\frac{\partial n_i}{\partial t} + \frac{n_i}{\tau_d} = A_d \exp(-U_d/kT_0) \beta T(t,r), \tag{2}
\]

where \( \tau_d \) is the relaxation time of defects, \( \tau_e \) is the relaxation time of the system electrons-lattice, \( A_{d,e} \) are constants, \( U_{d,e} \) is the activation energy of a defect or electron, \( k \) is the Boltzmann constant, \( T_0 \) is the average temperature of the heated region, and \( \beta T \) is the variable component of the temperature.

The activation energy \( U \) for electrons or defects depends on deformation, and, consequently, on stresses \( U \approx U_0 + V \sigma \), where \( V \) is the so-called activation volume.

Assuming periodic temperature oscillations with a cyclic frequency \( \omega \), it is possible, taking into account the solution of equation (2), to introduce the effective coefficient of thermal expansion, which also includes the contributions from the diffusion of defects and thermally delocalized electrons. In the linear approximation with respect to residual stresses for \( V \sigma \ll kT_0 \) it can be represented as

\[
\alpha_{\text{eff}}(\omega) \equiv \alpha(\omega) - \frac{\sigma}{E^2} \frac{\partial E}{\partial T} - \frac{\sigma}{E^2} \frac{\partial E}{\partial T} \left[ \sum_d \frac{\Omega_d A_d \tau_d (U_{d0} - kT_0)}{(1 + (\omega \tau_d)^2)kT_0^2} e^{-U_{d0}/kT_0} + \frac{2E_F A_V \tau_e (U_{e0} - kT_0)}{3k(1 + (\omega \tau_e)^2)kT_0^2} e^{-U_{e0}/kT_0} \right]
\]
Here $\alpha(\omega)$ is the effective coefficient of thermal expansion in the absence of stresses. The second term on the right side represents the classical thermoelastic dependence of the coefficient of thermal expansion on mechanical stresses. The expression in square brackets corresponds to the contribution from the imperfection of the crystal lattice to this dependence. The sign and magnitude of this contribution may be different for different types of defects, as well as for conductive materials. It should be emphasized that we are talking about dynamic effects comparable in time with the relaxation time of the excitation of defects. For the obtained experimental data for Si$_3$N$_4$ ceramics, the second term in square brackets is obviously zero, and the first should be negative in order to enhance the contribution from the thermoelastic component. According to the proposed model, both additional terms will be zero for an ideal crystal, and the change of the PA signal at the maximum possible load will not exceed several percent. In the case of a metal sample, the expression in square brackets should be positive and significantly larger than the thermoelastic component. This is possible, given that $U_{e0} > kT_0$, since in the opposite case, the defect electrons are delocalized already in the initial state. Thus, the proposed model of laser sound generation allows a qualitative explanation of the experimental data for ceramics and real metals.

5. Conclusions
A comparison of experimental and theoretical results shows that it is not enough to use a purely thermodynamic approach to explain the influence of mechanical stresses on optoacoustic signals. A correct explanation of this dependence requires taking into account the relaxation behavior of the defect and electron subsystems of the metal.

Thus, taking into account the formation of dynamic defective states during laser irradiation allows us to describe the experimental data on the excitation of acoustic vibrations quite well. Theoretical and experimental results show that the optoacoustic method in combination with the method of drilling holes can be used to assess mechanical stresses in various materials.

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