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PHOTOTHERMAL DETERMINATION OF THE THERMOPHYSICAL CHARACTERISTICS OF SOLID-STATE OBJECTS

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The possibilities of determining the heat conduction, thermal diffusivity, and specific heat of thin solid-state layers by a photothermal method are investigated theoretically and experimentally.

Photothermal heating methods based on the optical generation of thermal and acoustic waves in condensed media have been developed intensively and are extensively utilized in scientific investigations and applications [1]. These methods are highly informative, local, contact-free, possess high fast-response, and in principle, permit the determination of the thermal properties of substances in a broad range of state parameters [2].

The development of photothermal diagnostic methods for solids with spatially inhomogeneous thermal parameter distribution draws great interest. The physical principles of such measurements have been developed sufficiently weakly even for the simplest kinds of thermal inhomogeneities. In this paper the problem of the simultaneous determination of the thermal diffusivity, thermal conductivity, and specific heat coefficients is examined for a layer located on a substrate with known thermophysical characteristics.

For definiteness we will examine the gas microphone method of recording [2]. The structure being investigated (layer + substrate) is here placed in a photoacoustic cell (Fig. 1) and exposed to light with the modulated intensity $I = I_0/2(1 + \cos \omega t)$. Pressure fluctuations Δp associated with the thermal expansion of the gas layer heated from the specimen u_t and the mechanical displacement of the specimen surface being exposed u_m [3] occur in the cell

$$\Delta p \sim \frac{P_0}{l_0}\,(u_t(0) + u_m(0)) = \frac{P_0}{l_0}\left(\eta_g^{-1}\,\frac{\Delta T(0)}{T_0} + u_m(0)\right), \tag{1}$$

where $\eta_g = (1 + j)(\omega/2\chi_g)^{1/2}$.

Formula (1) is valid when the thickness of the gas layer is large as compared with the thermal length and small as compared with the corresponding sound wavelength $\ell_0 \gg \ell_g$, $\ell_0 \ll \lambda_g$, $\ell_g = (2\chi_g/\omega)^{1/2}$. The quantities $\Delta T(0)$, $u_m(0)$ depend in a complex manner on the thermal, acoustic, optical properties and the geometric dimensions of the layer and substrate, the heat transfer and clamping conditions on the interfacial boundaries, which makes analysis of the dependence of the photoacoustic response $\Delta p \sim u_t(0) + u_m(0)$ on the thermal parameters of interest to us difficult in the general case.

Let us examine the case, which is of practical interest, of surface ($x = 0$) generation of heat and long sound waves in a layer and substrate $\ell \ll \lambda$, $\ell_1 \ll \lambda_1$. Let us neglect heat transfer to the specimen outer boundaries, and we consider the temperature and heat flux continuous on the interfacial boundary $x = \ell$. Let the surface $x = 0$ be free of mechanical stresses and $x = \ell + \ell_1$ clamped stiffly. In this case the mechanical vibrations of the specimen surface are not important and the pressure in the gas is determined by just the temperature of the surface being exposed

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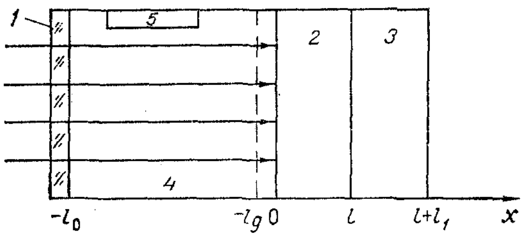

Fig. 1. Mathematical model of a photoacoustic cell: 1) input window; 2) layer; 3) substrate; 4) chamber; 5) microphone.

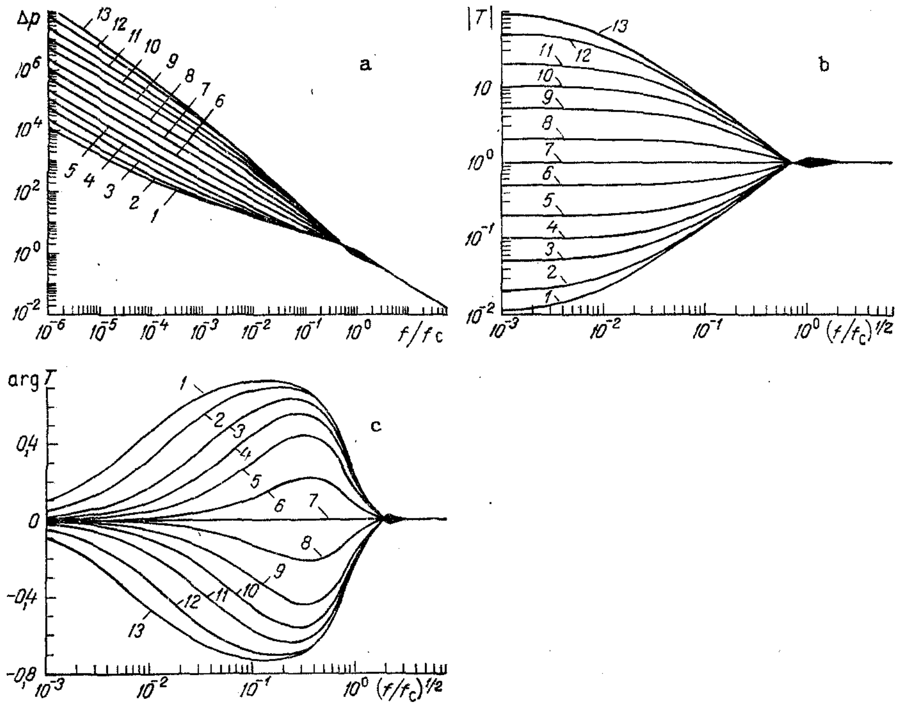

Fig. 2. Frequency dependences of the photoacoustic effect in a two-layered structure a) $\Delta p(f/f_c)$; b) $|T(f/f_c)|$; c) arg $T(f/f_c)$). 1) m = 100; 2) 50; 3) 20; 4) 10; 5) 5; 6) 2; 7) 1; 8) 0.5; 9) 0.2; 10) 0.1; 11) 0.05; 12) 0.02; 13) 0.01. Δp relative units; arg T, rad; f/f_c is the dimensionless frequency.

$$T = \frac{\Delta T(0)}{\Delta T_\infty} = 1 + 2\left[\exp(2\eta l)\left(\frac{1 + m\,\mathrm{th}\,\eta_1 l_1}{1 - m\,\mathrm{th}\,\eta_1 l_1}\right) - 1\right]^{-1},$$
$$\eta = (1+j)a, \quad \eta_1 = (1+j)a_1, \quad a = (\omega/2\chi)^{1/2}, \tag{2}$$
$$a_1 = (\omega/2\chi_1)^{1/2}, \quad m = \frac{\varkappa_1\eta_1}{\varkappa\eta} = \left(\frac{\varkappa_1 C_1}{\varkappa C}\right)^{1/2},$$
$$\Delta T_\infty = I_0/\varkappa\eta.$$

Formula (2) is obtained in a one-dimensional approximation, is valid if the transverse dimensions of the microphone chamber exceed the diameter of the light spot and the thermal lengths in the gas and the specimen [4].

In the case of a "thermally thick" substrate ($a_1\ell_1 \gg 1$)

$$T = 1 + 2\left[\exp(2\eta l)\left(\frac{1+m}{1-m}\right) - 1\right]^{-1}. \tag{3}$$

This simplification yields the lower bound of the radiation modulation frequency at $\chi_1/\pi\ell_1^2$.

Formula (3) was obtained first by Rosenzwaig and Gersho [5] and relates the relative temperature of the exposed surface to the thermal characteristics of the layer and substrate.

TABLE 1. Thermophysical Characteristics of Silicon and Copper Plates

Parameter	Temperature, K	Measured value		Literature value	
		Silicon	Copper	Silicon	Copper
χ, m²/sec	300	$0{,}93\cdot 10^{-4}\pm 5\%$	$0{,}73\cdot 10^{-4}\pm 6\%$	$(0{,}82-0{,}94)\cdot 10^{-4}$ [11]	$(0{,}3-1{,}2)\cdot 10^{-4}$ [11, 14]
$\varkappa$, W/(m·K)	300	$150\pm 8\%$	$240\pm 11\%$	120—208 [12, 13]	100—400 [14]
C, J/(m³·K)	300	$1{,}7\cdot 10^{6}\pm 9\%$	$3{,}3\cdot 10^{6}\pm 13\%$	$1{,}77\cdot 10^{6}$ [12]	$3{,}39\cdot 10^{6}$ [14]

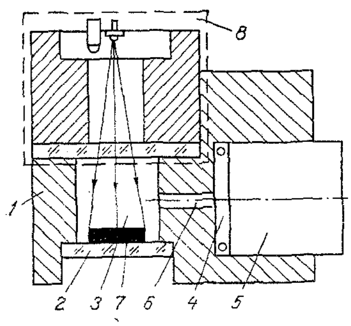

Fig. 3

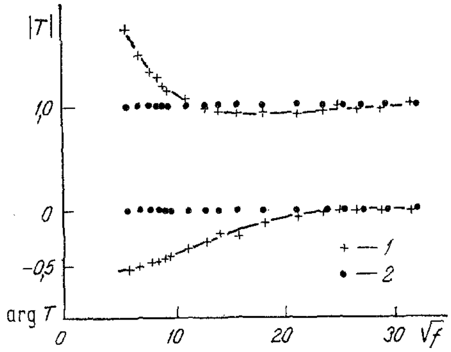

Fig. 4

Fig. 3. Gas-microphone photoacoustic cell with light diode as radiation source: 1) housing; 2) exit window; 3) specimen chamber; 4) microphone chamber; 5) MKÉ-10 microphone with preamplifier; 6) connecting channel; 7) object; 8) illuminator.

Fig. 4. Frequency dependences of the photoacoustic effect of solid-state structures 1) silicon on quartz; 2) silicon on silicon; arg T, rad; $\sqrt{f}$, $Hz^{1/2}$.

The Rosenzwaig–Gersho formula and its corollary were discussed and investigated experimentally [6-8], however, literature data available at this time are few, contradictory, and do not permit making unique deductions about the possibility of determining the thermophysical characteristics of solid-state structures from photoacoustic measurements.

The thermal characteristics of the layer and substrate enter into (3) by means of two parameters, m and the critical frequency $f_c = \chi/\pi \ell^2$. In meaning this is the frequency at which the thermal wavelength in the layer is compared with its thickness, evidently $a\ell = (f/f_c)^{1/2} = \Omega^{1/2}$ and T is a complex quantity characterized by the amplitude $|T|$ and phase arg T with respect to the reference signal, the harmonically modulated light intensity. In principle, two independent information sources permit the simultaneous determination of the values of m and f_c in measurements of the amplitude and phase of the photoacoustic signal at one modulation frequency.

Let us analyze numerically the frequency dependences of the photoacoustic effect. Graphs of the functions $|T(\Omega^{1/2})|$, arg $T(\Omega^{1/2})$, $\Delta p(\Omega)$ computed by means of (3) for the most widespread values of m are represented in Fig. 2 [9].

The frequency characteristics of the microphone response $\Delta p(\Omega)$ in the long ($\Omega \ll 1$) and short ($\Omega \gg 1$) thermal wave domains have the form $\Delta p \sim 1/\Omega$, which corresponds to a slope n = 1 in logarithmic coordinates (Fig. 2a), the transition domain $\Omega \sim 1$ occupies a broad frequency range whose boundaries depend on the parameter m.

The relative temperature of the surface being exposed changes from 1 (thermal waves do not reach the substrate) to m, the thermal wavelength is much greater than the layer thickness and the quantity T is determined by the substrate properties. For m < 1 the substrate reflects hear while for m > 1 it efficiently eliminates heat from the layer. In the domain $\Omega \sim 1$ the dependence $|T(\Omega^{1/2})|$ has an extremum associated with the thermal wave interference. The family of curves in Fig. 2b has two singular points whose coordinates are practically independent of m, these are the extremum pointand the point $\Omega^{1/2} \approx 0.8$, where $|T| = 1$. If the location of either of these points is successfully determined exactly in test, then a simple calculation of the thermal diffusivity χ of the layer is possible. Two independent measurements of $|T|$ for different values of the argument Ω are necessary as a minimum for the determination of both thermophysical characteristics of the layer.

In practice, both the amplitude and the phase characteristics are utilized; the latter are apparently more preferable since the value of the argument $\arg \Delta T_\infty = -\pi/4$ is known while the measurement of $|\Delta T_\infty|$ is not always possible. The phase dependences in Fig. 2c have the form of curves with an extremum whose magnitude and location are related uniquely to the parameters m and f_c. If the location and magnitude of the extremum are successfully determined exactly in test, the values of the desired parameters are found by using special tables [8]. In the general case, as above, two independent measurements of the phase of the photoacoustic response are necessary as a minimum.

An experimental confirmation of the amplitude and phase methods of measurement was performed on two pairs of model structures [9]. The first pair, silicon on quartz and silicon on silicon, was a p-Si plate 0.36 mm thick which was glued alternately on thermally thick ($\approx$ 2 mm) quartz or silicon substrates. The glued surfaces were polished optically. The second pair, copper on quartz and copper on copper, had an analogous construction, the thickness of the glued copper plate was 0.34 mm. The transverse dimensions of all the specimens were ~ 1 cm and salol, vacuum grease, and indium–gallium paste were used as glueing material.

The structures being investigated were placed in a gas-microphone photoacoustic cell with light diode as radiation source (Fig. 3) [9]. Frequency dependences of the amplitude and phase of the photoacoustic response were recorded for each of them in the 30-1000 Hz band. The amplitude and phase were measured with ±1% and ±1° accuracy. The cell was calibrated by copper and brass substrates. Later all the experimental data were normalized by the calibration characteristics according to the method described in [8].

The amplitude–frequency and phase–frequency characteristics of the silicon on quartz and silicon on silicon structures are presented in Fig. 4. They differ substantially from each other; the characteristics of the last structures agree with the calibration characteristics of the homogeneous thermally thick substrate. The presence of layer-substrate interfacial boundaries did not appear anywhere in the test data since the condition $R_a < R_1$ [10] was apparently satisfied in the frequency band utilized.

Theoretical curves verified by experimental points by least squares are also presented in Fig. 4. The sum of the squares of the deviations were minimized by using the gradient method of steepest descent. The error in calculating the parameters m and f_c was computed by using the three sigma rule, which yields a 0.99 fiducial probability for 17 measurements. Numerical values of the parameters m and f_c were $m = 0.11 \pm 4\%$ and $f_c = 229$ Hz $\pm 2\%$. As was assumed they simultaneously satisfy the experimental dependences of the photoacoustic response amplitude and phase.

Analogous tests and computations were performed for a copper on quartz structure for which the values of the adjustment parameters were $m = 0.6 \pm 10\%$ and $f_c = 200$ Hz $\pm 5\%$.

The thermal diffusivity, thermal conductivity, and specific heat of silicon and copper were determined from the formulas $\chi = \pi\ell^2 f_c$, $\kappa = x_1/m(\chi/\chi_1)^{1/2}$, $C = \kappa/\chi$; their values are presented in Table 1. The following thermophysical characteristics of fused quartz were used $\kappa_1 = 1.34$ W/(m·K), $C = 2.1 \cdot 10^6$ J/(m³·K) [14, 15]. It is seen from Table 1 that the computed values are in good agreement with the literature data.

We also examined the case of light absorption and heat generation on the layer-substrate interfacial boundary $x = \ell$. Their results of numerical computations of the frequency dependences of the amplitude and phase of the photoacoustic responses showed that simultaneous determination of the thermal diffusivity, thermal conductivity, and specific heat of a layer is also possible in the frequency band $\Omega \lesssim 1$ by determining the parameters m and f_c by least squares on the amplitude or phase measurements.

Therefore, an experimental confirmation strengthened the correctness of the method elucidated for determining the thermophysical characteristics of thin solid-state layers during their photothermal heating. The graphical dependences presented (see Fig. 2) can be utilized to estimate the amplitude and phase of the photoacoustic responses of the most widespread pairs of coatings–substrates. The developed programmed support permits determination of the parameters m and f_c just by the amplitude or phase characteristics which is convenient in practice.

Let us mention that processes of temperature wave propagation and the thermophysical characteristics of solid-state interfacial boundaries can be investigated by photoacoustic methods. Evidently necessary for this is compliance with the condition $R_a \sim R_1$. Application of photoacoustic methods to study the interfacial boundary structure seems to us quite important and special investigations are needed in this area.

The development of other methods of photothermal diagnostics (optical and piezoelectrical recording of thermoelastic strains of a specimen, etc.) for problems on the quantitative determination of the thermophysical characteristics of thin solid-state layers merits attention.

NOTATION

I, intensity; I_0, amplitude value of the intensity; $\omega = 2\pi f$, circular radiation modulation frequency; t, time; κ, κ_1, thermal conductivity; χ, χ_1, thermal diffusivities; C, C_1, bulk specific heats; η, η_1, thermal-wave wave vectors; λ, λ_1, sound wavelengths; ℓ, ℓ_1, layer and substrate thicknesses, respectively; Δp, variable pressure in the cell; u_t, effective thermal expansion of the gas; u_m, mechanical shift of the specimen surface; P_0, T_0, equilibrium pressure and temperature in the gas; χ_g, thermal diffusivity; ℓ_g, thermal wavelength; η_g, thermal-wave wave vector; λ_g, sound wavelength in the gas, respectively; ℓ_0, cell length; $\Delta T(0)$, ΔT_∞, amplitudes of temperature fluctuations of exposed two-layered and infinitely thick layer surfaces; T, dimensionless amplitude of the temperature fluctuations of the exposed two-layered structure surface; m, ratio of the layer and substrate thermal activities; f_c, critical frequency; Ω, dimensionless frequency; R_a, thermal resistance of the glueing, and R_1, characteristic thermal resistance of the substrate.

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