# QUASI-STATIONARY THERMAL REGIME IN A FILM-SUBSTRATE SYSTEM DURING PERIODIC-PULSED HEATING BY LASER RADIATION

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*The effect of thermal conductivity of the film-substrate contact boundary on the establishment of the quasi-stationary thermal regime during periodic-pulsed heating is studied.*

The influence of the nonideality of the film-substrate thermal contact on the temperature field configuration T(t, x) has been considered in connection with the development of the laser heat treatment process [1-3], the evolution of thermal flaw detection methods [4, 5], and measurement of adhesion characteristics of coatings [6, 7]. Film heating by radiation of lasers which operate in the pulsed and continuous regime has been extensively investigated. However, until the present time there has been no work done to study heating by radiation of lasers operating in the periodic-pulsed regime. It is known [8] that during such heating three stable quasi-stationary thermal regimes for $T(t,x) = \overline{T}(x)+\Theta(t,x)$; $|\Theta| << \overline{T}$ arise in the plates: the periodic-pulsed regime, the regular regime of the second kind, and the periodic-stationary regime; the form of the temperature pulsations $\Theta(t,x)$ of these regimes remains constant within a wide range of variation in the repetition pulse period of laser generation $t_n$ and the duty factor $\gamma$. The stability region for thermal regimes is determined by the pulse-repetition rate, the plate thickness, and thermophysical characteristics of the material.

We may expect that in the film-substrate system the character of the relation $\Theta(t,x)$ will be defined also by the thermal conductivity of the contact boundary $\alpha_{12}$. The question now arises on the stability of the quasi-stationary thermal regime and the form of the temperature pulsation with changes in the parameter $\alpha_{12}$. In this connection we performed a mathematical simulation of the periodic heating process of the film-substrate system by surface heat sources under the condition that the contact boundary is nonideal. The quasi-stationary thermal regime at different values of $\alpha_{12}$ is experimentally and theoretically investigated for the tungsten film-silicon substrate system widely used in the production of integrated microcircuits [9]. The obtained analytical relations are applied to solve the inverse problem for determining the thermal conductivity of the contact boundary $\alpha_{12}$ by the known relation $\Theta(t,x)$.

To derive the computational formulas, we write the linearized system of equations for the temperature pulsations

$$\frac{\partial \Theta_j}{\partial t} = a_j \frac{\partial^2 \Theta_j}{\partial x^2},\ \Theta_j = \Theta_j(t,\ x),\ j = 1,\ 2;\ t \geqslant 0,\ \ \Theta_j(0,\ x) = \Theta_2(t,\ \infty) = 0,$$

$$\left(-\lambda_1 \frac{\partial \Theta_1}{\partial x} + \alpha \Theta_1\right)\Big|_{x=0} = \overline{q}(t);\ \ \overline{q}(t) = q(t) - \overline{q},\ \alpha = 4\sigma\varepsilon \overline{T}^3(0), \tag{1}$$

$$q(t) = q(t + \nu t_n);\ \nu = 1,\ 2,\ \ldots,\ \ \overline{q} = \sigma\varepsilon\,[\overline{T}^4(0) - T_0^4] - \lambda \overline{T}_x' = 0.$$

Here indices 1 and 2 refer to the film and substrate, respectively. The conditions at the contact boundary of $x=\xi$ have the form

$$\left(\lambda_1 \frac{\partial \Theta_1}{\partial x} - \lambda_2 \frac{\partial \Theta_2}{\partial x}\right)_{x=\xi} = 0, \tag{2}$$

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$$\left[\lambda_1 \frac{\partial \Theta_1}{\partial x} + \alpha \Theta_1 + \alpha_{12}(\Theta_1 - \Theta_2)\right]_{x=\xi} = 0, \quad 0 \leqslant \alpha_{12} < \infty.$$

At an ideal thermal contact $\alpha_{12} \to \infty$, $a_1 = a_2$ and $\lambda_1 = \lambda_2$ the system (1) coincides with that used in [10] for a semilimited sample. When $\alpha_{12} = 0$ the film and substrate laminate, and the equations for $\Theta_1(t,x)$ pass to those given in [8] for describing the temperature pulsations in the plates.

In order to find the solution asymptotics at $t \to \infty$, which simulate the established quasi-stationary thermal regime, we write the solution of the problem in terms of the contour integral

$$\Theta_j(t, x) = \frac{1}{2\pi i} \int\limits_{p_0 - i\infty}^{p_0 + i\infty} H_j(p, t, x) \bar{\bar{q}}(p)\, dp, \quad p_0 > 0, \tag{3}$$

where

$$H_1(p, t, x) = \{A(p) \operatorname{ch}(x \sqrt{p/a_1}) + B(p) \operatorname{ch}[(\xi - x)\sqrt{p/a_1}]\} \exp(pt);$$

$$H_2(p, t, x) = \{C(p) \exp[-(x - \xi)\sqrt{p/a_2}]\} \exp(pt);$$

$$\bar{\bar{q}}(p) = \frac{\bar{c}_0}{p} + \sum_{k \neq 0} \frac{c_k}{p - i\omega_k}; \quad \omega_k = 2\pi k / t_n,$$

while the unknown coefficients A, B, and C are determined from the boundary conditions:

$$A(p) = -(1 + \alpha/\alpha_{12})/\alpha\Delta(p), \quad B(p) = \left[\left(\sqrt{\frac{D_1}{D_2}} + \frac{\lambda_1}{\alpha_{12}} \sqrt{\frac{p}{a_1}}\right) \operatorname{sh} u + \right.$$

$$\left. + (1 + \alpha/\alpha_{12}) \operatorname{ch} u\right] \Big/ \alpha\Delta(p), \quad C(p) = \left[\sqrt{\frac{D_1}{D_2}} (1 + \alpha/\alpha_{12}) \operatorname{sh} u\right] \Big/ \alpha\Delta(p),$$

$$\Delta(p) = \left[\left(\sqrt{\frac{D_1}{D_2}} + \frac{\lambda_1}{\alpha_{12}} \sqrt{\frac{p}{a_1}}\right) \operatorname{sh} u + (1 + \alpha/\alpha_{12}) \operatorname{ch} u\right] \times$$

$$\times \left[\frac{\lambda_1}{\alpha} \sqrt{\frac{p}{a_1}} \operatorname{sh} u + \operatorname{ch} u\right] - (1 + \alpha/\alpha_{12}), \quad u = \xi \sqrt{\frac{p}{a_1}}.$$

Points $p_k = 2\pi i k / t_n$ and $k \neq 0$ make the main contribution to the integral. As $\bar{c}_0 = 0$ [10], the contribution of the point $p = 0$ is equal to zero. Therefore, by computing residues, for the asymptotics we obtain

$$\Theta_j(t, x) = 2 \sum_{k=1}^{\infty} \operatorname{Re}[c_k H_j(p_k, t, x)]. \tag{4}$$

For the periodic-pulsed heating

$$q(t) = \begin{cases} q_0, & 0 \leqslant t \leqslant t_u, \quad q(t) = q(t + \nu t_n), \\ 0, & t_u < t < t_n, \quad \nu = 1, 2, \ldots, \end{cases}$$

the coefficients $c_k$ of the Fourier expansion function of q(t) are

$$c_k = q_0 \frac{\sin \psi_k}{\pi k} \exp(-i\psi_k); \quad \psi_k = \pi k \gamma, \quad k \neq 0. \tag{5}$$

Substituting (5) into (4), for the temperature pulsations we have

$$\Theta_j(t, x) = \frac{2\Theta_0}{\pi} \sum_{k=1}^{\infty} \frac{\sin \psi_k}{k} \operatorname{Re} \frac{L_k^j(x)}{\Delta_k} \exp\{i(\omega_k t - \psi_k)\}, \tag{6}$$

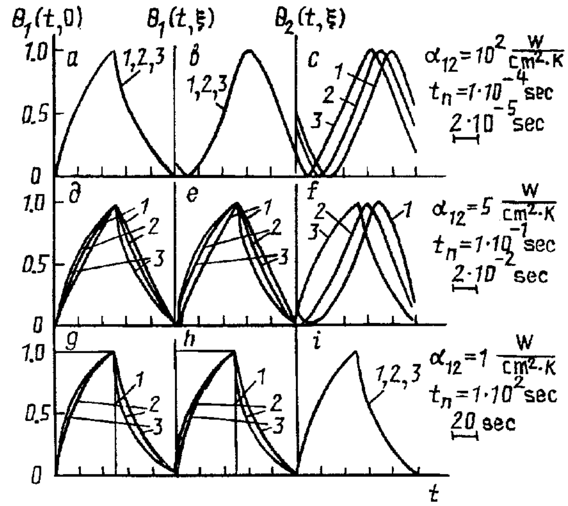



Fig. 1. The normalized form of the temperature pulsation on the film surface (a, d, g) at the film-to-substrate (b, e, h) and the substrate-to-film (c, f, i) contact boundary: 1) $\alpha_{12} = 0$; 2) interstitial values of $\alpha_{12}$ (indicated in the figure); 3) $\alpha_{12} = \infty$. Tungsten is the material of the film and substrate; the film thickness $\xi = 50\,\mu$m. $\bar{T} = 1600$ K.

where the function $L^j_k(x)$ has the form:

at the free surface of the film x = 0

$$L^1_k(0) = \frac{1}{2}\left(\sqrt{\frac{D_1}{D_2}} + h_{12}\sqrt{ik}\right)\mathrm{sh}\,(2\beta\sqrt{ik}) + (1 + \alpha/\alpha_{12})\,\mathrm{sh}^2\,(\beta\sqrt{ik}),$$

at the film-to substrate contact boundary $x = \xi - 0$

$$L^1_k(\xi) = \left(\sqrt{\frac{D_1}{D_2}} + h_{12}\sqrt{ik}\right)\mathrm{sh}\,(\beta\sqrt{ik}),$$

at the substrate-to film contact boundary $x = \xi + 0$

$$L^2_k(\xi) = \sqrt{\frac{D_1}{D_2}}\,(1 + \alpha/\alpha_{12})\,\mathrm{sh}\,(\beta\sqrt{ik})$$

and

$$\Delta_k = \frac{1}{2}\left[\sqrt{\frac{D_1}{D_2}} + (h_1 + h_{12})\sqrt{ik}\right]\mathrm{sh}\,(2\beta\sqrt{ik}) +$$

$$+\left[\left(\sqrt{\frac{D_1}{D_2}} + h_{12}\sqrt{ik}\right)h_1\sqrt{ik} + (1 + \alpha/\alpha_{12})\right]\mathrm{sh}^2\,(\beta\sqrt{ik}),$$

$$\beta = \xi\sqrt{\omega/a_1}; \quad h_1 = \alpha^{-1}\sqrt{D_1\omega}; \quad h_{12} = \alpha_{12}^{-1}\sqrt{D_1\omega}; \quad \Theta_0 = q_0/\alpha; \quad \omega = \frac{2\pi}{t_n}.$$

Figure 1 shows the character of dependences of $\Theta_1(t, 0)$, $\Theta_1(t,\xi)$, and $\Theta_2(t,\xi)$ calculated by formula (6) for different values of the thermal conductivity $\alpha_{12}$. To compare the results of the calculations and those of [8] ($\alpha_{12}$ = 0) and of [10] ($\alpha_{12} = \infty$), the film thickness is taken to be equal to $\xi = 50\,\mu$m, and tungsten is chosen as the material for the film and substrate. For visualization, the curves are normalized to unity and the duty factor is taken as $\gamma$ = 0.5. The repetition pulse periods $t_n = 10^{-4}$ sec (Fig. 1a-c), $10^{-1}$ sec (Fig. 1 d-f), and $10^2$ sec (Fig. 1 g-i) correspond to three stable forms of the temperature pulsations found in [8] for the plate. We see that during the periodic-pulsed heating regime (Fig. 1a-c) the form of the pulsation at the film surface x = 0 and at the contact boundary $x = \xi - 0$ is independent of the thermal conductivity $\alpha_{12}$. At the substrate-to-film contact boundary $x = \xi + 0$ (Fig. 1c) a pulsation shift in time which decreases with growth of $\alpha_{12}$. At the pulse-repetition rate 100 Hz (Fig. 1d-f), when $\alpha_{12}\to 0$ the periodic-pulsed heating regime of the film transforms to the second-kind thermal regime (curves 1 in Fig. 1 d, e) and the form of the pulsation depends on $\alpha_{12}$ for all the three surfaces. At the rate $10^{-2}$ Hz (Fig. 1 h-i) with the change in $\alpha_{12}$ from 0 to $\infty$ the п-shaped form of the pulsation in the film, corresponding to the periodic-stationary heating regime, is degenerated and the character of the variation of $\Theta_j(t)$ becomes close to that calculated for the periodic-pulsed heating regime at the surface x =0 (a).

The quasi-stationary periodic-pulsed heating regime and the regular thermal regime of the second kind were studied in [8, 10-12]. Therefore, let us consider in more detail the periodic-stationary regime. At this heating regime of the free film, thermal inertia of the medium is absent and the temperature over the whole film thickness instantly assumes only two fixed values:

$$\Theta_1(t,\ x) = \begin{cases} \Theta_1,\ 0 \leqslant t \leqslant t_u, & \Theta_1(t,\ x) = \Theta_1(t+\nu t_n,\ x),\ \ \nu = 1,\ 2,\ \ldots, \\ -\Theta_2,\ t_u < t < t_n, & \Theta_0 = \Theta_1 + \Theta_2. \end{cases} \tag{7}$$

Now we find out how the representation of (7) is related to formula (6), which prescribes the function $\Theta_1(t,x)$ for $\alpha_{12} \in [0,\infty[$. For this purpose we emphasize that at $\alpha_{12}=0$ and $t_n \gtrsim 10^2$ sec we have, to a good approximation, $L_k^1(0) = L_k^1(\xi) = \Delta_k = \sqrt{iD_1\omega_k}\ \mathrm{sh}(\beta\sqrt{ik})$. Upon reducing fractions, we come to the representation of (7) in the form

$$\Theta_1(t,\ x) = \frac{2\Theta_0}{\pi} \sum_{k=1}^{\infty} \frac{\sin\psi_k}{k} \exp\{i(\omega_k t - \psi_k)\}.$$

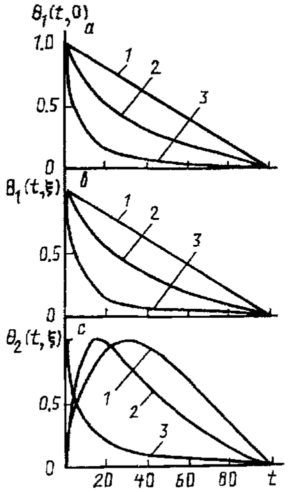
It is evident from Fig. 1 h-i that the phase shift of the temperature pulsations is absent within the whole range of change of the thermal conductivity $\alpha_{12}$ in the contact boundary. In particular, for the case of ideal contact $\alpha_{12}\to\infty$ at the chosen calculation conditions $L_k^1(0) = L_k^2(\xi) = \sqrt{D_1/D_2}\ \mathrm{sh}(\beta\sqrt{ik})$ and $\Delta_k = h_1\sqrt{ik}\ \mathrm{sh}(\beta\sqrt{ik})$. Therefore, $\Theta_1(t,x) = \Theta_2(t,\xi) = \Theta(t)$, where

$$\Theta(t) = \frac{2\Theta_0}{\pi} \sum_{k=1}^{\infty} \frac{\sin\psi_k}{k} \mathrm{Re} \frac{\exp\{i(\omega_k t - \psi_k)\}}{h_2\sqrt{ik}},\quad h_2 = \alpha^{-1}\sqrt{D_2\omega},$$

i.e., the form and amplitude of the temperature pulsations are independent of the film thickness $\xi$ and of its thermophysical characteristics. This allows us to assume (by analogy with the propagation of electromagnetic oscillations) that at frequencies less than $10^{-2}$ Hz the film becomes "transparent" for the temperature pulsations.

We performed the experimental investigations of the quasi-stationary thermal regime on the tungsten film ($\xi$=0.8 $\mu$m) - silicon substrate system using an YAG-laser LTI-502 operating under single-mode conditions ($\lambda$=1.06 $\mu$m, $t_u$ = 1.6 $\mu$sec, $t_n$ = 100 $\mu$sec, $\overline{P}$ = 16 W). The diameter of the heating spot was 30 $\mu$m. The mean temperature in the heating spot was kept constant and equal to $\overline{T}_1(0)$ = 1600 K. The measurement procedure of the mean temperature and the temperature pulsations in the heating spot $\Theta_1(t,0)$ was not different from that described in [8].

The basic problem associated with the application of indirect heating for annealing ion-implanted layers consists in finding the temperature of the silicon substrate at the contact boundary with the film $\overline{T}_2(\xi)$, because it determines the annealing kinetics. However, we may measure with a small error only the mean temperature and the temperature pulsations in the heating spot. As the value of $\overline{T}_2(\xi)$ at a given thermal conductivity $\alpha_{12}$ of the contact boundary is calculated simply from the values of $\overline{T}_1(0)$, the solution of the problem is reduced to the determination of $\alpha_{12}$ from the experimental relation $\Theta_1(t,0)$.



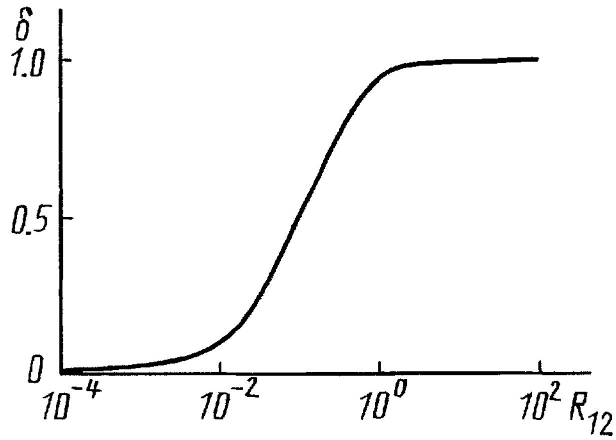



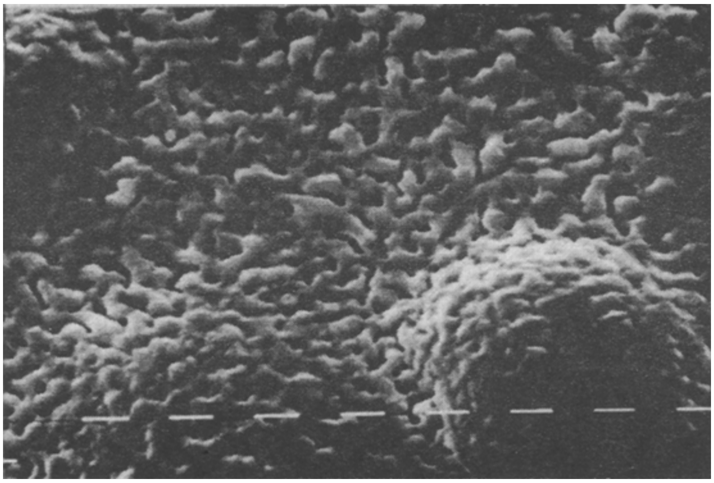

Fig. 2. The normalized form of the pulsation in the tungsten film (0.8 $\mu$m) - silicon substrate system at the film surface (a), at the film - to substrate (b), and at the substrate - to film (c) contact boundary: 1)$\alpha_{12}=0$; 2)$\alpha_{12}=10$ W/(cm$^2$·K); 3)$\alpha_{12}=\infty$; $\overline{T}=1600$K, $t_u = 1.6\,\mu$sec, $t_n = 100\,\mu$sec. t, $\mu$sec.

Fig. 3. Theoretical dependence of the relative pulsation area $\delta=S/S_0$ at the tungsten film surface on the magnitude of the contact boundary thermal resistance $R_{12}$: $S_0$ is the area of curve 1 in Fig. 2 a; S is the area of curves corresponding to variation in the thermal conductivity $0\leq\alpha_{12}<\infty$. $R_{12}$, cm$^2$·K/W.

To define the sensitivity of the form of the pulsation to the change in the value of $\alpha_{12}$, we perform a mathematical simulation of the quasi-stationary thermal regime. In the computations, the duration and repetition pulse period, as well as the mean temperature at the surface x = 0, corresponded to those chosen in the experiment. The data of [13, 14] were used for thermophysical constants. Figure 2 illustrates the computational results; here the values for the thermal conductivity of $\alpha_{12}=0$ and $\infty$ correspond to curves 1 and 3, while 10 W/(cm$^2$·K) to curves 2. From the figure it is evident that with decrease in the parameter $\alpha_{12}$, the quasi-stationary periodic-pulsed thermal regime [10] changes to the regular thermal regime of the second kind [11, 12]. At a pulse-repetition rate of 10 kHz, heat losses in the film at a thickness of $\xi=0.8\mu$m are small. Therefore, in the case of ideal contact at the surfaces x =0 and $x=\xi\pm0$, the mean temperatures and the form of the pulsation (curves 3) virtually coincide. During film peeling, the heat flux falling on the silicon substrate surface decreases by almost two orders and becomes equal to $q(t,\xi)=\sigma\varepsilon T_1^4(t,\xi)$. This leads to a sharp reduction of the mean substrate temperature $\overline{T}_2(\xi)$ with respect to the film temperature $\overline{T}_1(0) \simeq \overline{T}_1(\xi)$ and to a change in the form of the pulsation (curve 1 in Fig. 2c). According to Fig. 2a, in order to determine the sensitivity of the form of the pulsation $\theta_1(t, 0)$ to a change in $\alpha_{12}$, we may introduce the parameter $\delta = S/S_0$, which varies within the limits $0<\delta\leq1$. From formula (6) it follows that

$$S = \int_0^{t_n} [\Theta_1(t, 0) - \Theta_1(t_n, 0)]\, dt = -t_n\Theta_1(t_n, 0).$$

Hence, denoting $\Theta_1(t_n,0) = \Theta(\alpha_{12})$, we obtain the theoretical dependence for the parameter $\delta = \Theta(\alpha_{12})/\Theta(0)$. In the derivation of this dependence, it is more convenient to use, instead of $\alpha_{12}$, the inverse quantity which is equal to the thermal resistance of the contact boundary $R_{12}= \alpha_{12}^{-1}$. The character of the variation of $\delta$ from $R_{12}$ for the system under study is shown in Fig. 3. In the figure we may separate three regions of the change in $\delta$: the regions of existence of the stable quasi-stationary regimes $R^{12}\lesssim R_{12}^1=10^{-2}$ cm$^2$·K/W and $R_{12}\gtrsim R_{12}^2=1$



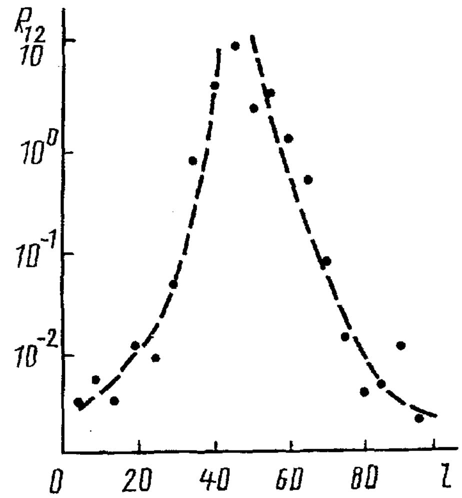



Fig. 4. Morphology of the tungsten film surface (0.8 $\mu$m) obtained by deposition on the silicon substrate (0.5 nm).

Fig. 5. Variation of contact boundary thermal resistance $R_{12}$ during scanning. $R_{12}$, $cm^2 \cdot K/W$; $l$, $\mu$m.

$cm^2 \cdot K/W$, and the transient region $R^1_{12} \lesssim R_{12} \lesssim R^2_{12}$ in which a sharp growth of $\delta$ occurs with increasing $R_{12}$. The values of $R_{12}$ may be calculated precisely from the form of the pulsation in the transient region. But as the relative measurement error of $\Theta_1(t,0)$ is of the $10^{-2}$ order, the range of the determinable values for $R_{12}$ under the given experimental conditions is within the limits of $10^{-4}-10^2$ $cm^2 \cdot K/W$. The tungsten film was deposited on a 0.5-mm-thick silicon substrate in a magnetron-type evaporator. The substrate surface was first polished and then cleaned in the argon plasma medium. Figure 4 shows the film surface relief obtained on the PSEM-500 scanning electron microscope . It is seen from this figure that the surface is of the "orange peel" structure with globe-shaped formations of diameter ~10 $\mu$m. The film was scanned by a laser beam with a scanning pitch of 5 $\mu$m and a time delay at each point of ~5 min necessary for establishment of the quasi-stationary thermal regime in the heating spot. The value of $\delta$ was determined in terms of the ratio of the pulsation area S to the pulsation area $S_0$ in the heating spot of the free tungsten film. Then, by the known dependence $\delta$ $(R_{12})$ (see Fig. 3), we calculated the thermal resistance $R_{12}$. Figure 5 shows the change in the thermal resistance when scanning the globe-shaped formation (Fig. 4). The character of the relation obtained allows us to assert that this formation has an internal cavity.

Thus, we have carried out investigations into the stable forms of the temperature pulsations arising during the periodic-pulsed heating of the film-substrate system. The possibility has shown for the quantitative estimation of the contact (adhesion) quality by measuring the form of the temperature pulsation in the heating spot.

## NOTATION

$q(t)$, absorption density of laser radiation power; $q_0$, radiation power density in a pulse; $\bar{q}$, power density of heat losses; $\bar{P}$, mean power of laser radiation; $\Theta_j$ and $\bar{T}_j$, temperature pulsations and mean temperature in the film (j = 1) and in the substrate (j = 2); $D_j=\lambda_j C_j \rho_j$, square of the thermal activity coefficient; $\alpha$, radiation heat loss coefficient; $\alpha_{12}$ and $R_{12}$, thermal conductivity and thermal resistance of the contact boundary; $c_k$, coefficients of the Fourier expansion function of $q(t)$; S and $S_0$, temperature pulsation area in the heating spot at values of thermal conductivity $0 \leq \alpha_{12} < \infty$ and $\alpha_{12}=0$, respectively.

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