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ОПРЕДЕЛЕНИЕ ПАРАМЕТРОВ ТЕПЛОПЕРЕНОСА В ТОНКОЙ ПОГЛОЩАЮЩЕЙ ПЛЕНКЕ НА ПОДЛОЖКЕ МЕТОДОМ ДИНАМИЧЕСКИХ РЕШЕТОК

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Продемонстрированы преимущества использования метода динамических решеток для бесконтактного изучения тонкопленочных материалов. Рассмотрены тонкие (микронные) пленки алмазоподобного углерода и термоэлектрические материалы на основе теллурида свинца. Установлено, что при поверхностном поглощении лазерного импульсного излучения наносекундной длительности в тонкопленочном материале формируется динамическая решетка, время релаксации которой позволяет определить параметры теплопереноса и рассчитать коэффициенты температуропроводности. Увеличение периода динамической решетки сопровождается переходом от измерения температуропроводности тонкой пленки к установлению эффективного значения температуропроводности, в которую начинает вносить вклад подложка, и при большом периоде решетки можно определять температуропроводность самой подложки. Особый интерес вызывает эффект возбуждения акустической волны в приповерхностном слое воздуха, который ранее рассматривался как фактор, искажающий динамику дифрагированного сигнала. Показано, что в этом случае появляется возможность измерить температуру пленки при ее нагреве лазерным импульсом.

Ключевые слова: голография; динамическая решетка; теплоперенос; температуропроводность; алмазоподобный углерод; теллурид свинца; термоэлектрик; акустическая волна.

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DETERMINATION OF HEAT TRANSFER PARAMETERS IN ABSORBING THIN FILMS ON SUBSTRATE BY THE TRANSIENT GRATING METHOD

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Advantages of the transient grating method for contactless study of thin-film materials have been demonstrated. Thin films of diamond-like carbon and thermoelectric lead telluride have been studied. Transient gratings are formed on a surface of film due to absorption of nanosecond pulsed laser radiation. The diffracted signal character enables the heat transfer parameters and thermal diffusivity determination. The transient grating period increase leads to transfer from thermal diffusivity of thin film measurements to its effective value determination, with a contribution made by the substrate. At large period of the grating one can realise measurements of thermal diffusivity of the substrate itself. Acoustic wave excitation in the near-surface thin layer of the air, which earlier has been considered as a factor distorting the diffracted signal dynamics is of special interest. In this case there is a possibility to temperature measurement of the film when it is heated by a laser pulse.

Keywords: holography; transient grating; heat transfer; thermal diffusivity; diamond-like carbon; lead telluride; thermoelectric; acoustic wave.

Introduction

Studies of the features of heat transfer in thin films are of topical importance in the field of micro- and nanoelectronics, material science. Examination of materials and measurements of the heat transfer characteristics are conducted with the use of different physical methods, among which contactless procedures using optical radiation are the most attractive approaches [1–6].

The paper presents optical studies of thermal properties of diamond-like carbon and lead telluride based thermoelectric. These materials are synthesised to find numerous applications in various branches of science and technology including the formation of protective films, manufacturing of heat-conducting coatings, development of solar power engineering, etc. Consideration is given to a thin film that is in a double sided thermal contact with the air and the substrate.

According to the method used, two coherent light beams of a pulsed laser are interfering on the surface of a sample under study. Heating of the excitation area results in the formation of a surface-relief grating. First-order diffraction of a light beam from the continuous-wave laser by the created grating is observed. The diffraction signal intensity is photometrically measured by a detector of high temporal resolution. The lifetime of a transient grating depends on its spatial period and thermal diffusivity of film under study.

When the thermal grating lifetime is measured, one can have information about physical processes proceeding in the material under study. In particular one can measure relaxation time of thermal grating and the thermal diffusivity of film studied. All the experiments have been performed with the use of a system, shown in a series of works [7–9].

Influence of the thermal contact between substrate and film on the in-plane heat transfer

Free standing thin films growth and their subsequent usage is rarely realised in practice because of range of technological problems. In this paper the heat transfer is studied in films that are in thermal and mechanical contacts with substrate. Pulsed heating of the film is of essentially surface character and the initial penetration depth of exciting radiation should be much less than film thickness. Heat transfer takes place in the directions of the thermal gradient formed, i. e. parallel and normal to sample surface.

In the process of the in-plane thermal diffusivity χ_x measuring, the depth of heat penetration L_{th} into a film of thick h in the direction normal to the sample surface during relaxation of the transient grating is a parameter of great importance. For reliability of the measurements of χ_x, it is necessary that the condition $L_{\text{th}} < h$ is well satisfied. We have $L_{\text{th}} = \sqrt{4\chi_z \tau_x}$ [10–12], where χ_z – thermal diffusivity in the direction normal to the film surface, τ_x – relaxation time, $\tau_x = \dfrac{\Lambda^2}{4\pi^2 \chi_x}$, Λ – grating period. Most commonly $\chi_z = \chi_x$ (thermal isotropy) and in this case the influence of substrate on kinetics of thermal relaxation is insignificant if the thermal grating period $\Lambda < \pi h$.

As heat penetrates deeper into the sample, amplitude of the thermal profile is lowered due to thermal diffusion in the in-plane direction x. Naturally, relaxation kinetics in the case, when a film with high thermal diffusivity is formed on the substrate with a relatively low thermal diffusivity, is mainly determined by the in-plane heat transfer in film.

When the period Λ of a thermal grating, and hence its life time τ_x, are large enough and when during the period of the diffracted signal observation heat has a chance to reach the substrate and to exert its thermal effect, we measure the effective value averaged over the heat penetration depth instead of a real thermal diffusivity of film. In this case kinetics of thermal relaxation is weakly dependent on the film properties and in fact is determined by thermal parameters of the substrate itself.

A transformation the effective thermal diffusivity (χ_{eff}) of the film under study to its actual value is illustrated in fig. 1, showing the result of excitation of thermal transient gratings with different periods in a film of diamond-like carbon (DLC). The grating is excited by laser pulses of width 10 ns at the wave length 532 nm. The radiation absorption factor μ of the film in this spectral range is $\approx 10^5$ cm^{-1}.

It is well seen that when the grating period Λ decreases from 20 to 2 μm, the in-plane thermal diffusivity χ_x is varying from thermal diffusivity of the glass substrate to the same parameter of diamond-like carbon film [13]. If $\Lambda \gg h$, there is practically no heat transfer in direction parallel to sample surface, whereas the decay of a grating is mainly determined by heat transfer in the substrate.

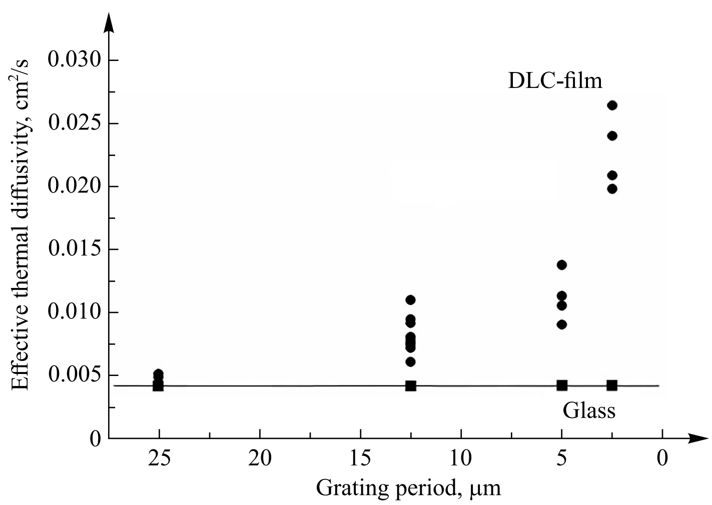

Fig. 1. Effective thermal diffusivity of the system *diamond-like carbon film – glass* as a function of the period of a thermal transient grating formed on surface of the absorbing film of thick 2 μm

Role of the air in relaxation kinetics formation of transient surface grating lead telluride film

Lead telluride (PbTe) based thermoelectrics are now actively studying because this narrow-band semiconductor shows promise for the technologies of renewable energy engineering. Recently the attention has been attracted to lead telluride doped with indium or bismuth that reveals advantageous functional qualities. Some portion of thermal energy stored by the absorbing surface of a PbTe film is transferred to the adjacent air layer and acoustic grating is excited in thin layer.

To estimate a contribution of the air into kinetics of the diffraction signal attenuation, one should take into consideration that, according to the data from [14] obtained by the photorefraction method, thermal diffusivity of the air on normal atmospheric pressure comes to 0.2–0.4 cm^2/s depending on its temperature. The same parameter for the tested film is within the range 0.015–0.025 cm^2/s [15]. Consequently, the life time of a thermal grating in air is one-tenth as long as the grating in film under study. Thus, to exclude the influence of a thermal grating in the air on the measurement results during experimental studies of heat transfer in samples based on PbTe, the initial point of the analysed kinetics should be shifted by lifetime of air grating after the moment when the diffraction signal arises.

As it seen in fig. 2, *a*, at the initial part of the kinetics curve one can observe an acoustic attenuating component of diffraction. Ultrasonic oscillations period is about 0.12 μs.

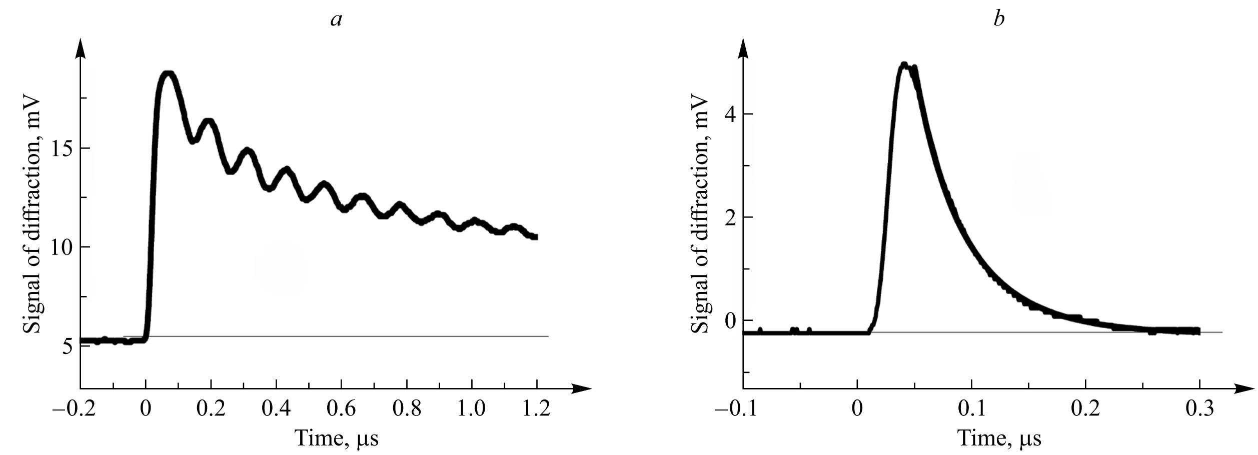

Fig. 2. Diffraction kinetics of a probe beam for the film of PbTe : Bi thick 2 μm on glass. The grating periods are 41.7 μm (*a*) and 2.5 μm (*b*)

It has been found experimentally that when the spatial grating period is below 5 μm, no acoustic oscillations are observed due to not enough temporal resolution of a recording system (fig. 2, *b*). Indeed, for $\Lambda = 2.5$ μm, the period of acoustic oscillations in the air is 9 ns which is much less then temporal resolution of the detection system used. Figure 2, *b*, shows that relaxation time of a dynamic grating comes to 0.13 μs. Then, for the grating period 2.5 μm, thermal diffusivity of the film comes to $0.93 \cdot 10^{-2}$ cm^2/s.

Ultrasonic estimation of PbTe : Bi film temperature

The possibility to measure frequency of an ultrasonic wave as excited by of the transient grating method offers estimation of the speed of sound in a near-surface layer of the air. Considering that the ultrasound propagation speed in the air is linearly growing with temperature over the range up to 500 K [16], the proposed method may be used for estimation of the film temperature at the initial moment of dynamic grating recording. It is assumed that temperature of a thin near-boundary layer of the air in intimate contact with the heated surface of the film is close to temperature of the film surface.

Frequency of the diffracted signal acoustic oscillations has been found by means of the fast Fourier transformation (fig. 3, *a*), and the propagation speed of ultrasound in air $V = \Lambda f = 347$ m/s has been calculated. By using the graph in fig. 3, *b*, it has been found that the film temperature at $V = 347$ m/s is approximately 310 K. This value should be taken as an average over the probing spot.

As the energy of laser pulses which record the grating grows by a factor of 1.5, the propagation speed of ultrasound increases up to 368 m/s. According to fig. 3, *b*, of the near-boundary layer temperature of the air and hence of the film increases to 330 K.

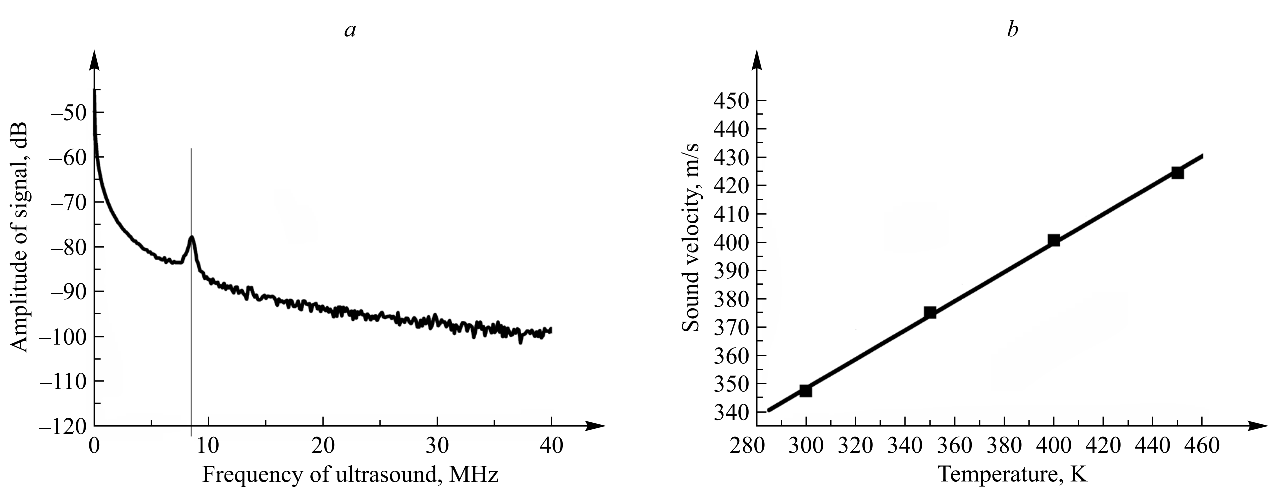

Fig. 3. PbTe : Bi film: Fourier spectrum of the diffracted signal for PbTe : Bi film
(vertical line denotes the component of acoustic oscillations
in air at frequency 8.32 MHz) (*a*)
and temperature dependence of the ultrasonic speed in air
(■ is reference points [16]) (*b*)

To improve reliability of the results obtained during the conducted measurements, the film temperature has been also estimated with regard to the fact that the diffraction efficiency η of grating is determined by the amplitude of the formed surface thermal relief. For the 100 % contrast of interference pattern in the film under study, this amplitude is completely determined by all the absorbed energy of laser radiation. This excitation mode allows for calibration of the diffraction efficiency of a relief transient grating and for its evaluation.

As it is demonstrated by the experiments, in a linear mode of the dynamic grating recording its initial amplitude indicated on the oscilloscope screen should not to exceed 7–8 mV. To find which diffraction efficiency of the grating is associated with this amplitude, the probe beam was calibrated with the use of a set of neutral optical filters, preliminary certified for transmission. The number of filters was selected so that the signal amplitude on the oscilloscope screen was the same as the diffraction-signal amplitude recorded. The overall transmission was found to be 10^{-5}. The diffraction efficiency of the relief-reflection type grating for diffraction efficiency $\eta \ll 1$ is given by the well-known relation [17]:

$$\eta = \left(\frac{\pi \Delta h}{\lambda}\right)^2. \quad (1)$$

Here $\Delta h = l \Delta T \beta$, where Δh surface relief height, l – heated layer depth taken equal to the film thickness 2 μm, $\beta = 20 \cdot 10^{-6}\ \mathrm{K}^{-1}$ – thermal linear-expansion coefficient of lead telluride, λ – wavelength of probe radiation. According to (1), for $\eta \sim 10^{-5}$ and the probe-radiation wave length 635 nm, the thermal relief height comes to $\Delta h \sim 0.7$ nm. Then, accordingly to the equation (1), the temperature increase of the film due to absorption of pulsed laser radiation above the room temperature ΔT, averaged over the depth and the excitation spot, is about 30–40 °C. This is in a good agreement with the result obtained through ultrasonic application.

Conclusions

In this way the experiments with thin films of diamond-like carbon and PbTe based thermoelectric have demonstrated the potentialities for application of the transient grating method in contact-free testing of heat transfer in thin-film materials. It has been shown that with surface radiation absorption and with the grating period Λ less than film thickness, relaxation time of a transient grating is determined by thermal diffusivity of film in the direction along the film surface. Rising up the value of Λ is accompanied by transition to the effective value of thermal diffusivity of the system film substrate. If the period Λ much greater than the film thickness we can go to measurements of thermal diffusivity of the substrate. Of particular interest is the acoustic wave identification in the near-surface layer of the air. It offers a unique possibility to measure the frequency and speed of ultrasound, enabling one to film temperature estimation at the moment of it heating by a laser beam radiation.

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