Thermal diffusion of a two-layer system

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Thermal conductivity and thermal diffusivity of a two-layer system are examined from the theoretical point of view. We use the one-dimensional heat-diffusion equation with the appropriate solution in each layer and boundary conditions at the interfaces to calculate the heat transport in this bounded system. We also consider the heat flux at the surface of the sample as a boundary condition instead of using a fixed temperature. From this, we obtain an expression for the effective thermal diffusivity of the composite sample in terms of the thermal diffusivity of its constituent materials without any approximations.

I. INTRODUCTION

During the last decade, several methods have been developed to determine thermal diffusivities and conductivities with high precision by means of photothermal effects.[1] The most widely used method is based on the photoacoustic effect. The principle of the effect is that when a sample in a closed cell is illuminated by light modulated or chopped at audio frequencies, an acoustic signal is produced. The application of the photoacoustic effect to the measurement of thermal diffusivities for thin films has been made by Adams and Kirkbright.[2] They have used the method called rear-surface illumination. When the rear surface of a sample is illuminated with the chopped light beam, heat oscillations generated therein propagate from the surface into the sample. Pressure oscillations of the same frequency are induced in the gas chamber by the temperature oscillations at the surface interface between the sample and the gas, where they can be detected by a microphone. The photoacoustic signals obtained have a certain phase shift relative to the signal detected from front-surface excitation. Besides that, since the phase shift of the signal does not depend on the optical properties of the sample, it is simpler to extract information on the thermal diffusivity from the experimental results. Phase shift lag measurements are then most suitable for determining the thermal diffusivity of the material. Charpentier et al.[3] made an analysis of the pressure variations considering rear- and front-surface excitation and gave a formula relating the lag shift of the photoacoustic signal using high modulation frequency. The refinement of the theory in terms of the Rosencwaig and Gersho theory[4] was given by Pessoa et al.;[5] they showed that the relative phase lag exhibits no explicit dependence on the absorbed power and surface conditions so that a single modulation frequency measurement is sufficient to determine the thermal diffusivity.

In recent years, there has been some interest in studying the thermal characterization of two-layer systems of variable thickness using the photoacoustic effect. Tomi-

naga and Ito[6] used the Rosencwaig-Gersho model for a two-layer system under rear illumination and looking at the phase angle behavior as a function of the modulation frequency. They showed that, at high modulation frequencies, the rear-illumination phase angle depends upon a critical frequency above which one of the materials becomes thermally thick. From the analogy between thermal and electrical resistances widely used in heat-transfer problems,[7] Mansanares et al.[8] calculated the effective thermal diffusivity of the two-layer system as a function of the filling fraction of the composite sample and the ratio of the thermal conductivities of each material. Recently Christofides and Seas[9] made an extension of the theoretical model of photopyroelectric spectroscopy of solids[10] to investigate the optical and thermal properties of a two-layer sample. They examined the case where the optical absorption coefficients of the substrate and film vary in different ways; several computer simulations were performed in order to examine the correctness of this model for a wide range of wavelengths and modulation frequencies.

In the present paper, we take a different approach than previous investigations to the heat transport in bounded systems. We restrict our analysis to the case of solution of the heat transport equation considering only the continuity of the heat flux at the interface of the two-layer system. In addition, we only take into account the decreasing exponential term of the dynamical part of the temperature fluctuation in each layer, which gives the physical solution of the heat flux. It is shown that if the ratio of the square root of the diffusivity is equal to the ratio of the thermal conductivity of each layer, then the temperature distribution is continuous at the interface; otherwise the temperature is discontinuous. In both cases, the effective diffusivity of the two-layer system is the same.

II. ONE-LAYER SYSTEM

It is well known that heat transport in solids is carried out by various quasiparticles (electrons, holes, phonons,

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magnons, plasmons, etc.). Frequently the interactions between these quasiparticles are such that each of these subsystems can have its own temperature and the physical conditions at the boundary of the sample can be formulated separately for each temperature. For example, the physical conditions resulting in heat transport by electrons and phonons are given by Granovski and Gurevich[11] and those for the transport by electrons and magnons are given in Ref. 12, where the appropriate boundary conditions are formulated. The theory of heat conduction in solids was developed by Gurevich and Kaganov[13] using the two-temperature approximation. They showed that, in general, the temperatures of carriers and phonons in anisotropic semiconductors are unequal even in the interior of a bulk sample. The expression for the temperature distribution for electrons and phonons was obtained in bounded semiconductors using adiabatic boundary conditions. It is shown that the accepted assumptions about the constancy of the temperature gradient are only valid under certain limits.[14]

We restrict ourselves for definiteness to the case when the quasiparticle systems are electrons and phonons. Let T_p be the characteristic phonon temperature. Then the momentum $q \propto \frac{T_p}{s}$ (s is the sound velocity) sets a limit to the phase space volume occupied by phonons. Hereinafter, we shall consider two limiting cases.[15]

(i) Short wavelength (SW) phonons occupy a large volume in phase space. This case is represented by the the inequality

$$2p \ll \frac{T_p}{s}, \tag{1}$$

where p is the average electron momentum, namely, $p \propto \sqrt{2mT_e}$ for the nondegenerate electron gas and $p = p_f$, the Fermi momentum, for the degenerate electron gas; here T_e is the electron temperature.

(ii) Large wavelength (LW) phonons occupy a large volume in phase space. This is the case when electrons interact with all phonons,

$$2p \gg \frac{T_p}{s}. \tag{2}$$

From the Boltzmann equation, it is known that the degree of nonequilibrium of the phonons is determinated by the relationship between phonon-electron (ν_{pe}) and phonon-phonon (ν_{pp}) relaxation frequency. ν_{pe} determines the degree of phonon disturbance by the electrons and it decreases rapidly, tending to zero for $q > 2p$, while the other, ν_{pp}, describes the tendency of phonons to come to equilibrium as a result of the energy distribution. Therefore, in the region $q > 2p$, only the following inequality holds:

$$\nu_{pp} \gg \nu_{pe}, \quad q > 2p. \tag{3}$$

In the region $q < 2p$, the inequality

$$\nu_{pp} \gg \nu_{pe}, \quad q < 2p \tag{4}$$

can hold as well as the reverse inequality

$$\nu_{pp} \ll \nu_{pe}, \quad q < 2p. \tag{5}$$

In the limit $\nu_{pp} \gg \nu_{pe}$, the phonon-phonon interactions are more frequent than phonon-electron collisions and more efficient in terms of energy relaxation than energy transfer from the electron to the phonon subsystem. It should be noted that the electron and phonon subsystems, generally speaking, cannot be characterized by a single temperature. Therefore steady state heat conduction can be described by the following system of equations:

$$\text{div}\mathbf{Q}_e = P_{ep}(T_e - T_p), \quad \text{div}\mathbf{Q}_p = -P_{pe}(T_e - T_p) . \tag{6}$$

The term $P_{ep}(T_e - T_p)$ describes the transfer of heat between electrons and phonons. Here P_{ep} is a parameter proportional to the frequency of phonon-electron collisions ($P_{ep} = P_{pe}$) and the heat flux of electron $\mathbf{Q}_e$ and phonon $\mathbf{Q}_p$ subsystems are described by the usual relationships

$$\mathbf{Q}_e = -k_e \text{div}T_e, \quad \mathbf{Q}_p = -k_p \text{div}T_p , \tag{7}$$

where k_e (k_p) is the electron (phonon) thermal conductivity. If in addition, we consider large specimens such that the dimension $l \gg L^*$, where L^* is the scale of the electron-phonon energy interaction, referred to as the cooling length then the temperatures of the two subsystems are equal, i.e., $T_e = T_p = T$.[14] In this case we obtain, after summing Eqs. (6), the following equation:

$$\text{div}\mathbf{Q} = 0 , \tag{8}$$

where $\mathbf{Q} = \mathbf{Q}_e + \mathbf{Q}_p$ is the total heat flux carried by electrons and phonons.

Now, consider the situation represented by the inequality of Eq. (5). This is the case when phonon-phonon collisions alone cannot bring the phonon subsystem to an internal equilibrium. If Eq. (1) is true, i.e., LW phonons occupy a much smaller phase volume than SW phonons, then the SW system has enough time to redistribute the energy received from the LW phonons between its constituent quasiparticles. As a result, the distribution function of SW phonons becomes Planckian (see Ref. 15). Then the electron–LW phonon interactions relax their energy more efficiently than the phonon subsystem and the LW phonons emitted by electrons of temperature T_e are characterized by the same temperature $T_e = T_p^{\text{LW}}$. In this situation we also have two different subsystems; one corresponds to the SW phonons with temperature T_p^{SW} and the other one corresponds to electrons and LW phonons with a characteristic temperature $T_e = T_p^{\text{LW}}$, and they satisfy the following heat transport equations:

$$\text{div}(\mathbf{Q}_e + \mathbf{Q}_p^{\text{LW}}) = -P_{pp}(T_e - T_p^{\text{SW}}),$$
$$\text{div}\mathbf{Q}_p^{\text{SW}} = P_{pp}(T_e - T_p^{\text{SW}}) , \tag{9}$$

where the term $P_{pp}(T_e - T_p^{\text{SW}})$ represents the transfer of heat from LW phonons to SW phonons and P_{pp} is calculated in Ref. 16. If the size of the sample is greater than the cooling length ($l \gg L^*$) of this system then we

obtain the condition $T_e = T_p^{SW} = T$, and after summing Eqs. (9) the total heat flux satisfies div$\mathbf{Q} = 0$.

When $\nu_{pe} \gg \nu_{pp}$ and Eq. (2) holds, we have only LW phonons and they interact efficiently with electrons. In this case all the quasiparticle systems have the same temperature $T_e = T_p = T$ and the equation for heat flux is similar to Eq. (8).

From this brief discussion, we have shown that under certain conditions on the realaxation frequency of the electron and phonon subsystems and size of the sample the total system can be described by the same temperature T and the total heat flux satisfies div$\mathbf{Q} = 0$.

So far, we have only described the static contribution of the heat transport, i.e., the heat flux is independent of time. However, in the photoacoustic experiments, the incident radiation is modulated in time by the chopper, and in this case it is necessary to consider the dynamic contribution to the heat transport in the electron and phonon systems. It is worth mentioning that even the external perturbation depends on time, the dynamic contribution of the heat flux is only important when the frequency ω of the incident electromagnetic field is of the same order as the characteristic relaxation energy frequency of the electronic system, ν_ε.[17] If $\omega \gg \nu_\varepsilon$ (high frequency limit), the electron temperature cannot follow the variation of the field and assumes an average value. Since phonons can receive energy only from electrons, the phonon temperature should also remain constant as a function of time. Otherwise, if $\omega \ll \nu_\varepsilon$ (low frequency limit), the variation of the electron and phonon temperatures is quasistatic. That means that the static quasiparticle temperature oscillates with the same frequency as the radiation.

In Eq. (8), we are not taking into account the distributed heat source resulting from the light absorption. Assuming that the temperature of the electron and phonon systems are equal, which is usually the condition in most of the photothermal experiments, the equation of heat conduction in solids (that is valid for $\omega \le \nu_\varepsilon$) can be written as

$$\frac{\partial T(\mathbf{r},t)}{\partial t} = \alpha \nabla^2 T(\mathbf{r},t). \tag{10}$$

In this equation, we are considering that the variation of the temperature as a function of x is such that the heat conductivity k is independent of the coordinates; otherwise, we have to solve a nonlinear heat equation. Here the diffusivity α is given as $\alpha = \frac{k}{\rho c}$ where ρ is the density and c is the specific heat of the sample. Consider the photoacoustic cell geometry for the heat transmission configuration shown schematically in Fig. 1(a). The temperature fluctuation is obtained from the solution of Eq. (10) in one dimension. The solution $T(x,t)$ should be supplemented by boundary conditions at $x = 0$. In the photoacoustic experiments, the most common mechanism to produce thermal waves is the absorption by the sample of an intensity modulated light beam with frequency modulation $\omega \le \nu_\varepsilon$. It is clear that when the intensity of the radiation is fixed, the light-into-heat conversion at the surface of the sample can be written as

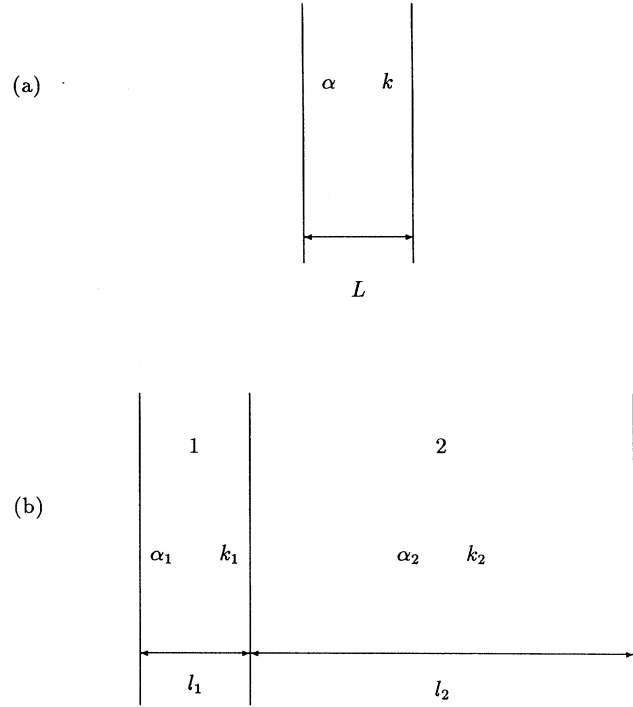$$Q(x,t)|_{x=0} = Q + \Delta Q e^{i\omega t} , \tag{11}$$

FIG. 1. Geometry for (a) a one-layer system and (b) a two-layer system.

where Q is proportional to the intensity of high frequency light ($\Omega \gg \nu_\varepsilon$) and the other term represents the modulation of this light. The temperature is not used as a boundary condition because it is usually an unknown parameter in the experiments, and besides that it is necessary to know the temperature on both surfaces. It is only important in thermoelectric phenomena in semiconductors when the specification of the temperatures on the surfaces of the sample must be known.

The general solution of the heat-diffusion equation for a one-layer system is given by

$$T(x,t) = T_0 + T_1 x + T_2 e^{i\omega t - \sigma x}. \tag{12}$$

The parameter σ is determined by forcing Eq. (12) to satisfy Eq. (10) for one-dimensional heat flux and is equal to $\sigma = (1 + i)\sqrt{\omega/2\alpha}$. Using the boundary condition at $x = 0$, the constants T_1 and T_2 are given by

$$T_1 = -\frac{Q}{k}, \quad T_2 = \frac{\Delta Q}{2k}\left[\frac{2\alpha}{\omega}\right]^{1/2}(1 - i). \tag{13}$$

In arriving at Eq. (12), we assume that the sample is optically opaque to the incident light (i.e., all the incident light is absorbed at the surface). Here T_0 is a constant which cannot be determined from these boundary conditions and it is not important in obtaining the physical results. It is worth mentioning that the increasing exponential term with distance in the dynamical part of Eq. (12) has not been considered because this term represents a macroscopic heat flux from the lower to higher temperature region (heat flux cannot be reflected).

Once we know the temperature distribution in the sample, we can assume the acoustic piston model for evaluating the pressure fluctuation in the cell. Acording to this model,[4] the oscillating component of the temperature attenuates rapidly to zero with increasing distance from the sample surface. The thin gas boundary layer at the interface then acts as a vibrating piston. The displacement of this piston is estimated using the ideal gas law $(PV = Nk_BT)$ for the boundary layer. As a result of this gas piston oscillation, a pressure fluctuation $\delta P(t)$ is produced in the cell and is given by

$$\delta P(t) = \frac{P_0}{T_0}\frac{\Delta Q}{2k}\left(\frac{2\alpha}{\omega}\right)^{1/2}\left[\cos\left(\omega t - \sqrt{\frac{\omega}{2\alpha}}L\right) + \sin\left(\omega t - \sqrt{\frac{\omega}{2\alpha}}L\right)\right]\exp\left(\frac{\omega}{2\alpha}\right)^{1/2}L \,, \quad (14)$$

where P_0 and T_0 are the ambient pressure and temperature, respectively. Then Eq. (14) may be evaluated for the magnitude and phase of the acoustic pressure wave produced in the cell by the photoacoustic effect.

III. TWO-LAYER SYSTEM

Let us consider the two-layer system shown schematically in Fig. 1(b) consisting of a material 1 of thickness l_1 and a material 2 of thickness l_2, both having the same cross section. Let $L = l_1 + l_2$ denote the total sample thickness, α_i the thermal diffusivity, and k_i the thermal conductivity of the material i $(i = 1,2)$. The system of heat-diffusion equations describing the heat transfer through the various layers of the one-dimensional photoacoustic configuration is given by

$$\frac{\partial T_i}{\partial t} = \alpha_i\frac{\partial^2 T_i}{\partial x^2}. \quad (15)$$

The boundary conditions of the thermal-diffusion equation (15) are obtained from the requirement of heat flux continuity at the interfaces of the two materials and Eq. (11). The solutions of the heat transport equations can be written as

$$T_1(x,t) = T_0 - \frac{Q}{k_1}x + \frac{\Delta Q}{2k_1}(1 - i)\left(\frac{2\alpha_1}{\omega}\right)^{1/2}e^{i\omega t - \sigma_1 x},$$

$$0 < x < l_1, \quad (16)$$

$$T_2(x,t) = \theta_0 + \theta_1(x - l_1) + \theta_2\exp\left[i\omega t - \sigma_2(x - l_1)\right],$$

$$l_1 < x < L, \quad (17)$$

where

$$\theta_1 = -\frac{Q}{k_2} \,, \quad (18)$$

$$\theta_2 = \frac{\Delta Q}{2k_2}(1 - i)\left(\frac{2\alpha_2}{\omega}\right)^{1/2} \times \exp\left[-\left(\frac{\omega}{2\alpha_1}\right)^{1/2}(1 + i)l_1\right] \,, \quad (19)$$

with

$$\sigma_i = \left(\frac{\omega}{2\alpha_i}\right)^{1/2}(1 + i). \quad (20)$$

Comparing the temperature distribution Eq. (17) with Eq. (12) at $x = L$, we can write the effective thermal diffusivity and conductivity of the two-layer system as

$$\frac{L}{\sqrt{\alpha}} = \frac{l_1}{\sqrt{\alpha_1}} + \frac{l_2}{\sqrt{\alpha_2}} \quad (21)$$

and

$$\frac{\sqrt{\alpha}}{k} = \frac{\sqrt{\alpha_2}}{k_2} \quad (22)$$

or

$$\frac{L}{k} = \frac{\sqrt{\alpha_2}}{k_2}\left(\frac{l_1}{\sqrt{\alpha_1}} + \frac{l_2}{\sqrt{\alpha_2}}\right). \quad (23)$$

It is worth mentioning that the effective thermal parameters Eqs. (21)–(23) have been obtained without any approximations about the thermal thin and thickness of the materials,[2] analogy between thermal and electrical resistances used in heat-transfer problems,[4] some critical frequencies above which one of the layers becomes thermally thick,[5] or continuity of the temperature distribution at any interface.

However, if the thermal parameters of each layer satisfy

$$\frac{\sqrt{\alpha_1}}{k_1} = \frac{\sqrt{\alpha_2}}{k_2} \quad (24)$$

and using Eq. (23), the effective thermal conductivity can be written as

$$\frac{L}{k} = \frac{l_1}{k_1} + \frac{l_2}{k_2} \,. \quad (25)$$

Then, from Eqs. (16) and (17) we obtain that $T_1(l_1, t) = T_2(l_1, t)$, i.e., the temperature is continuous at the interface $x = l_1$; otherwise, in general, the temperature will be discontinuous at the interface of the two materials and the effective thermal conductivity and diffusivity are given by Eqs. (21) and (23). This discontinuity in the temperature can be expressed mathematically as follows:

$$Q(x,t)|_{x=l_1} = \lim_{\varepsilon\to 0} -k\frac{T_2(l_1 + \varepsilon) - T_1(l_1 - \varepsilon)}{\varepsilon}$$
$$= \eta(T_2 - T_1) \,, \quad (26)$$

where η is the surface thermal conductivity at the interface.[14] Note that when η goes to infinity, since Q is finite, the temperature distribution must be continuous at the interface, and for finite surface thermal conductivity, in general, we have that $T_1(x,t)|_{x=l_1} \neq T_2(x,t)|_{x=l_1}$.

IV. CONCLUSIONS

A theoretical analysis of the photoacoustic effect in a two-layer sample has been studied. Using the appropriate boundary conditions, we obtain the effective thermal diffusivity and thermal conductivity of the two-layer system without any approximation in the thermal parameters. The continuity or discontinuity of the temperature at the interface of the two-layer system depends on the relationship of the thermal parameters of both layers. In general, the heat flux is defined as the product of the thermal surface conductivity and the difference of the temperatures at the interface.

Mansanares et al.[8] demonstrated the usefulness of a single modulation frequency method for measuring the thermal diffusivity of solid samples. The method consists of measuring the relative phase between the rear-surface illumination and the front-surface illumination. Using the thermal-diffusion model of Rosencwaig and Gersho[4] for the production of the photoacoustic signal, the ratio of the signal amplitude and the phase lag for rear- and front-surface illumination is given as a function of the sample thickness and the sample thermal-diffusion coefficient. The theory for the relative phase lag will be studied using our model in a forthcoming paper. Finally, its important to mention that our model is valid for modulated frequency of the incident light, ω, of the same order as the frequency of the relaxation energy between the quasiparticle systems ν_ϵ. In the limit $\omega \gg \nu_\epsilon$ the system cannot respond to this external perturbation; therefore the average in time of the dynamical part of the heat flux is negligible and the transferred heat is only static. For $\omega \ll \nu_\epsilon$, the heat flux is quasistatic, i.e., $\partial T/\partial t = 0$.[17] Then the solution of Eq. (10) for one layer is given by $T(x,t) = C_1 + C_2 x$ and from Eq. (11) we finally obtain the solution in this regime as $T(x,t) = (C_1' + C_2' x)e^{i\omega t}$. This represents an oscillation of the temperature distribution in every point of the layer.

ACKNOWLEDGMENT

[*] On leave from the Institute for Radiophysics and Electronics of the Academy of Sciences of Ukraine, Kharkov, 310085, Ukraine.

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