

Harmonic heat flow in isotropic layered systems and its use for thin film thermal conductivity measurements

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A theoretical model is presented describing harmonic heat flow in a two layer system heated by a modulated Gaussian laser beam. Amplitude and phase of the modulated temperature rise in the layers, as well as in the backing substrate and adjacent atmosphere, are calculated by solving the three-dimensional heat conduction equation with a source term including exponential absorption of the laser light in one or two layers. Heat conduction is assumed to be isotropic throughout the system, however, a thermal contact resistance between the two layers can be taken into account. Results are presented for single and double layer systems of gold and various dielectric thin film materials on glass substrates. Amplitude and phase of the harmonic temperature variation are calculated either as a function of position in the sample system or at the surface as a function of the laser beam modulation frequency. It is found that both amplitude and phase of the calculated temperature rise exhibit typical thin film features in their frequency dependence, however, the phase is more sensitive to thin film phenomena than the amplitude. The phase shows typical extrema in that frequency region, where the thermal diffusion length in the film is equal to the film thickness. Based on these findings, we demonstrate how these calculations can be utilized for the interpretation of thin film thermal parameter measurements. The influence of thermal wave interference is demonstrated, and it is discussed how the main thermal parameters like conductivity, effusivity, and thermal contact resistance of the thin film system can be extracted from measurements by a fit of theoretical curves to experimental data. Applying a simple one-dimensional thermal expansion model, surface displacements for thin film systems are calculated and the applicability of photothermal surface displacement for thin film conductivity measurements is discussed.

I. INTRODUCTION

The measurement of thin film conductivities is a long standing problem in many areas of physics and engineering. As one example, we mention the interest in thermal conductivities of dielectric thin film materials used for optical coatings.¹ Another important field of interest is films used in thermal management applications with semiconductor circuits.² Recently, such measurements gained a tremendous interest in connection with the development of thin film diamond-like materials promising excellent thermal properties.³⁻⁶

The experimental methods used for the determination of thin film conductivities may roughly be divided into two classes: first, methods involving a direct contact between a heating element and a thermocouple with the sample; and second, contactless photothermal techniques where the heating is provided by the absorbed light of a pump laser beam and the temperature is measured either directly by radiation or via secondary effects probed by a second laser beam. The main advantage of the first class of experiments^{2,7-11} is their conceptual simplicity leading to a direct result that may often be interpreted without elaborate theoretical modeling. However, contacting thin film samples often bears the risk of damage and change in sam-

ple properties yielding misleading results for thermal parameters.

These risks are avoided by photothermal techniques. On the other hand, data interpretation for this kind of measurement very much relies on a precise theoretical model for the thermal transport in the sample, since results are obtained in an indirect way. Therefore, it is one intention of this paper to provide a sound theoretical basis for the interpretation of photothermal conductivity measurements on simple thin film systems applying modulated laser beams.

Thermal conductivity measurements have been performed under steady state conditions; i.e., by measuring the temperature equilibrium established by continuously heating with a cw laser beam and cooling by heat conduction and radiation to the surroundings.¹² Steady state calculations have been presented for simple bulk materials,^{13,14} as well as complicated multilayer coating systems.^{15,16}

By far the most photothermal conductivity measurements reported in literature are based on modulated techniques, where the amplitude or more often the phase of a periodic temperature variation on the sample surface induced by a harmonically or square wave modulated cw laser beam is monitored utilizing photothermal effects like radiometry,¹⁷ mirage effect,¹⁸⁻²⁰ thermoreflectance,²¹ surface displacement,^{21,22} interferometry,²³ or pyroelectric detection.²⁴

The main advantage of modulated techniques is the

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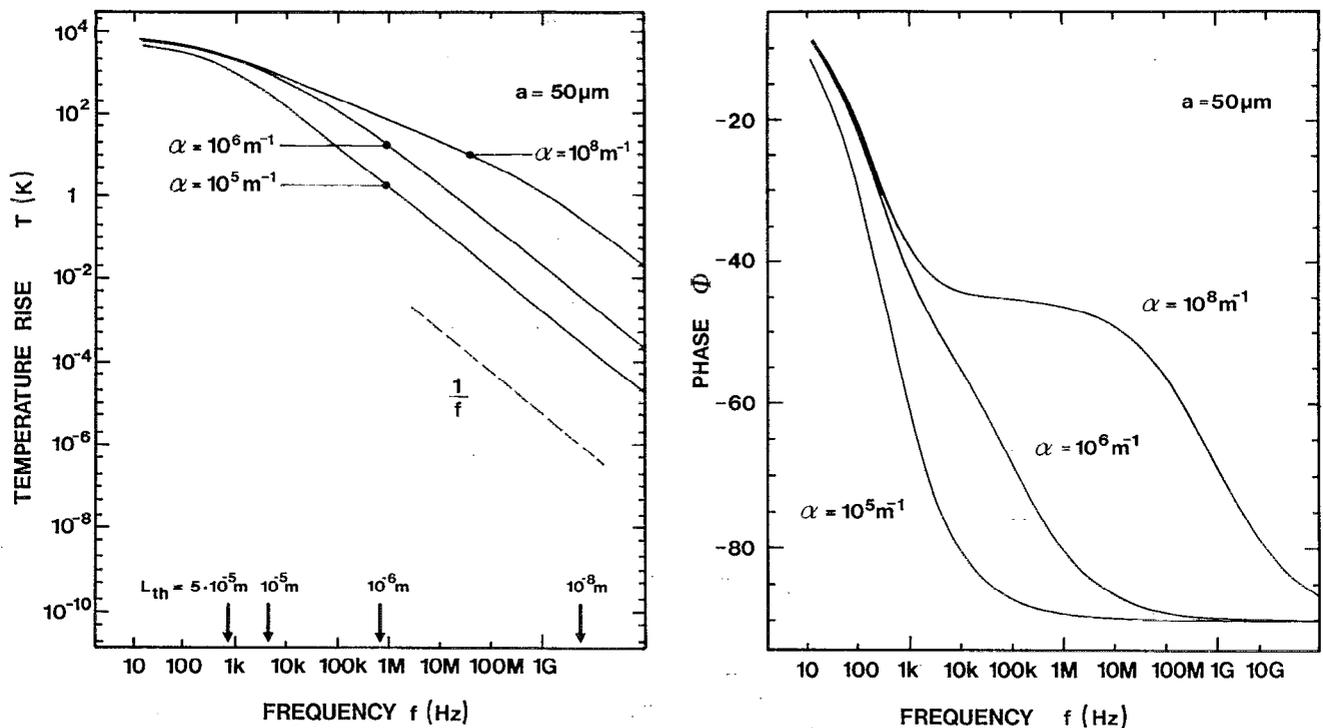


FIG. 1. Illustration of the frequency dependent amplitude and phase response of the surface temperature of a bulk material subject to modulated laser heating. Curves were calculated from theory using thermal parameters of fused silica and assuming three different values for the absorption coefficient α .

fact that by a variation of the modulation frequency the penetration depth of the heat wave can easily be controlled and the measured, i.e., harmonic part of the heat flow, can be restricted to the areas of interest, e.g., confined to a thin film. In a simple one-dimensional heat flow model,²⁵ a thermal length can be defined as

$$L_{th} = \sqrt{\frac{\kappa}{\pi f \rho c}}$$

L_{th} is the decisive parameter connecting material properties like conductivity κ , density ρ , and heat capacity c , with the modulation frequency f as the main experimental parameter. When adjusting the thermal length to other experimental parameters like pump beam diameter or thin film thickness, the photothermal response of the system often exhibits certain features as a function of parameters varied that allow an extraction of thermal parameters from the measurement.

This idea is also the basis for the present paper, where we present plots of the phase of the harmonic surface temperature variation as a function of the modulation frequency. For bulk materials these plots are generally characterized by two features demonstrated in a model calculation shown in Fig. 1.

For high frequencies the amplitude of the temperature variation exhibits a $1/f$ behavior while the phase remains at a constant value of -90° with respect to the excitation.²⁶ This arises from the fact that in this regime the temperature rise is solely determined by the energy deposited in the absorbing volume (defined by the pump beam diameter

and the light penetration depth or sample thickness). This amount of energy is proportional to the inverse frequency and will not be decreased by heat diffusion during a modulation period, provided the thermal length is small compared to typical dimensions of the absorbing volume.

In the low frequency limit, i.e., when the thermal length is much larger than pump beam diameter and/or absorption length, heat diffusion leads to an extension of the thermally active volume far beyond the absorbing volume during the modulation period, resulting in a decreased temperature rise and finally a saturation in the amplitude for $f \rightarrow 0$. Due to rapid diffusion, the temperature rise instantaneously follows the heating and the phase approaches a value of 0° in this regime.

The frequency range for the transition between the two regimes is determined by the material constants and the pump beam diameter as a result of lateral heat diffusion. Additional features in the frequency behavior arise from vertical heat flow when the thermal length passes the optical penetration depth. For surface absorption (see $\alpha = 10^8 \text{ m}^{-1}$ curve in Fig. 1) the phase exhibits a plateau at -45° in the intermediate frequency range, where the diffusion length L_{th} is larger than the pump beam diameter a , but smaller than the light penetration depth α^{-1} .

In thin film systems the flow of heat is influenced by a mismatch of thermal effusivities²⁷ at the interfaces between adjacent layers, leading to reflection of the heat waves, and under certain circumstances to thermal wave interference.^{28,29} By introducing the film thickness as a new characteristic length of the system, additional structures in

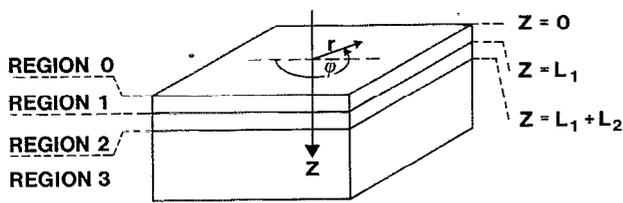


FIG. 2. Cross-sectional view of the model sample system. Region 0 (semi-infinite): atmosphere, region 1 (thickness L_1): first layer, region 2 (thickness L_2): second layer, region 3 (semi-infinite): backing substrate. For single layer calculations region 2 becomes the substrate and region 3 represents the backside atmosphere.

the frequency curves are expected. These structures contain information about the vertical heat diffusion rather than the lateral. In fact, it has been shown theoretically³⁰ and experimentally²¹ that the phase of the surface temperature modulation of a one layer system exhibits a characteristic minimum in that frequency range where the diffusion length is about equal to the film thickness. This observation can be used for a determination of thin film thermal parameters by a fit of theoretical frequency response curves to experimental data. In such measurements the harmonic heat flow is mainly restricted to the thin film and its vicinity and, therefore, the thin film response can be separated from that of the substrate. Often lateral heat diffusion is suppressed by choosing a large beam diameter. In that case the heat flow is nearly one-dimensional and the effectively measured conductivity is that in the vertical direction. Nonetheless, in many situations it is important for data interpretation to develop the theoretical model beyond the one-dimensional approximation, as will be shown in a forthcoming paper.³¹

Various calculations for temperature distributions relevant for modulated photothermal measurements have been published in the literature.³²⁻³⁴ A general approach for multilayer systems based on a one-dimensional diffusion model³⁰ has meanwhile been extended to the full three-dimensional solution of the heat diffusion equation.³⁵

The present work is an extension of the approach used by Jackson *et al.*,³⁶ which was originally developed for the interpretation of photothermal mirage measurements. In our calculations we introduce a new boundary, allowing calculations for two layer systems. Light absorption in both layers or in one layer and the substrate is taken into account. An important new feature covered by our theory is the possibility to incorporate a thermal resistance³⁷ between the layers, resulting in a finite temperature step at one interface.

II. ANALYTICAL MODEL

The geometrical model used for our calculations is sketched in Fig. 2. The system consists of four regions with arbitrary but homogeneous and isotropic thermal properties (conductivity κ_i , density ρ_i , and heat capacity c_i . The index i denotes the number of the layer) that are in thermal contact with each other. A semi-infinite substrate ma-

terial (region 3) is followed by two layers (regions 1 and 2) of arbitrary thickness (L_1, L_2), the latter in thermal contact with the adjacent atmosphere (region 0). A thermal resistance R_{th} may be present between the layers. We assume that light absorption takes place in these two layers that can be characterized by absorption coefficients α_i and reflectivities R_i , respectively. For cases with one layer on an absorbing substrate, region 2 is taken as the substrate and region 3 is either neglected or represents a backside atmosphere. The sample is assumed to be infinitely extended in lateral direction r . This approximation does not introduce any significant error, provided that laser beam diameter and thermal diffusion lengths are small compared to the lateral extension of the actual sample, a condition fulfilled in most cases of practical interest. Thermal transport is described in a pure thermal diffusion model neglecting any convective or radiative contributions. Neglecting convection is justified, since we are only dealing with the harmonic part of the total heat flow at frequencies far away from time constants typical for convection. Radiative energy transport has no relevance in our case, since only small amounts for the modulated temperature rise are expected.³⁸

Within this model the flow of heat in the system may be described in terms of four equations of heat diffusion for the respective regions. Here T_i denotes the temperature rise in region i , $k_i = \kappa_i / \rho_i c_i$ the diffusivities and Q_i the source terms for the energy input into the respective region:

$$\begin{aligned} \nabla^2 T_0 - \frac{1}{k_0} \frac{\partial T_0}{\partial t} &= 0, & \nabla^2 T_1 - \frac{1}{k_1} \frac{\partial T_1}{\partial t} &= -\frac{Q_1}{\kappa_1}, \\ \nabla^2 T_2 - \frac{1}{k_2} \frac{\partial T_2}{\partial t} &= -\frac{Q_2}{\kappa_2}, & \nabla^2 T_3 - \frac{1}{k_3} \frac{\partial T_3}{\partial t} &= 0. \end{aligned}$$

The solutions are determined by the boundary conditions at the three interfaces expressing conservation of the flux of heat through the interface and continuity of the temperature throughout the sample. Only at the interface between region 1 and region 2 we assume a finite temperature step determined by a thermal resistance R_{th} :

$$\begin{aligned} T_0 &= T_1, & \kappa_0 \frac{\partial T_0}{\partial z} &= \kappa_1 \frac{\partial T_1}{\partial z} \Big|_{z=0}, \\ -\kappa_1 \frac{\partial T_1}{\partial z} &= \frac{1}{R_{th}} (T_1 - T_2), & \kappa_1 \frac{\partial T_1}{\partial z} &= \kappa_1 \frac{\partial T_2}{\partial z} \Big|_{z=L_1}, \\ T_2 &= T_3, & \kappa_1 \frac{\partial T_2}{\partial z} &= \kappa_3 \frac{\partial T_3}{\partial z} \Big|_{z=L_1+L_2}. \end{aligned}$$

The source terms are described by linear absorption (absorption coefficients α_i) of a Gaussian laser beam with radius a , modulated with the frequency $f = \omega / 2\pi$. The coefficients of reflectivity for both layers R_1, R_2 are taken into account, however, multiple reflections leading to a nonexponential energy deposition profile are neglected. This assumption might impose limits of applicability of the model to some thin film systems, especially if internal reflectivities

are high. We denote the total power of the incident Gaussian laser beam by P and write, for the source terms,

$$Q_1 = (1-R_1) \frac{2\alpha_1 P}{\pi a^2} e^{-2(r/a)^2} e^{-\alpha_1 z} \times \frac{1}{2} \left(1 + \frac{1}{2} (e^{i\omega t} + e^{-i\omega t}) \right),$$

$$Q_2 = (1-R_1)(1-R_2) \frac{2\alpha_2 P}{\pi a^2} e^{-2(r/a)^2} e^{-(z-L_1)\alpha_2} e^{-\alpha_1 L_1} \times \frac{1}{2} \left(1 + \frac{1}{2} (e^{i\omega t} + e^{-i\omega t}) \right).$$

Due to the cylindrical symmetry of the problem, the differential equations describing the diffusion process can be solved by standard Hankel transform techniques. The solutions for the temperature rise are governed by an integral over a single variable δ , representing the Hankel variable in the radial direction:

$$T_0(r, z) = \int_0^\infty \delta d\delta J_0(\delta r) E(\delta) e^{\beta_0 z} e^{i\omega t} + \text{c.c.},$$

$$T_1(r, z) = \int_0^\infty \delta d\delta J_0(\delta r) [\Gamma(\delta) e^{-\alpha_1 z} + A(\delta) e^{-\beta_1 z} + B(\delta) e^{\beta_1 z}] e^{i\omega t} + \text{c.c.},$$

$$T_2(r, z) = \int_0^\infty \delta d\delta J_0(\delta r) [\Sigma(\delta) e^{-\alpha_2(z-L_1)} + L(\delta) e^{-\beta_2(z-L_1)} + G(\delta) e^{\beta_2(z-L_1)}] e^{i\omega t} + \text{c.c.},$$

$$T_3(r, z) = \int_0^\infty \delta d\delta J_0(\delta r) M(\delta) e^{-\beta_3(z-L_1-L_2)} e^{i\omega t} + \text{c.c.},$$

where

$$\Gamma(\delta) = (1-R_1) \frac{\alpha_1 P}{2\pi\kappa_1} \frac{e^{-(\delta a)^2/8}}{\beta_1^2 - \alpha_1^2}$$

and

$$\Sigma(\delta) = (1-R_1)(1-R_2) \frac{\alpha_2 P}{2\pi\kappa_2} \frac{e^{-(\delta a)^2/8}}{\beta_2^2 - \alpha_2^2} e^{-\alpha_1 L_1}$$

are the coefficients for the contributions to the temperature rise due to direct heating, while

$$\beta_i(\delta)^2 = \delta^2 + i \frac{\omega}{k_i} = \delta^2 + i \frac{2}{L_{th,i}^2}$$

may be regarded as a generalized wave number of the heavily damped thermal waves in positive and negative z directions, respectively. In regions 0 and 3, that we assume to be infinitely extended, only thermal waves in one direction are present. Expressing β in terms of the thermal diffusion length L_{th} reveals the dependence of the thermal signal on this parameter also in the three-dimensional case. The remaining coefficients A , B , E , G , L , and M are determined by applying the six boundary conditions to the solutions for T_i :

$$A = - \frac{(1-g)[b\gamma(1-nv) - v] e^{-\alpha_1 L_1 - \beta_1 L_1} + (g+v)[1+b\gamma(1+n)]}{H(\delta)} \Gamma(\delta) + \frac{(1-g)b[v-s+\gamma(1+v)] e^{-\beta_1 L_1}}{H(\delta)} \Sigma(\delta),$$

$$B = - \frac{(1+g)[b\gamma(1-nv) - v] e^{-\alpha_1 L_1 - \beta_1 L_1} + (g+v)[1-b\gamma(1-n)] e^{-2\beta_1 L_1}}{H(\delta)} \Gamma(\delta) + \frac{(1+g)b[v-s+\gamma(1+v)] e^{-\beta_1 L_1}}{H(\delta)} \Sigma(\delta),$$

$$L = \frac{1}{b(1-\mu)} [v\Gamma e^{-\alpha_1 L_1} + A e^{-\beta_1 L_1} - B e^{\beta_1 L_1} + b(v-s)\Sigma],$$

$$G = v\Sigma + \mu L,$$

$$M = \Sigma e^{-\alpha_2 L_2} + L e^{-\beta_2 L_2} + G e^{\beta_2 L_2},$$

$$E = \Gamma + A + B,$$

$$H = (g+1)[1+b\gamma(1+n)] - [1-b\gamma(1-n)] \times (1-g) e^{-2\beta_1 L_1},$$

$$g = \frac{\kappa_0 \beta_0}{\kappa_1 \beta_1}, \quad b = \frac{\kappa_2 \beta_2}{\kappa_1 \beta_1}, \quad v = \frac{\alpha_1}{\beta_1},$$

$$n = \kappa_1 \beta_1 R_{th}, \quad s = \frac{\alpha_2}{\beta_2}, \quad d = \frac{\kappa_3 \beta_3}{\kappa_2 \beta_2},$$

$$\mu = \left(\frac{1-d}{1+d} \right) e^{-2\beta_2 L_2}, \quad \gamma = \frac{1-\mu}{1+\mu},$$

where

$$v = \left(\frac{s-d}{1+d} \right) e^{-(\alpha_2 + \beta_2) L_2}$$

Note that the coefficients are governed by the ratios of κ_i/β_i at the respective boundaries, complex numbers determined by the thermal effusivities $(\kappa_i \rho_i c_i)^{1/2}$, as expected from simple qualitative considerations. Hence, the phase of the thermal signal will be influenced by the reflection of thermal waves at the interfaces. The thermal conductivity κ alone appears only in the prefactor of the temperature expressions, and, among others, determines the amplitude of the temperature variation. Therefore, with equipment providing absolute temperature calibration, it is possible to determine both parameters κ and k independently, e.g., by measurements of amplitude and phase of the surface temperature variation as a function of the modulation frequency. Practically absolute calibration is difficult and measurements are mostly restricted to the determination of the effusivity. Such measurements require only relative measurements as a function of frequency or other experimental parameters.

III. NUMERICAL RESULTS

The model described above yields complex numbers for the modulated temperature variation as a function of experimental and system thermal parameters. For comparison with experiments, these numbers are converted into values for the amplitude of the temperature variation and the phase of the thermal response relative to the modulation of the pump beam.

All plots presented below are representations of the amplitude or phase of the harmonic temperature variation in the center of the incident pump beam and at the surface of the respective thin film system; i.e., at the location of maximum temperature amplitude. Results are presented as a function of the modulation frequency.

Figure 3 shows (a) amplitude and (b) phase curves for gold films of different thickness on BK7 glass substrates, in comparison with results for a gold bulk sample. Absolute temperature values are given for the amplitude while the thin film phase values are taken relative to the bulk phase. Thin film amplitude as well as phase curves approach gold bulk behavior in the limit of high frequencies, and are identical if the thermal diffusion length is much smaller than the film thickness, as expected. In the low frequency region, the heat is able to penetrate into the backing glass, and the thermal behavior is dominated by substrate properties. High amplitudes arise from the small effusivity of the substrate. Note, that for the 500 nm film even at 1 kHz frequency a temperature amplitude of nearly 100 K results from 100 mW laser power focused into a 40 μm spot. The phase behavior is typical for a surface absorber with small diffusivity. The transition between substrate and thin film behavior occurs in that frequency region, where the thermal length is equal to the film thickness. A characteristic minimum in the phase curve is observed that can be used for a thermal characterization in photothermal diffusivity measurements. The crossing of thin film amplitude curves with the bulk curve and the

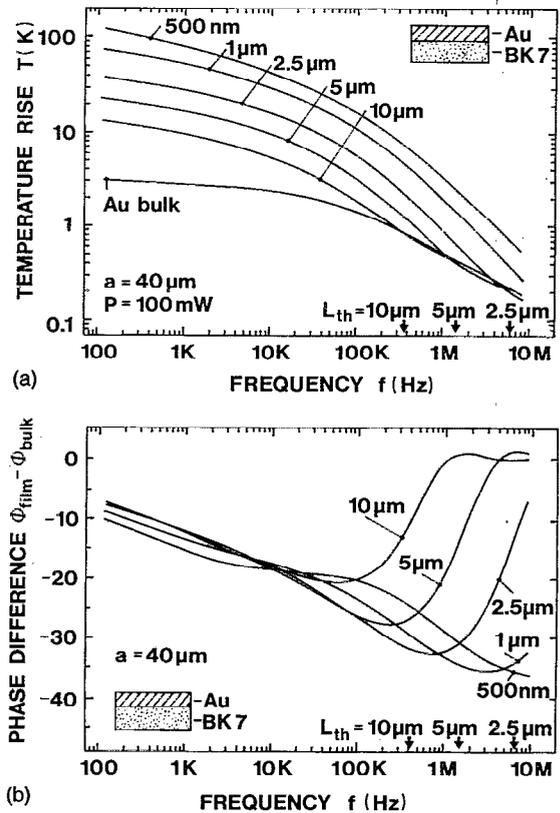


FIG. 3. (a) Amplitude and (b) phase response of the surface temperature for gold films of different thickness as a function of the pump beam modulation frequency f , in comparison with the curve for a bulk gold sample. Amplitudes are displayed in absolute units while the phase values are normalized to the bulk response. Arrows indicate frequencies where the film diffusion length is equal to the film thickness.

local maxima in the phase behavior are regarded as effects resulting from thermal wave interference of the heat wave excited at the film surface, with the wave reflected from the interface between film and substrate. Since heat waves are always heavily damped,²⁵ interference phenomena are only very weakly pronounced.

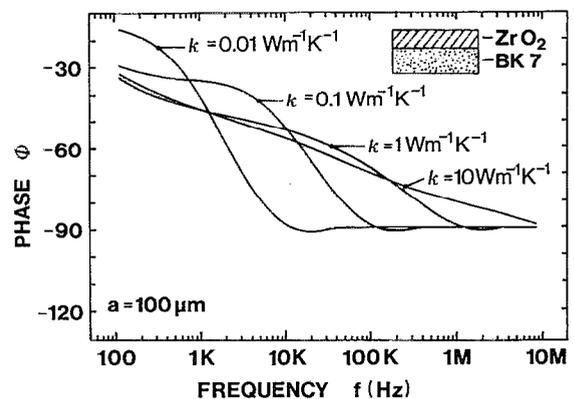


FIG. 4. Phase response of the surface temperature for a 1 μm ZrO_2 film on a BK7 glass substrate as a function of pump beam modulation frequency f . The film conductivity κ_1 was varied from 0.01 to 10 $\text{W m}^{-1} \text{K}^{-1}$; for the other thermal parameters literature values for ZrO_2 were used.

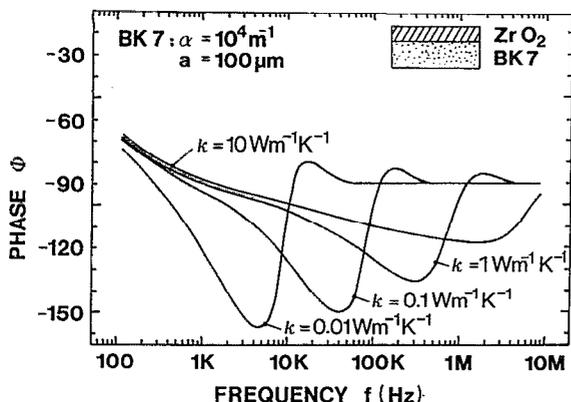


FIG. 5. Phase response curve for the same system as in Fig. 4, but with an absorbing substrate (filter glass, absorption coefficient $\alpha = 10^4 \text{ m}^{-1}$).

Our main interest on thin film conductivities is focused on optical coatings and diamond thin films, i.e., low absorbing dielectric materials on glass substrates. In this case the laser light is absorbed throughout the whole film and the frequency response is different from that of a surface absorber. Figure 4 shows a typical example for a $1 \mu\text{m}$ ZrO_2 film on a BK7 glass substrate. We performed the calculation for various values of the thin film conductivity above and below the bulk literature conductivity of $1 \text{ W m}^{-1} \text{ K}^{-1}$, assuming that density and heat capacity are equal to the respective bulk values.³⁹ From our model calculations we find that frequency-dependent conductivity measurements have to be performed for modulation frequencies up to 100 kHz to cover the full thin film effect in the phase behavior, provided the thin film conductivity is reduced by at least one order of magnitude compared to the bulk value.

A much more pronounced phase variation is observed if we assume substrate absorption for the same system, as demonstrated in Fig. 5. For low frequencies the absolute phase values are lowered by about 30° compared to the case of pure thin film absorption. The additional phase shift is a result of the fact that now the heat wave is excited at the

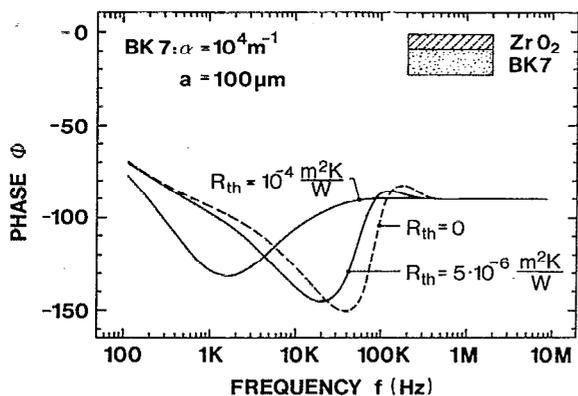


FIG. 6. Phase response curves for one system from Fig. 5 ($\kappa_1 = 0.1 \text{ W m}^{-1} \text{ K}^{-1}$) for different values of a thermal resistance R_{th} at the interface between ZrO_2 film and a glass substrate.

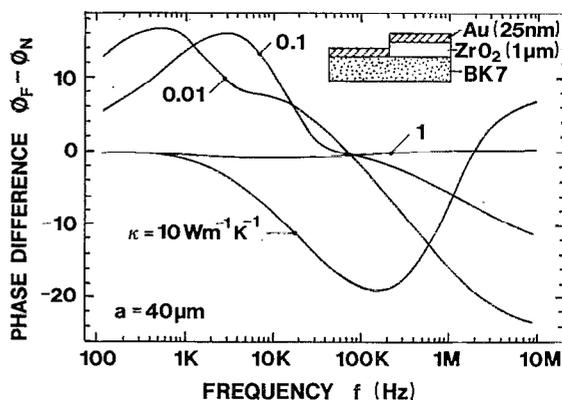


FIG. 7. Phase response curves for a double layer sample $\text{Au}/\text{ZrO}_2/\text{BK7}$ based on the systems from Fig. 4 with an additional 25 nm Au overlayer. Phases are normalized to the response of a single 25 nm Au film on a BK7 glass substrate.

interface and has to travel through the entire film until it reaches the sample surface. In the high frequency limit, however, the phase approaches the -90° value also found in Fig. 4 that is typical for a weakly absorbing bulk material. Here, the thermal signal is completely decoupled from the substrate thermal source and purely determined by thin film heating and diffusion. The interplay between thin film heating, substrate heating, and thermal interference between the excited heat waves results in the pronounced non monotonic phase behavior in the intermediate frequency region.

Next, we investigated the influence of a thermal interface resistance on the phase of the surface temperature rise, again for the $\text{ZrO}_2/\text{BK7}$ system with substrate absorption from the previous example. Phase curves for two different values of the thermal resistance are shown in Fig. 6. It is seen that the phase minimum is shifted toward lower frequencies for increasing thermal resistance, since the effective thermal conductivity of the composite system is lowered by the introduction of R_{th} . The exact phase behavior, however, may not be fitted by a phase curve, assuming a reduced thermal conductivity without thermal resistance at the interface. Thus, the interface resistance introduces a unique feature and one can expect that it may independently be determined by photothermal measurements of the precise frequency dependent phase shift.

One of the most convenient photothermal techniques is the method of modulated reflectance,⁴⁰ since there the surface temperature is directly measured and no complicated theory for signal generation is required. However, reliable measurements require materials with a sufficiently high temperature coefficient of the reflectivity.⁴¹ Thus, for conductivity measurements it is desirable to cover the dielectric thin film under investigation with an opaque but thermally thin metal overlayer providing the reflectance signal. Therefore, we calculated frequency response curves for two-layer systems consisting of a dielectric film on a glass substrate covered with a thin gold overlayer,⁴² as shown in Fig. 7. We have taken the same conductivity values for ZrO_2 as in Fig. 4, however, in Fig. 7 the surface tempera-

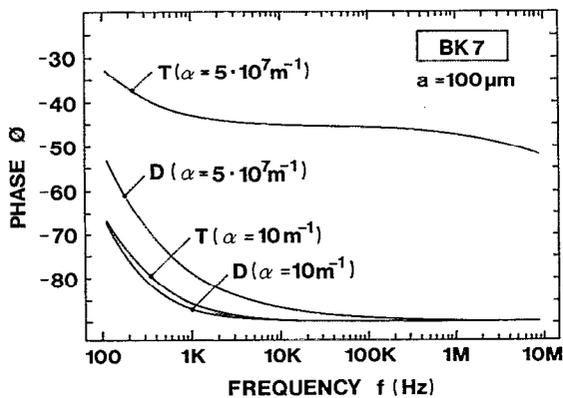


FIG. 8. Frequency dependence of the phase of surface temperature (T) and surface displacement (D) of a BK7 bulk material for the cases of surface absorption ($\alpha = 10^7 \text{ m}^{-1}$) and bulk absorption ($\alpha = 10 \text{ m}^{-1}$). The calculation was performed using the model from Ref. 45.

ture phase of the layered system is normalized with respect to the phase of a single gold layer directly deposited onto the BK7 substrate. It can be shown⁴³ that the exact properties of the overlayer of the two-layer system do not change the temperature response dramatically, and, therefore, need not to be known exactly for data interpretation of reflectance measurements based on this sample geometry.²¹ The phase response curve from Fig. 7 is mainly determined by the surface absorption in the gold overlayer and the thermal response of the ZrO_2 film. Of course, additional structures are introduced by the effusivity steps at the two interfaces. However, the calculation demonstrates that also for more complicated sample geometries the frequency dependent photothermal phase provides information about the thermal properties of a specific layer in the system.

IV. SURFACE DISPLACEMENT

For *in situ* measurements of dielectric thin film thermal properties a technique that requires a metallization of the layer-like thermoreflectance is not appropriate. In such cases the mirage technique⁴⁴ or the surface displacement technique⁴⁵ have been shown to be applicable.^{46,47} Surface displacement is most interesting due to its simple alignment, high frequency capability, and unique sensitivity. Therefore, we also investigated the harmonic surface displacement response of thin film systems. Although the surface displacement has been calculated for bulk materials by solving the Navier–Stokes equation for a semi-infinite medium,⁴⁵ for thin films only approximate analytic solutions are available.⁴⁸ This is due to the complexity of the general thermoelastic problem, leading to an extremely complicated analytical solution if all boundary conditions are taken into account. On the other hand, it has been shown that under proper experimental conditions a simple first order approximation of the complete thermoelastic equation of motion yields reasonably good results for the surface displacement.⁴⁹ This approximation can be regarded as a linear expansion model, since we assume that

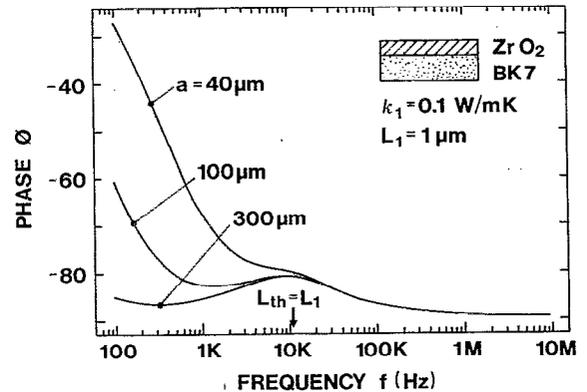


FIG. 9. Surface displacement calculated for a $1 \mu\text{m}$ ZrO_2 film on BK7 with the linear expansion model. The thin film effect due to vertical heat flow at $L_{\text{th}} = L_1$ is only very weakly developed, and may be hidden by lateral heat flow effects depending on pump beam radius a .

all thermoelastic shear stresses and strains can be neglected, and the surface displacement simply results from linear expansion along the z direction. Since all heated layers contribute to the expansion, the effective displacement at the sample surface has to be represented as a sum of z integrals over the temperature distributions multiplied by the thermal expansion coefficients and a Poisson factor for the respective region:

$$u_z(r,0) = \sum_i \alpha_{\text{th},i} \frac{1+\nu_i}{1-\nu_i} \int_{L_i} T_i(r,z) dz.$$

Although this is not an exact representation for the surface displacement, the equation retains the most important features of the problem, especially if the pump beam radius is large compared to the thermal length and the light penetrates the whole film. The former condition leads to small stress gradients in lateral direction while the latter provides a smooth profile for the depth dependence; both together result in a substantial reduction of shear phenomena.

A striking consequence of the integral representation of the surface displacement is the fact that any displacement measurement on a bulk sample is not at all sensitive to vertical heat flow. This effect is demonstrated in Fig. 8, where we calculated both the surface temperature and the surface displacement for a bulk material for two different absorption coefficients representing surface absorption and bulk absorption, respectively. For the bulk absorption case the phase curves for surface temperature and surface displacement are almost identical since there appears only a negligible vertical temperature gradient. In the case of surface absorption vertical heat diffusion is dominant and the two curves differ considerably. In the limit of high frequencies the displacement curve approaches that for the bulk absorbing case, indicating the integrative nature of the displacement signal. The deviation for low frequencies is due to lateral heat flow and, hence, dependent on the pump beam radius.

These considerations have to be kept in mind when applying the displacement method on conductivity measurements based on the technique described in this paper. Since, this technique is based on the measurement of vertical heat flow reliable results can only be expected if the coefficients of thermal expansion of both substrate and thin film are well known and differ from each other. Otherwise no thin film features will be detected. As an example for a displacement calculation based on the linear expansion model, we present the phase behavior of the surface displacement for the $ZrO_2/BK7$ system in Fig. 9. As expected, there appears a thermal interference feature in the region, where the diffusion length is equal to the film thickness, however, it is only weakly developed. This calculation also demonstrates that for a safe interpretation of such measurements it may be important to suppress lateral heat flow by choosing a large pump beam diameter; otherwise the thin film effect will be hidden in the general low frequency phase behavior.

V. CONCLUSIONS

A complete three-dimensional heat diffusion model for isotropic two-layer systems has been presented describing harmonic heat waves excited by a Gaussian laser beam. Absorption in both layers as well as changes in the thermal properties at three boundaries and a thermal contact resistance at one interface have been taken into account. The frequency dependence of amplitude and phase of the harmonic temperature variation have been calculated for various sample configurations consisting of low absorbing dielectric thin films on glass substrates. The frequency response curves reveal characteristic features in that frequency region, where the thermal length in the film is equal to the film thickness. These features are a result of steps in the effusivity at the interfaces leading to the reflection of heat waves and thermal interference. They appear in both the amplitude and phase response curves, however, in the phase behavior they are more pronounced. The strongest effect is found for a low absorbing thin film on a substrate material with a much higher absorption coefficient. A thermal interface resistance introduces a clear shift and deformation of the thin film feature in the phase curve, while the amplitude behavior is only slightly affected. If the thin film system is covered by an additional, thermally thin but absorbing metallic overlayer, the frequency dependent thermal response becomes more complicated, however, the main phenomena are still determined by the thin film. The effects quantitatively described in this paper can be used for the interpretation of conductivity measurements on thin film systems utilizing any photothermal method that is sensitive to sample surface temperature variation and can be operated in the appropriate frequency range. The required frequency interval depends on thin film conductivity and thickness. With the help of a simple one-dimensional thermal expansion model it was shown that the surface displacement technique is not sensitive to vertical heat flow and thin film conductivity measurements based on the described technique are only possible if the

coefficients of thermal expansion of thin film and substrate are well known and differ from each other considerably.

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