Albert Reimer
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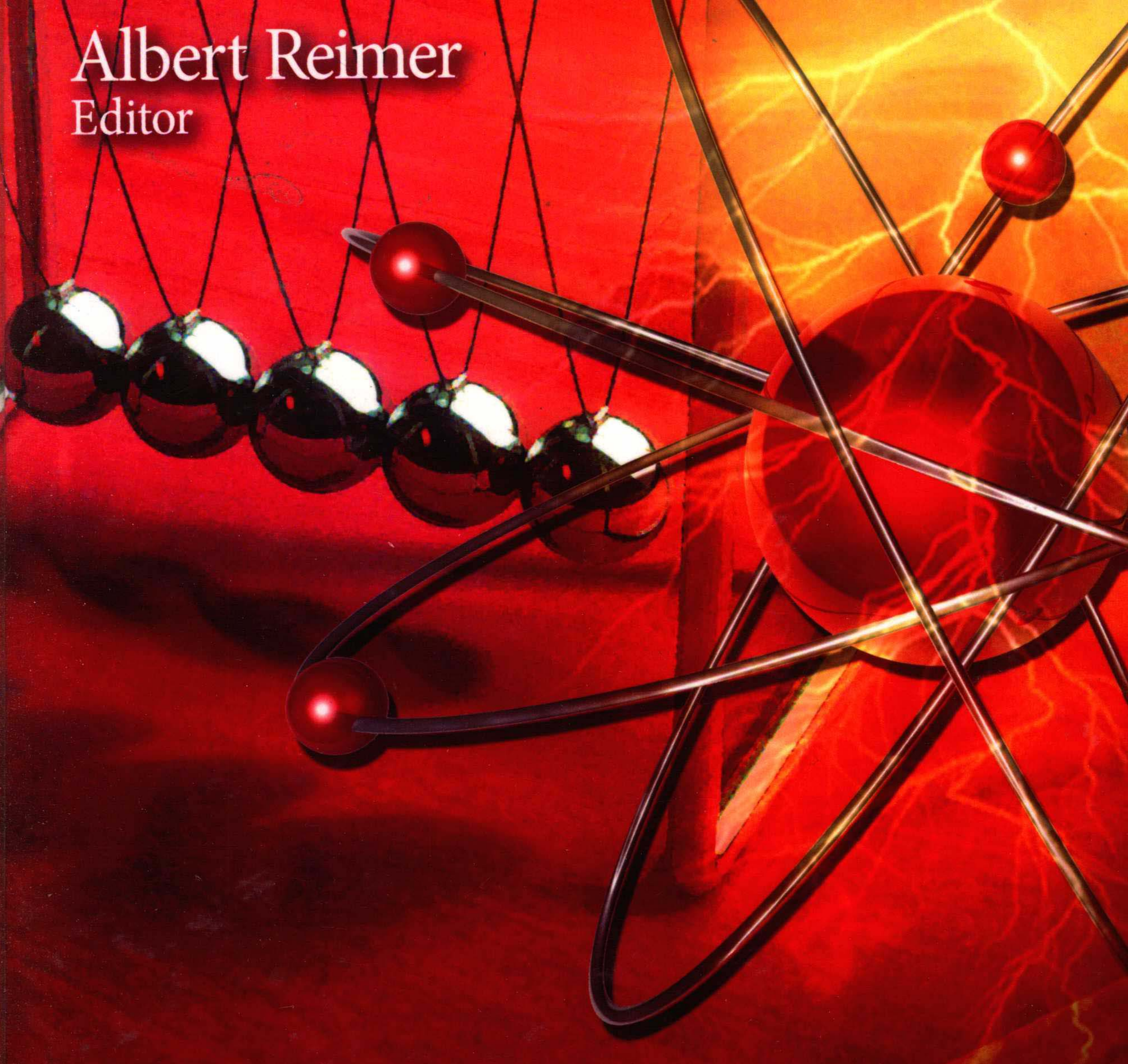

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VOLUME 276

ALBERT REIMER
EDITOR

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PREFACE

This book presents original research results on the leading edge of physics. Each article has been carefully selected in an attempt to present substantial research results across a broad spectrum. Topics included in this compilation include radiative transfer and its impact on thermal diffusivity in remote sensing; ferromagnetism in oxide based diluted magnetic semiconductors; the microstructure properties and critical damage of metals and the relativistic spin-polarization in quark matter.

Chapter 1 - Thermal diffusivity is usually determined from measured temperature evolution when a sample is exposed to a transient energy source. In almost all cases reported in the literature, heat flow through the sample is considered as purely conductive. Instead, this chapter, on an introductory level, investigates to which extent conduction coupled to radiation is reflected by the temperature evolution of thin films, and how this alters diffusivity if it is extracted using standard procedures from transient temperature.

The analysis is applied to flash experiments using constant or wavelength-dependent extinction properties of thin films, and to periodic (intensity modulated) energy sources. Focussed on samples with optical thickness between 1 and 50, the results demonstrate that coupling between conduction and radiation may significantly alter thermal diffusivity. This would occur if diffusivity is extracted from data taken at single surface positions only, or by any other technique that relies on coupling between surface temperature and temperature of a medium above the surface, like in mirage experiments. Uncertainties are expected also in intensity modulated experiments when diffusivity is determined from correlations with phase shifts. A front-face flash method recently introduced by the authors can be more reliable.

Chapter 2 - This chapter reviews the unusual ferromagnetism in the research of diluted magnetic semiconductors (DMS), mainly focusing on the oxide based DMSs. The unusual ferromagnetism includes non-intrinsic ferromagnetism, such as ferromagnetism induced by magnetic element formed clusters, nonmagnetic element formed clusters, structure defects, such as cation vacancies, anion vacancies, cation and anion interstitial. In addition, research has shown that perfect single crystal film with magnetic element doping does not exhibit ferromagnetism, suggesting that the way for magnetic element doped oxide semiconductor may not be applicable for achieving high quality DMS for spintronics. In addition, the unusual ferromagnetism also includes light element, such as, C and N as dopant in oxide semiconductor induced room temperature ferromagnetism. It is due to the coupling between p orbital of light element, such as C and N and p orbital of O in oxide semiconductor, which is different from traditional p-d or s-d coupling. Moreover, alkaline metal earth element doped

ZnO has shown hole-mediated ferromagnetism at room temperature and the ferromagnetism is originated from cation vacancies. Furthermore, the different possible mechanisms for the ferromagnetism in pure oxide semiconductors are discussed. Finally, the giant moment of dopants is also introduced and discussed.

Chapter 3 - Fundamental issues of the growth of InAs/GaSb superlattice (SL) structures on a GaSb buffer layer using molecular beam epitaxy (MBE) have been investigated. It has been observed that high quality substrates are crucial to obtain a GaSb buffer without crystal defects. The investigations by high resolution X-ray diffractometry showed that the presence of mosaity or cracks in the substrate crystal leads to the deterioration of crystal quality of GaSb buffer layer. The substrate temperature and Sb-flux were varied during GaSb growth steps to determine the optimal oxide desorption, growth parameters and conditions under which Sb sublimation out of material and Sb condensation on the surface are in equilibrium. It was found that both the oxide desorption and the post-growth cooling run without Sb surface stabilization. The pre-growth annealing reduces significantly the contamination of impurities at the substrate - epi interface. Such growth conditions allowed investigating the periodic structure of type-II InAs/GaSb superlattices. During the strain balancing process, the correlation between beam equivalent pressure (BEP) of arsenic, interruption time at the interfaces, and superlattice strain was found. Based on reproducibility of epitaxial processes, the optimal growth conditions were specified and lattice matched InAs/GaSb superlattices were obtained. These results assure the relevant control of the MBE process of fabrication superlattices suitable for mid-infrared detection.

Chapter 4 - Metal structures experience gradual change in deformation properties under loading. These changes are negligible in the elasticity zone; they do not affect the properties of the material. Beyond the elastic deformation, the microstructure damage starts to accumulate. Stress - strain curve becomes nonlinear. The amount of microstructure damage is calculated with the methods of stochastic micro-heterogeneous media mechanics. The probability distribution of random microstructure stresses is obtained by an integral operator equation, which includes the Green's tensor. The microstructure damage is calculated as the probability of random stress exceeding the random strength in elements of microstructure. The normal distribution and the Weibull distribution are used to obtain the random microstructure strength condition.

For every loading cycle, the calculation of the microstructure damage makes it possible to find the macroscopic properties of the composite and to plot the corresponding section of the stress-strain curve. The emergence of macroscopic cracks in the material corresponds to the termination point of stress-strain curve after transition to a negative Young's modulus. At the same time, the stress-strain curve transitions to a descending branch. The corresponding stress is equal to the tensile strength of the material at this loading condition. The stress-strain curves drawn for the shear modulus and volumetric strain modulus of metals are also studied. The influence of Poisson's ratio on the character of damage is discussed.

Calculations show that materials with considerable heterogeneity of microstructure have higher coefficients of variation of the strength condition. Such materials accumulate damage faster. Under the same conditions, their stress-strain curves are lower than stress-strain curves for metals with smaller spread of the microstructure properties.

The matrix of microstructure stresses is considered for the analysis of damage under complex stress condition. Critical damage corresponds to zero and infinite eigenvalues of the matrix. The corresponding eigenvectors determine the three-dimensional surface of strength.

The degree of metal damage that is deemed critical is defined by the proposed use of the structure and is determined from experiment and engineering expertise. For statistical problems, this value is related to the safety factor from deterministic problems. The relationship between the critical microstructure damage of the composite and the margin of safety for strength with variable coefficients of variation of structural stresses is determined. The authors show that the increased safety factor corresponds to a decrease of the critical damage allowed in the use of structure. Taking into account the microstructure properties of the material makes it possible to reduce amount of material and weight of the structure, while maintaining its strength.

Chapter 5 - In modern tennis, the essential part of executing high-performance shots is generating topspin and backspin on the ball. The purpose of this paper is to present a rigorous kinetic analysis of the flick-motion of the tennis racket, based on the original concept of the vector of mass/inertia moments coupled to the pole and for the corresponding axis. The forward racket flick-motion generates topspin on the ball, while the backward racket flick-motion generates backspin (also known as slice) on the ball. To describe both kinds of the racket flick-motion we use rigorous kinetic analysis. This advanced rigid-body analysis includes the six degrees-of-freedom (DOF) Newton-Euler dynamics, a new sophisticated form of vectors/tensors of the racket mass inertia moments, impact forces during the racket-ball contact, and mass-deviational moment vectors of the racket and ball rotation before, during and after contact.

Chapter 6 - Relaxor ferroelectrics of perovskite oxides show interesting dielectric and electromechanical properties of practical applications, where polar nano-domains play a crucial role. The origins of this polar domain in the perovskite relaxors of cubic symmetry are not fully understood although some models proposed in analogy to the spin glass or the extended random field models are in partial success. On the basis of our experimental observations of electron diffraction and Raman scattering measurements we want to show that the relaxor ferroelectric behavior may be rooted from the local flexoelectricity. The flexoelectric effect has been long understood in liquid crystal systems in terms of directors and strain gradients.

This talk will start with Introduction [I], where the authors' problem of local domains of polarization in cubic symmetry of crystals is defined in terms of specific relevance to perovskite oxides, polar nano-domains and solid state flexoelectricity. Two essential ingredients for the flexoelectric polarization will be then discussed in [II] Directors in solid and [III] Strain gradients, then they propose for the origin of the formation of local polar domains in [IV] Flexoelectric polarizations in solid, and details will be given for probing the effects of the local flexoelectric polarization in [V] Experimental verifications: electron diffraction and Raman scattering measurements.

The authors close this chapter with a Conlusion [VI], where they conclude that flexoelectric polarization in solids is responsible for the polar nano-domains in the ferroelectric relaxors. However, Raman optical activity(ROA), a variation of Raman scattering measurement, seems to be better in the signal to noise (S/N) ratio than the present measurement of Raman band depolarization ratios to remove the homogeneous strain contribution to the observed Raman bands as a background in addition to the inhomogeneous strain(strain gradient) contribution of the flexoelectric polarization.

Chapter 7 – The authors study the magnetic properties of quark matter, ferromagnetic ordering and spin density wave. The mechanism of the spontaneous spin polarization in the relativistic framework is quite different from that in the nonrelativistic one. There are two types of the spin polariszations: one is that through the axial-vector interaction, and the other is through the tensor interaction. In this text, first, they explain these mechanisms within the relativistic mean-field theory and compare them. Next we discuss somewhat different subject of possibility of the dual chiral density wave, one of the non-uniform phases induced by chiral transition, within the NJL model. Treating it by using the mean-field approximation, we show its possible formation and another interesting magnetic property like spin density wave in it. Finally the authors explain a recent development of their study about ferromagnetic transition in quark matter, where magnetic susceptibility is calculated within the Landau Fermi-liquid theory, based on the one-gluon-exchange interaction. The authors figure out important and interesting roles of screening on gluon propagator.

Chapter 8 - The issue of controllability of ferromagnetism in graphene-based samples is addressed. To study the magnetic properties in graphene-based samples, the authors systematically carry out quantum Monte Carlo simulations of the Hubbard model on a honeycomb lattice. In graphene, the structure of the honeycomb lattice leads to the well known massless-Dirac-fermion -like low energy excitations and two Van Hove singularities in the density of states at electron filling hni = 0:75 and 1.25, which determine much of system's magnetic properties. In the filling factor at hni = 0:75, graphene shows a short-range ferromagnetic correlation, which is slightly strengthened by the on-site Coulomb interaction and markedly by the next-nearest-neighbor hopping, and a possible flat-band ferromagnetic solution is discussed. Moreover, the ferromagnetic properties depend strongly on the electron filling, which may be manipulated by the electric gate. The possible experiment to realize controllability of ferromagnetism has been proposed based on our results. Due to its resultant controllability of ferromagnetism, graphene-based samples may facilitate the development of spintronics and many other applications

Chapter 9 - A synthesis of the present knowledge on gamma-ray emission from the magnetosphere of a rapidly rotating neutron star is presented, focusing on the electrodynamics of particle accelerators. The combined curvature, synchrotron, and inverse-Compton emissions from ultra-relativistic positrons and electrons, which are created by twophoton and/or one-photon pair creation processes, or extracted from the neutron-star surface, provide them with essential information on the properties of the accelerator, in which the electro-static potential drops along the magnetic field lines. It is demonstrated that any particle accelerator models adopting a thin geometry in the meridional direction cannot reproduce the observed fluxes of gamma-ray pulsars. It is then pointed out that the pair-starved polar-cap model requires the Goldreich–Julian charge distribution that is incompatible with the Poisson equation for the electrostatic potential, one of the Maxwell equations. Finally, a new accelerator model is proposed, by solving the Poisson equation for the electrostatic potential together with the Boltzmann equations for particles and gamma-rays in the three-dimensional pulsar magnetosphere. This scheme is applied to the Crab pulsar, and shown that the obtained solution corresponds to a quantitative extension of the previous outer gap models and reproduces the gamma-ray observations at least qualitatively. Relationship between the gap model and the force-free magnetosphere is discussed.

CONTENTS

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Chapter 1

RADIATIVE TRANSFER AND ITS IMPACT ON THERMAL DIFFUSIVITY DETERMINED IN REMOTE SENSING

***Harald Reiss*[1*] *and Oleg Yu. Troitsky*[2]**

[1]University of Wuerzburg, Wuerzburg, Germany

[2]Seversk State Technology Academy, Seversk, Tomsk Region, Russia

ABSTRACT

Thermal diffusivity is usually determined from measured temperature evolution when a sample is exposed to a transient energy source. In almost all cases reported in the literature, heat flow through the sample is considered as purely conductive. Instead, this paper, on an introductory level, investigates to which extent conduction coupled to radiation is reflected by the temperature evolution of thin films, and how this alters diffusivity if it is extracted using standard procedures from transient temperature.

The analysis is applied to flash experiments using constant or wavelength-dependent extinction properties of thin films, and to periodic (intensity modulated) energy sources. Focussed on samples with optical thickness between 1 and 50, the results demonstrate that coupling between conduction and radiation may significantly alter thermal diffusivity. This would occur if diffusivity is extracted from data taken at single surface positions only, or by any other technique that relies on coupling between surface temperature and temperature of a medium above the surface, like in mirage experiments. Uncertainties are expected also in intensity modulated experiments when diffusivity is determined from correlations with phase shifts. A front-face flash method recently introduced by the authors can be more reliable.

Keywords: Heat transfer, radiation, non-equilibrium states, Monte Carlo simulation, extinction, thermal diffusivity, remote sensing.

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* Address: Department of Physics, University of Wuerzburg, Am Hubland, D-97074, Wuerzburg, Germany.

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1. SURVEY

Intensive heat and mass transfer is involved during plasma spraying of protective coatings like Y-stabilized ZrO_2 on gas turbine blades or during chemical vapour deposition of amorphous carbon on combustion engine components. Like other crystalline ceramics, ZrO_2 due to its chemical stability and superior hardness, is a candidate for highly wear-resistant surface coatings, to protect metallic or glassy substrates from corrosion or erosion, or as buffer layers in semi-conductor manufacture or as solid electrolytes in fuel-cells.

Also in their use phase, protective thin films of ZrO_2 may be subject to very high temperatures and temperature gradients. Accordingly, there is a clear need for accurate, in-situ determination of the thermal diffusivity of ZrO_2, SiC or TiO_2, not only during preparation but also during ageing of the films. For these conditions, thermal wave technique and flash-methods are available for remote measurement of the thermal diffusivity of thin films.

Thermal wave technique, with intensity modulated energy sources, comprises scanning photo-acoustic microscopy, mirage effect, infrared radiometry or measurement of optical displacement. Flash methods apply very short energy pulses from laser radiation or from an electron beam. Both methods include investigation of sub-surface materials anomalies like detection of cracks and pores or regions of high mechanical stress that may arise during manufacture or result from materials fatigue. Roughly speaking, both methods, thermal wave technique and flash experiments, are sensitive to all disturbances that manifest themselves as variations of thermal diffusivity.

Thermal waves may be generated from localized intensity-modulated heat sources created by thin electrical heating elements prepared on sample surface, or from laser or electron beam irradiation; the thermo-reflectance microscope operates with even point-like heat sources. By variation of the modulation frequency, surface or in-depth scanning of the thermal transport properties including determination of interfacial resistances of layered samples can be performed.

Localised heat sources are applied also in flash technique but the heat source is generated from absorption of single, short energy pulses. Flash technique, accordingly, is rarely used for in-depth scanning of thermal transport properties; its advantage is the ease by which the experiments can be performed and the results analysed.

In the present state of the art, both methods work successfully under the following conditions:

a) stationary “background” sample temperature,
b) opacity of sample or substrate material, which means signals (temperature variations) arrive at a detector solely by thermal conduction,
c) laboratory conditions, like in the thermo-reflectance microscope.

Traditionally, isotropic transport properties, too, have been considered a prerequisite for successful application of thermal wave or flash methods, but approaches have been reported recently [1-3] that allow precise determination of diffusivity from flash experiments also in case of strongly anisotropic thermal conductivity, or in layered materials, or for detection of disturbances like cracks. This is a significant step forward in remote sensing since isotropic conduction properties can rigorously be fulfilled only with poly-crystalline thin films or with pure metals if film thickness is sufficiently large, in other words, in some rare cases only.

Condition (a) of the above neither may be fulfilled during sample manufacture nor during use phases. It is questionable whether the time interval, Δt, needed to reach stationary thermal wave propagation is short enough to avoid collisions with "internal" variations of sample properties (temperature, coating thickness, which increases during growth, or change of chemical composition). Plasma spraying of a protective coating on a gas turbine blade, for example, is performed by running a beam (droplets of molten metallic or ceramic powder) delivered by a spray gun line by line over the substrate. This not only induces strong variations of sample temperature, with a frequency in the order of seconds, but coating thickness rapidly increases during measurement, and crack formation may arise behind the solidification front due to thermal contraction or thermo-mechanical mismatch between coating and substrate. The time interval, Δt, to reach a stationary state scales with L^2/D, with L the momentary thickness of the coating and D its diffusivity. In case of large growth rate, like in plasma spraying, or if the diffusivity is small, like in thermal protective coatings for gas turbines, this means the interval Δt, too, can extend to the order of seconds so that the intensity-modulated thermal wave experiment cannot follow quickly enough variations of sample temperature and properties during its deposition.

Laboratory conditions, item (c) of the above, hardly can be realized if the sample is integrated in a running industrial process, or if the thin film shall be investigated during its preparation, which like plasma spraying or chemical vapor deposition proceeds at high temperatures and in closed deposition chambers.

The thin film may either be transparent or opaque to radiation. Clearly, the first case is not very suitable for application of thermal wave or of flash technique: The aim of the usual experimental setup is to determine the *solid thermal diffusivity*, and as a consequence, the applied method must respond *solely* to solid thermal conduction. Accordingly, it is item (b), the extinction coefficient of the material that must be very large, if the film thickness is given, and is the pre-requisite for successful application of wave and flash experiments. Naturally, this is the ideal situation, and tempting assumptions like "opaque sample" thus have often been made in the literature to simplify interpretation of results. It is condition (b) that will be preferentially discussed in this paper.

The pulse technique can be applied virtually to all solids, and attempts have been made to use this technique even for the investigation of the thermal diffusivity of liquid metals. While metals and alloys or opaque non-metals can be measured in a straightforward way, materials and coatings have mostly been investigated in a layered arrangement where an irradiated covering layer is applied that must be ultra-thin and opaque, or the investigated material simply must be blackened. We will later discuss that this practice could be of little value.

1.1. The Parker and Jenkins Approach

In a frequently referenced paper, Parker and Jenkins [4] derived a solution to the thermal conduction problem in a flat thin film sample of surface, A, under a transient surface heat source: Let x = L denote sample thickness, and d the thickness of a thin layer at the sample surface. A single heat pulse of magnitude Q/A [Ws m^{-2}] is delivered from an arbitrary energy source to this layer. Boundary conditions read

$$0 \leq x \leq d: T(x, t = 0) = (Q/A)/(\rho\ c_p\ d)$$

$$d < x \leq L; T(x, t = 0) = 0 \quad (1)$$

using t = time and ρ and c_p the density and specific heat of the sample material, respectively. The thickness d can tentatively be interpreted as the mean radiation penetration depth, l_m, below the sample surface. Under these conditions, the solution to Fourier's differential equation for all $0 \leq x \leq L$ is given by a series expansion

$$T(x,t) = [(Q/A)/(\rho\ c_p\ L)]\{1+2\sum[(\sin(n\ \pi\ d/L))/(n\ \pi d/L)]\cos(n\ \pi\ x/L)\exp(-n^2\ \pi^2\ D\ t/L^2)\} \quad (2)$$

In Eq. (2), the summation over n has to be taken from n = 1 to n → ∞. If, hopefully, d → 0, the factor sin(n π d/L) reduces to zero, and the solution converges to

$$T(x,t) = [(Q/A)/(\rho\ c_p\ L)]\ [1+2\sum \cos(n\ \pi\ x/L)\ \exp(-n^2\ \pi^2\ D\ t/L^2)] \quad (3)$$

which for x = L (the rear sample surface), and $\cos(n\ \pi) = (-1)^n$, yields

$$\Theta(L,t) = 1 + 2\sum(-1)^n \exp(-n^2\ \pi^2\ D\ t/L^2) \quad (4)$$

using the dimensionless temperature $\Theta(L,t) = T(L,t)/T_{max}(L)$; this can easily be measured.

Note that this comparatively simple result is based on the assumption d → 0. This ideal case completely neglects *any* radiative transfer in the thin film. The solution given in Eq. (4) further neglects radiative exchange between the sample and its environment, and possibly existing convective losses; appropriate corrections have been suggested in the literature for these items. The literature also discusses uncertainties that may result from too small a target spot in relation to the whole sample surface or in relation to the sample thickness. Eqs. (2) to (4) indeed are based on the assumption of strictly one-dimensional (1D) thermal conduction, which is hardly fulfilled if the target spot would be tiny; for this case, the above equations have been modified.

However, neither is it clear that the penetration depth, d, is zero under all circumstances nor can we assume that transfer of thermal energy proceeds by conduction only, regardless whether the energy is delivered as a periodic excitation or as a single pulse. The penetration depth, d, goes to zero *only* if the absorption coefficient, A_Λ, of the thin film material is very large, at *all* wavelengths, Λ, not only at the wavelength of the incoming laser radiation (in principle, also scattering contributions to a total extinction coefficient would have to be considered; this is neglected for the moment). If instead there are radiation transmission "windows" in some regions of the thermal spectrum, neither will penetration depth d → 0 nor will assumption (b) of the above (energy transfer solely by conduction) be fulfilled.

This also applies if a thin layer, strongly absorbing at all wavelengths, is prepared on the sample surface, to prevent direct transmission of the laser beam into the depth of the sample and, possibly, unto a detector positioned on the sample rear surface in the axis of the laser beam.

After absorption of the incoming radiation by this layer, with the radiation from *any* kind of source (spectral or integral, directional or diffuse, or even if the incoming flux is non-radiative like an electron beam), the thin layer itself will thermally and diffusely radiate at *all* (thermal) wavelengths, according to its temperature, including the wavelength of an incident beam. This means if there are transmission windows, the sample will be open to radiative transfer in the corresponding intervals, regardless from which source the original excitation comes from (laser or electron beam, a thin surface heater or even from the thin radiating absorbing layer, or from absorbing internal interfaces). Accordingly, there will be radiation heat transfer parallel to conduction, with or without application of a thin absorbing surface layer.

Parker and Jenkins also assumed that absorption of the pulse in the volume V = d A takes place within zero time interval, and that there is uniform absorption in this volume. Neither assumption is fulfilled in reality.

The question is whether all this could seriously affect accuracy of the standard methods to determine thermal diffusivity; this is the subject of this paper. For this purpose, we will discuss the problem how to conclude whether radiation plays an important role or not for determination of thermal diffusivity by

a) considering the absolute magnitude of surface temperature evolutions,
b) a front-face flash method recently introduced by the authors that mainly relies on *ratios* of transient surface temperature.

1.2. Different Propagation Velocities and Related Problems in Flash Experiments

Two more problems arise from the enormous difference of propagation velocities of thermal waves that are driven solely by conduction or by radiation. For simplicity, consider a flat, semi-infinite slab: Under pure 1D-conduction, a thermal disturbance propagates like a plane wave with a velocity

$$v_{Cond} = C\ (D/t)^{1/2} \tag{5a}$$

with C a constant (C = 1.8 for a flat sample), i. e. it travels at a finite speed that decreases with time, while under pure radiation heat transfer, radiation travels with constant velocity of light,

$$v_{Rad} = c/n, \tag{5b}$$

with n the refractive index (assumed isotropic, but this may be quite different in layered materials or under local density or electrical conductivity variations). Between these to limits, the velocity by which the interior of a thin film sample experiences a thermal disturbance is subject to absorption/remission properties of the material. The quantity that overall reflects the absorption properties is the complex index of refraction, and in Eq. (5b) the velocity of light that is reduced by its real part, n. This is also reflected by the total optical path length obtained from integration over all single mean free paths between interactions and by the coupling strength between conduction and radiation heat flow. Coupling accordingly affects not only the magnitude of the signal seen by a detector positioned at rear or front sides of a thin film, but also the evolution of the signal with time. The present paper delivers examples illustrating this problem (see later, e. g. Figure 9b).

Further, consider again a purely conductive, semi-infinite sample, and let it be exposed to a intensity modulated heat source at its boundary, with a frequency ω. Penetration depth, δ(ω), of the thermal disturbance into the sample, from the periodic thermal excitation, is given by the expression

$$\delta(\omega) = C\ (2D/\omega)^{1/2} \quad (6)$$

with C a constant (C = 4.6 for a flat, semi-infinite sample). In-depth thermal properties of the sample accordingly can be scanned by variation of the frequency. If ω is large, the scanning depth is restricted to a very small layer near the surface. Assume now for the moment that the extinction coefficient, E, of the sample is independent of temperature. Radiation emitted from the target spot neither obeys. Eq. (6) nor does it reflect a phase difference observed in the amplitudes of the local temperature variation if there is only conduction. Instead, volume power sources, now of periodically varying magnitude, are generated, on the statistical average, at the *constant* depth $l_m = 1/E$ (we will later come back to a discussion of l_m, the mean free path, that in general is not a constant but a statistical quantity). The value of l_m depends solely on the infrared optical properties of the sample material, neither on magnitude of the disturbance nor on its frequency (at least in linear optics). Both extinction coefficient, E, or depth, l_m, constant means that both infrared-optical quantities are independent of frequency ω. There could be a *spectral* frequency or wavelength dependence of E and l_m, but this is of course not correlated with the frequency by which the heat source is modulated. Periodically varying volume power sources will be responsible for triggering conduction deep in the interior of the sample if E is not too large and regardless whether the frequency, ω, of the modulated energy source, or the penetration depth, δ(ω), would be small or not. This aspect will again be discussed in Sect. 7 of this paper.

Radiation penetrating through transparent spectral windows or by absorption/remission and scattering of course arrives first at the detector. These signals will in almost all cases not be recognized by the detector (most presently developed infrared detectors also would not be able to do so). But evolution of the radiation signal will become the more important the shorter the pulses provided by advanced laser radiation sources, and the faster the response of the detectors.

Accordingly, contrary to the assumption of Parker and Jenkins, the non-reflected part of incident radiation is absorbed not at the sample surface, x = 0, but at *finite* locations beneath the surface. This creates radiative power sources that describe radiation remitted at all thermal wavelengths. Remitted radiation may be considered as a true black body spectral distribution. These sources contribute to evolution of local temperature distribution in the sample, which means immediately after absorption of the laser pulse, i. e. generation of local radiative power sources, thermal conduction will be initialized; possibly there are also phase changes of the thin film material.

Further, the incident spectral radiation neither could be spatially nor in time a Dirac pulse.

1.3. Conservation of Energy

Transfer of energy delivered from any kind of source to the thin film sample has to be rigorously described under conservation of energy. For this purpose, Fourier's differential equation, in three dimensions, reads

$$\rho\ c_p\ \partial T(\mathbf{x},t)/\partial t = \mathrm{div}(\mathbf{q}_{Cond}) \tag{7}$$

without sources. It describes coupling of the local temperature evolution, $T(\mathbf{x},t)$, to local variations of the conduction vector, $\mathbf{q}_{Cond}(\mathbf{x},t)$. If there is also radiation, as a second heat transfer mode, Eq. (7) has to be extended to a full "energy equation"

$$\rho\ c_p\ \partial T(\mathbf{x},t)/\partial t = \mathrm{div}[\mathbf{q}_{Cond}(\mathbf{x},t) + \mathbf{q}_{Rad}(\mathbf{x},t)], \tag{8}$$

still without sources, the solution of which delivers the local temperature evolution now with respect to the divergence of the total thermal transport vector, $\mathbf{q}_{Total} = \mathbf{q}_{Cond} + \mathbf{q}_{Rad}$.

The divergence $\mathrm{div}(\mathbf{q}_{Cond})$ of the vector $\mathbf{q}_{Cond}$ incorporates the thermal conductivity, λ, that may depend on temperature and possibly on local variations of materials composition.

For the following discussion, we will have to distinguish between solutions of the equation of radiative transfer (see standard volumes on this subject) and of the energy equation for

i. the general case (3-dimensional heat transfer) and
ii. the special case of flash experiments.

The temperature evolution, $T(\mathbf{x},t)$ is calculated from $\partial T(\mathbf{x},t)/\partial t$ provided the term $\mathrm{div}(\mathbf{q}_{Cond} + \mathbf{q}_{Rad})$ can appropriately be modelled. Unfortunately, this turns out to be difficult so that a variety of approximations has been proposed.

Consider for simplicity the time interval $t > t_p$, where t_p denotes the duration of the heat pulse. In $\mathrm{div}(\mathbf{q}_{Cond} + \mathbf{q}_{Rad}) = \rho\ c_p\ \partial T(\mathbf{x},t)/\partial t$, it is the temperature profile, $T(\mathbf{x},t)$ that couples both quantities if they are functions of temperature. If instead $\mathbf{q}_{Cond}$ would be calculated separately from $\mathbf{q}_{Rad}$, without taking notice of the temperature distribution, which means, without observation of the energy balance, Eq. (8), and both vectors simply added, conduction would be decoupled from radiation.

Traditional literature on coupled conductive and radiative heat transfer discusses extensively under which conditions this "additive approximation" is acceptable, e. g. in conductive, purely scattering, or in conductive, absorbing and/or scattering but non-transparent materials. However, thin films even if they have strong absorption properties may not be non-transparent at all.

For flash experiments, a frequently applied approximation replaces $\mathrm{div}(\mathbf{q}_{Cond})$ by $\lambda\ \partial^2 T(\mathbf{x},t)/\partial x^2$, assuming the conductivity, λ, is constant. Flash experiments applying e. g. the Parker and Jenkins approach, accordingly should be performed with only small temperature increase under radiation, to fulfil this condition.

Another approximation concerns the term $\mathrm{div}(\mathbf{q}_{Rad})$. Frequently, it is assumed $\mathrm{div}(\mathbf{q}_{Rad})$ can be replaced by simply taking the derivative of Beer's directional intensity decay law, in one dimension,

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