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One- and Two-Temperature Models in Solids Under Laser Pulse Irradiation Which Effectively Present as Two-Photon Absorption Coefficient Anisotropy

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In this paper we have developed a semi-analytical model to study the temperature distributions in infra red (IR) optical materials heated by laser pulses. Our model takes into account two-photon absorption and calculations are based on a three-dimensional model of heat diffusion in solids using the integral transform method. We found out that the rigorous semi-analytical expression of the thermal field when one considers both one and two-photon absorption. The model is valid for any laser-solid system whose interaction can be described by the generalized Beer-Lambert law. Specific results are presented for an application of the model to GaAs sample. We discovered that two-photon absorption can produce detectable temperature variation. The main goal of the present paper was to correlate the thermal field with the two-photon absorption anisotropy.

Keywords: Heat equation, nonlinear optical materials, laser pulses, multi-photon absorption, two-photon absorption, two-temperature model

1 INTRODUCTION

Multi-photon absorption processes in semiconductors have been the subject of extensive theoretical and experimental investigations since the advent of the laser over five decades ago. Nonlinear absorption plays a crucial role in

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high-power laser technology, as well as in many fundamental aspects of solid-state physics. Moreover, in the last decade the employment of semiconductor components as nonlinear elements in optical communication and information processing systems has become extensive and so a more precise and accurate knowledge of their nonlinear optical properties is needed. For this reason we consider in this context a study of two-photon absorption coefficients in crystalline solids, which are of unambiguous practical importance for a wealth of applications.

Two-photon absorption has proved to be a powerful spectroscopic tool: first, it is the only method available in the cases when one-photon absorption is forbidden by the selection rules and second, it gives complementary material information even in the cases in which the linear absorption is allowed because it permits the study of the crystalline volume and not only of its surface. two-photon absorption processes can also give fundamental information about the energy-band structures not easily obtained by using linear techniques.

The rapid development of high-power continuous wave (CW) and pulsed CO_2 lasers [1] was and still is limited more and more by the susceptibility of windows and mirrors to damage. The ideal high-power laser window material would have a low absorption coefficient at the laser wavelength. In this paper we consider the heating of a solid sample by laser pulses. In order to understand better the physical absorption process it is necessary to consider multi-photon processes. We will take into account one and two-photon absorption coefficients. From theory and experiment we know that the higher-order multi-photon transition probabilities decrease rapidly with increasing order.

During the last ten years advances in laser processing have encouraged the development of model calculations of spatial and temporal temperature fields in laser heated solids. In recent years the integral transform method has been successfully applied to the studies of thermal fields in laser-solid interaction [2–5]. Elaborated mathematical models, both analytical and numerical, have been developed to describe the heat flow under a large number of simplifying approximations and assumptions regarding the laser beam and the sample.

The role of two-photon absorption in semiconductors is of increasing importance as semiconductor components become employed as nonlinear elements in optical communications and information processing systems. two-photon absorption provides a fundamental limitation for optical switching in the transparent spectral region of semiconductors. two-photon absorption also has potential applications as a nonlinear spectroscopic technique, providing complementary material information to one-photon-absorption measurements.

In this work we carried out three-dimensional model calculations in which full account is taken not only of the two-photon absorption, but also of the time and space characteristic of the laser beam as the heating source. We have also discussed the influence of the two-photon absorption coefficient in

establishing the thermal profiles. In addition we discuss the influence of the heat transfer coefficient and consider the anisotropy of the two-photon absorption coefficient. Last we make a comparison with the very powerful "two-temperature model" [6].

2 THE SEMI-ANALYTICAL MODEL

Initially we have to say that we are dealing with a semi-analytical model. But, because it is very strongly convergent after just 10 iterations we can say that we are dealing with an analytical model. So, for our work the macroscopic heat equation, is employed to investigate the temperature field in a semiconductor [5] exposed to a Gaussian or flat spatial profile and a rectangular nanosecond pulse. The sample is supposed to be homogeneous and therefore there is no angular dependence of the temperature variation. The equation describing the heat diffusion inside a cylindrical IR optical material irradiated by a laser beam centred to the probe is fully described by the partial differential equation:

$$\frac{\partial^2 T}{\partial r^2}+\frac{1}{r}\frac{\partial T}{\partial r}+\frac{\partial^2 T}{\partial z^2}-\frac{1}{\gamma}\frac{\partial T}{\partial t}=-\frac{A(r\ ,\ z\ ,\ t)}{k} \tag{1}$$

where k is the thermal conductivity of the sample, γ is the thermal diffusivity of the sample given by $\gamma=\frac{k}{c\cdot\rho}$, c is the heat capacity of the sample, ρ is the mass density of the sample. Here $A(r; z, t)$ represents the heat rate (per unit volume and time) produced by the laser in the sample. The temperature T is a function of $(r; z, t)$ and is defined here as a temperature variation rather than an absolute temperature: $T(r, z, t)=T_{fi}-T_{in}$, where T_{fi} and T_{in} are the final and the initial absolute temperature of the sample. If we consider a linear heat transfer at the sample surface (the "radiation" boundary condition), we have

$$k\frac{\partial T(r\ ,z\ ,t)}{\partial r}\Big|_{r=b}+hT(b\ ,\ z\ ,\ t)=0 \tag{2a}$$

and

$$k\frac{\partial T(r\ ,\ z\ ,t)}{\partial r}\Big|_{z=0}-hT(r,\ 0\ ,\ t)=0 \tag{2b}$$

and

$$k\frac{\partial T(r\ ,z\ ,t)}{\partial r}\Big|_{z=a}+hT(r\ ,\ a\ ,\ t)=0 \tag{2c}$$

where h is the heat transfer coefficient of the sample surface; a and b are the thickness and radius of the sample, respectively. In the presence of both one- and two-photon absorption, described by coefficients α and β, respectively, the change in the intensity of the light as it passes through the sample is given

by generalized Beer-Lambert law: $\frac{dI}{dz} = -\alpha \cdot I - \beta \cdot I^2$ when free carrier absorption is negligible. The two-photon absorption coefficient can be calculated in second-order perturbation theory in terms of transition probabilities W_2: $\beta = 4W_2\hbar\omega / I^2$. The transition probability from an initial valence band to a final conduction band, accompanied by the simultaneous absorption of two photons, each of frequency W can be calculate as

$$W_2 = \frac{2\pi}{\hbar}\int \left| \frac{\sum_i <\psi_c|H|\psi_{i>}<\psi_i|H|\psi_{v>}}{(E_i - E_v - \hbar\omega)} \right|^2 \times \delta(E_c(\vec{k}) - E_v(\vec{k}) - 2\hbar\omega)\frac{d^3\vec{k}}{(2\pi)^3} \qquad (3)$$

where Ψ_i is the Bloch function of the crystalline electrons in band i whose energy is E_i and H is the interaction Hamiltonian. The calculation of numerical values for two-photon transition probability is extremely difficult. The straightforward solution to the Beer-Lambert equation is

$$\frac{I}{I_0} = \frac{(1-R)^2 \exp(-\alpha z)}{1+\beta \cdot I_0(1-R)[1-\exp(-\alpha z)]/\alpha} \qquad (4)$$

where R is the reflectivity at the wavelength of the incident radiation and z is the thickness of the sample.

In order to take into account the two-photon absorption the heat rate per unit volume and time will be determined by the laser intensity according to Beer-Lambert's law:

$$A(r,z,t) = (\alpha \cdot I_{00}(r,z) + \beta I_{00}^2(r,z)) \cdot (h(t) - h(t-t_0)) \qquad (5)$$

where t_0 is the pulse duration and $h(t)$ is the step function. The solution of the heat equation is

$$T_{\alpha\beta}(r,z,t) = \sum_{i=1}^{\infty}\sum_{j=1}^{\infty}\left[\frac{1}{\mu_i^2+\lambda_j^2} \cdot f_{\alpha,\beta}(\mu_i,\lambda_j) \cdot (1 - e^{-\theta_{ij}^2 t} - (1 - e^{-\theta_{ij}^2 (t-t_o)}) \cdot h(t-t_0)\right] \times K_r(\mu_i, r) \cdot K_z(\lambda_j, z) \qquad (6)$$

where

$$\theta_{ij}^2 = \gamma(\mu_i^2 + \lambda_j^2) \qquad (7a)$$

and

$$f_{\alpha,\beta}(\mu_i, \lambda_j) = \frac{1}{kC_iC_j}\int_0^a\int_0^b (\alpha I_{00} + \beta I_{00}^2) r \cdot K_r(\mu_i, r) \cdot K_z(\lambda_j, z) dr dz \qquad (7b)$$

The coefficients C_i and C_j are the normalizing coefficients. The Eigenvalues μ_i and λ_j correspond to the Eigenfunctions $K_r(r,\mu_i)$ and $K_z(z,\lambda_j)$, respectively.

The integral operators corresponding to the Eigenfunctions $K_r(r,\mu_i) = J_0(\mu_i r)$ and $K_z(z,\lambda_j) = \cos(\lambda_j z) + \frac{h}{\lambda_j \cdot k}\sin(\lambda_j \cdot z)$ are normalized by the following coefficients:

$$C_i = \int_0^b rK_r^2(r,\mu_i)dr = \frac{b^2}{2\mu_i^2}\left(\frac{h^2}{k^2} + \mu_i^2\right)J_0^2(\mu_i b) \qquad (8a)$$

and

$$C_j = \int_0^a K_z^2(z,\lambda_j)dz = \frac{1}{4\lambda_j^3}\left(2\frac{h}{k}\lambda_j + 2a\frac{h^2}{k^2}\lambda_j + 2a\lambda_j^3 - 2\frac{h}{k}\lambda_j\cos[2a\lambda_j] - \frac{h^2}{k^2}\sin[2a\lambda_j] + \lambda_j^2\sin[2a\lambda_j]\right) \qquad (8b)$$

The Eigenvalues μ_i and λ_j are determined from the boundary conditions by

$$\frac{h}{k}J_0(\mu_i b) - \mu_i J_1(\mu_z b) = 0 \qquad (9a)$$

and

$$2\cot(\lambda_j a) = \frac{\lambda_j k}{h} - \frac{h}{\lambda_j k} \qquad (9b)$$

3 THE RESULTS OF THE PROPOSED SEMI-ANALYTICAL MODEL

In the previous section the heat diffusion equation was analytically solved in order to determine the temperature field inside a semiconductor sample. One cylindrical sample made of GaAs is now considered with dimensions of radius, b, of 10 mm and a thickness, a, of 4 mm. The sample was supposed to be irradiated by a 250 ns pulse TEM_{00} CO_2 laser beam of 2 mm diameter and with a power of 100 W. We consider for the GaAs that $\beta = 15$ cm/MW. Under these conditions the temperature fields plotted in Figures 1 to 4 correspond to $z = 0$, $h = 6 \times 10^{-7}$ W/mm^{2}K and nil surface absorption.

A typical temperature distribution *versus* time and radial coordinate for the 250 ns pulse TEM_{00} CO_2 laser beam is shown in Figure 1, where it can be seen that the temperature distribution reaches its maximum in the sample centre - at the point where the power density has its maximum. The same temperature field as in Figure 1 is presented in Figure 2, but for a 0.2 ms pulse TEM_{00} CO_2 laser beam with 50 W power. As expected, the general shape of the thermal fields shown in Figure 2 are very similar but the pulse duration of the laser beam determines the temporal evolution of the temperature field. One can easily notice that in Figure 1 the cooling process is faster in comparison with that shown in Figure 2. Figure 3 presents the same temperature field as in Figure 1 but for $h = 3 \times 10^{-7}$ W/mm^{2}K. As one can see from Figure 3 the general increase in temperature with the shape of the field

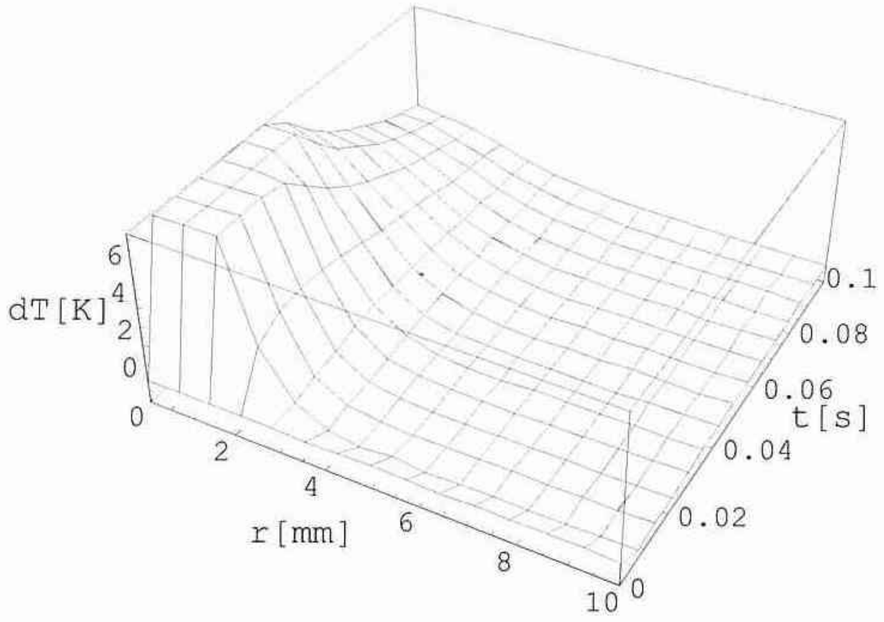

FIGURE 1
Computed temperature field inside the GaAs sample probe when exposed to a 250 ns pulse TEM_{00} CO_2 laser beam with a power of 100 W.

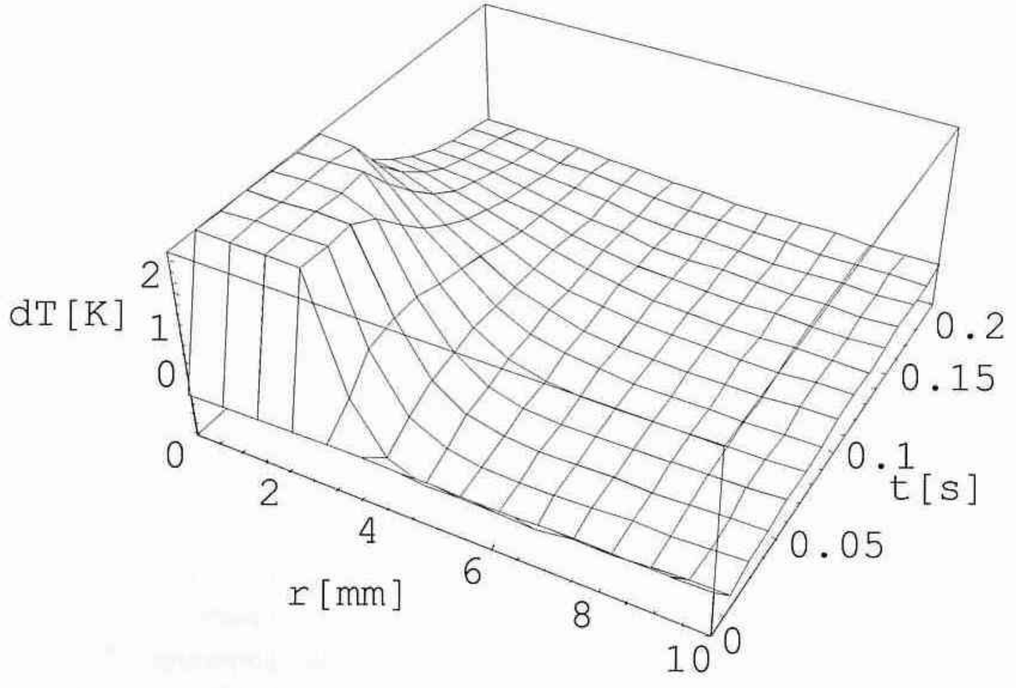

FIGURE 2
Computed temperature field inside the GaAs sample probe when exposed to a 0.2 ms pulse TEM_{00} CO_2 laser beam with a power of 50 W.

temperature remaining the same. The explanation for this observation is that h is the key factor which determines the energy losses by radiation and convection through the surface and therefore establishes a steady state regime where there is an equilibrium between the energy absorbed from the laser and

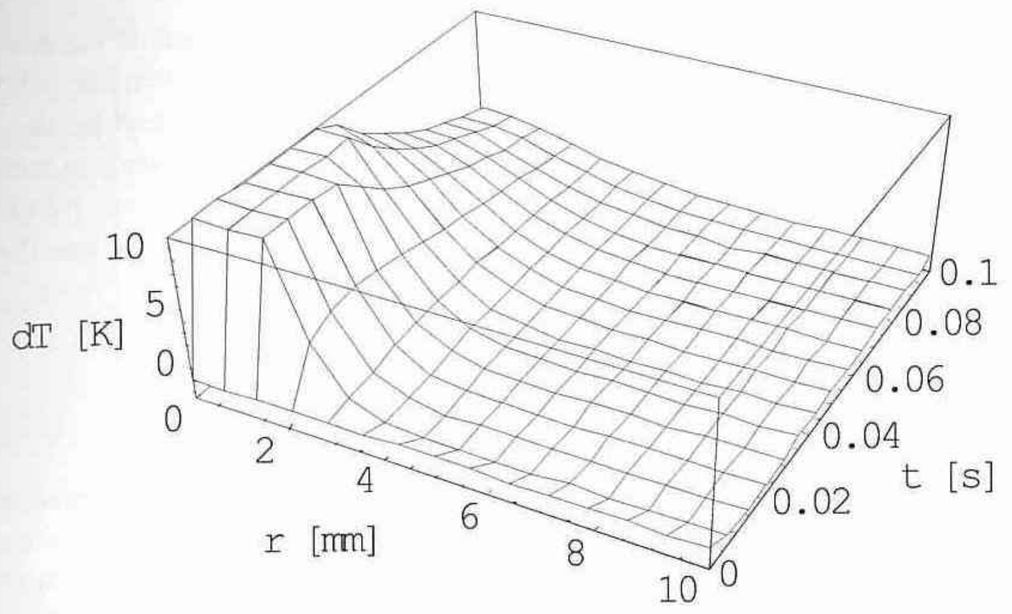

FIGURE 3
Computed temperature field inside the GaAs sample probe for $h = 3 \times 10^{-7}$ W/mm^{2}K when exposed to a 250 ns pulse TEM_{00} CO_2 laser beam with a power of 100 W.

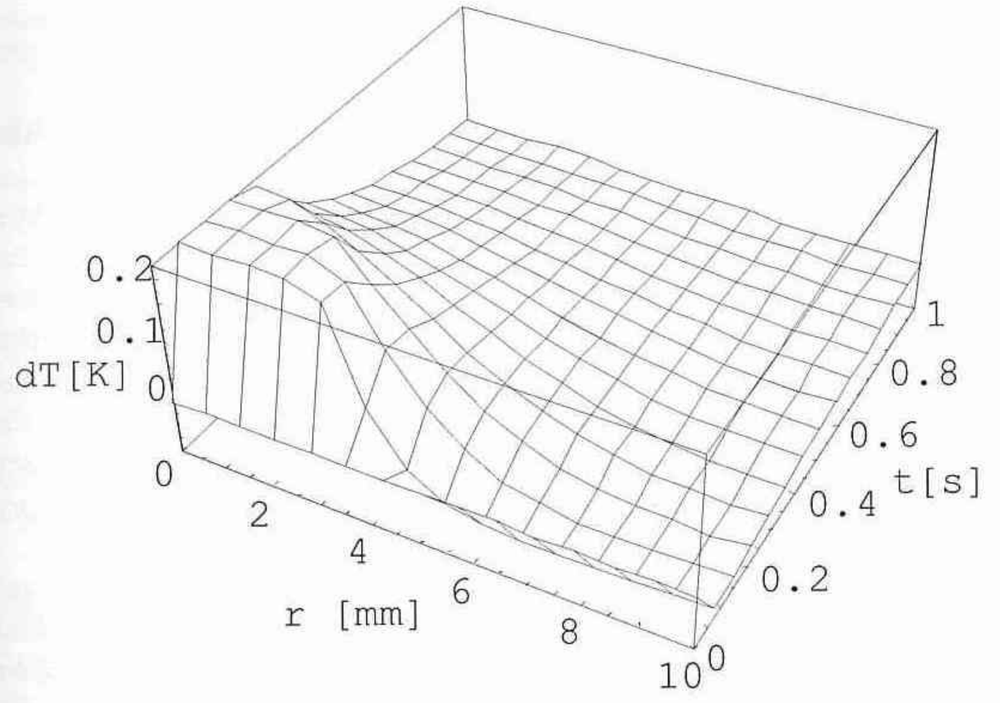

FIGURE 4
Computed temperature field inside the GaAs sample probe when exposed to a 250 ns pulse TEM_{00} CO_2 laser beam with a power of 100 W when $\alpha = 0$ and the two-photon absorption coefficient, β, is 15 cm/MW.

the energy losses. We present in Figure 4 the temperature field produced only by the two-photon absorption coefficient ($\alpha = 0$). One can notice from Figure 4 that the temperature field is less than 5% from the maximum temperature field produced in the case of $\alpha \neq 0$ shown in Figure 1.

This model assumes that the laser beam interacts with the sample *via* one- and two-photon absorption coefficient. Our study concludes that for solid materials the heat equation has a rigorous analytical expression and the two-photon absorption produce a detectable temperature field in comparison with the field produced by the one-photon absorption. The temperature profile model could be applied to any other semiconductor introducing the specific constants of the material.

4 THE TWO-TEMPERATURE MODEL

For the above we can apply more realistic models such as the two-temperature model. The spatial and the temporal evolution of the electron and the lattice temperatures can be described by the following one-dimensional, two-temperature diffusion models [10]:

$$C_e \frac{\partial T_e}{\partial t} = -\frac{\partial Q(z)}{\partial z} - \gamma(T_e - T_i) + S \tag{10a}$$

and

$$C_i \frac{\partial T_i}{\partial t} = \gamma(T_e - T_i) \tag{10b}$$

where z is the direction perpendicular to the target surface; $Q(z)$ is the heat flux and is given by $Q(z) = -k_e \partial T_e / \partial z$; S is the laser heating source term and is given by $S = I(t)A\alpha \exp(-az)$ with $I(t)$ being the laser heating source; A and α are the surface absorption coefficient and the material absorption coefficient, respectively; C_e and C_i are the heat capacities of the lattice and electron subsystems, respectively; γ is the parameter characterizing the electron-lattice coupling and k_e is the electron thermal conductivity. We used the simplest approximation of the two-temperature models in accordance with Nolte *et al* [6] and the results are shown in Figure 5.

Using the solutions of the thermal models one can easily find the one- and two-photon absorption coefficient using the like "source" term intensities given in the work of Dvorak *et al* [7]. Indeed, as one can se from Figure 5, we have obtained good agreement between the one-and two-temperature model.

For two-photon absorption we have the following source terms when the probe was polarized parallel to the pump:

$$\partial I_p / \partial \eta = -\beta(\theta) I_P^2 - 2\beta(\theta) I_e I_p - \alpha I_P \tag{11a}$$

and

$$\partial I_e / \partial \eta = -\beta(\theta) I_e^2 - 2\beta(\theta) I_e I_p - \alpha I_e \tag{11b}$$

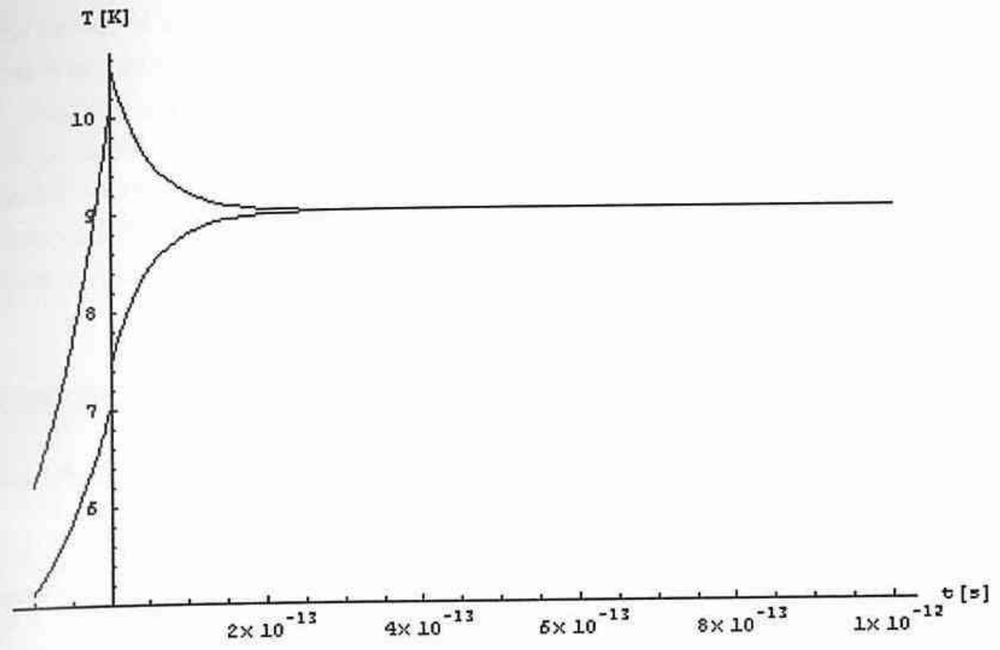

FIGURE 5
The thermal field *versus* time at the point x = 0 mm, y = 0 mm when the laser is acting in the flat mode during a 10 s exposure time. The simulation has been performed using two-temperatures model (the upper curve represents the electron temperature while the lower curve represents the lattice temperature).

When the measurements were then repeated with the probe polarization perpendicular to the pump we obtained

$$\partial I_p / \partial \eta = -\beta(\theta - \pi/2) I_P^2 - 2\beta^{\perp}(\theta) I_e I_p - \alpha I_P \tag{12a}$$

and

$$\partial I_e / \partial \eta = -\beta(\theta) I_e^2 - 2\beta^{\perp}(\theta) I_e I_p - \alpha I_e \tag{12b}$$

Using the most elementary algebra one can deduce the variation of the two-photon absorption coefficient with the angle θ.

5 THE ANISOTROPY OF THE TWO-PHOTON ABSORPTION COEFFICIENT

Using the notations from the work of Dvorak *et al* [7] we have: $\beta(\theta) = 19.5(1 + 0.38 \sin^2 2\theta)$ cm/GW, which allows the curve in Figure 6 to be plotted. We can observe from Figure 6 that we can have a variation of nearly 30%, which means a variation in the temperature of approximately 0.06 K, which can be detected as shown in Figure 7. The reverse situation (when we know the thermal field and we want to find out the "source" term) is a little bit more difficult because we have to find out the source term using numerical methods.

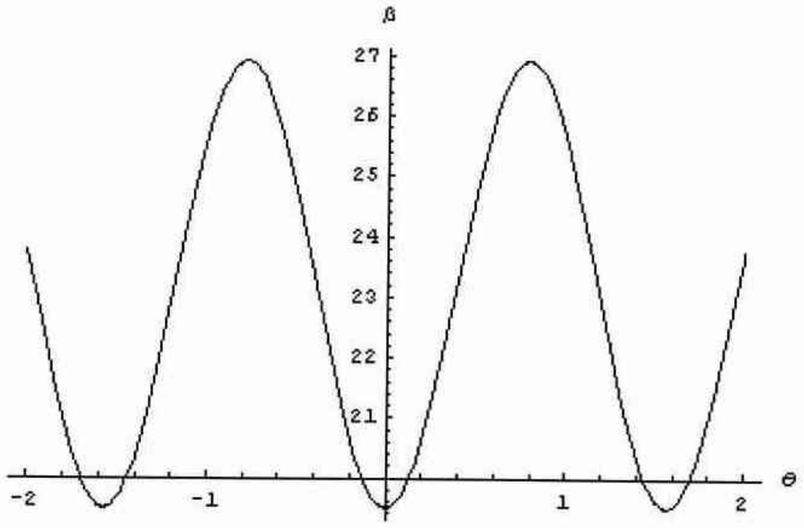

FIGURE 6
The dependence of two-photon absorption coefficient with the angle between the laser beam polarization and the GaAs axes.

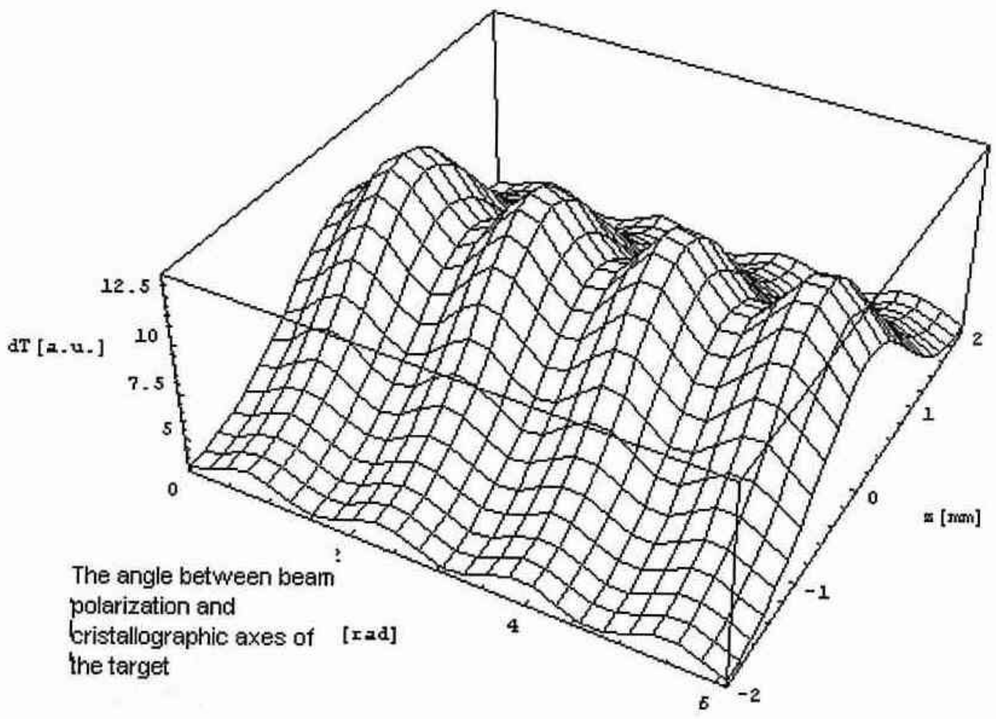

FIGURE 7
The temperature variation of the GaAs sample as a function of z (mm) and 0 (rad) for the situation presented in Figure 6.

6 CONCLUSIONS

The main conclusion to be made from this work is that laser calorimetry can be a powerful tool with regards to the connection between the laser polarization and the crystal orientation. We notice that the two-temperature model gives the same order of magnitude of the sample irradiated. We conclude that

our model is as general as possible (until the melting and vaporization starts and when our models "breaks down").

Another significant conclusion is that by using a two-temperature model is not possible at this time to have three-dimensional graphics, but with this model we obtained temperatures more close to reality. On the other hand, we observed that the thermal effects had a refreshment similar to those seen by other workers [8–15].

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