

## MATHEMATICAL MODELING OF THREE-PHOTON THERMAL FIELDS IN LASER-SOLID INTERACTION

S.-L. TSAO<sup>1</sup>, M. OANE<sup>\*2</sup>, LIN LI<sup>3</sup>, FL. SCARLAT<sup>2</sup>, F. SCARLAT<sup>2</sup>, C. OPROIU<sup>2</sup>,  
I. N. MIHAILESCU<sup>4</sup>

<sup>1</sup> *Institute of Electro-optical Science and Technology,  
National Taiwan Normal University, Taiwan*

<sup>2</sup> *National Institute for Laser, Plasma and Radiation Physics, Accelerator Laboratory,  
P.O. Box Măgurele-36, RO 76900, Bucharest, Romania*

<sup>3</sup> *Manchester Laser Processing Research Centre, UK*

<sup>4</sup> *National Institute for Laser, Plasma and Radiation Physics, Laser Dept.,  
P.O. Box Măgurele-36, RO 76900, Bucharest, Romania*

*\* Corresponding author: Mihai Oane, National Institute for Laser,  
Plasma and Radiation Physics, Accelerator Laboratory P.O. Box Măgurele-36,  
RO 76900, Bucharest, Romania, E-mail: oane.mihai@k.ro; Tel/Fax: 004021-457 42 43*

(Received July 15, 2005)

*Abstract.* In this paper we have developed an analytical model to study the temperature distributions in optical materials heated by laser pulses. Our model takes into account the three-photon absorption. The calculations are based on a three-dimensional model of heat diffusion in solids using the integral transform method. We find out the rigorous analytical expression of the thermal field when one considers three-photon absorption. The model is valid for any laser-solid system in which interaction can be described by generalized the Beer-Lambert law. Specific results are presented for an application of the model to  $\text{Al}_2\text{O}_3$  sample. We find out that three-photon absorption can produce detectable temperature variation.

*Key words:* three-photon absorption, temperature profiles,  $\text{Al}_2\text{O}_3$ .

### 1. INTRODUCTION

Multiphoton absorption (MPA) processes in semiconductors have been the subject of extensive theoretical and experimental investigations since laser was invented over four decades ago. Nonlinear absorption plays a crucial role in the high-power laser technology, as well as in many fundamental aspects of solid-state physics [1–4].

Moreover, as in the last decade the employment of semiconductor components as nonlinear elements in optical communication and information processing systems became very extensive, a more precise and accurate knowledge of their nonlinear optical properties is needed. This is why we consider that in this

context, the study of three-photon absorption coefficients in crystalline solids can be of an unambiguous practical importance for a variety of applications.

For example, three photon absorption is 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 which the linear absorption is allowed, because it permits the study of the crystalline volume and not only of its surface. Three photon processes can also give fundamental information about the energy-band structures not easily obtained by using linear techniques.

MPA techniques are used for producing population inversion in semiconductors, for controlling laser pulse intensities and pulse duration and for color-center creation in alkali-halide crystals. This mechanisms can also be used as negative-feedback elements in laser cavities and in studying processes associated especially with ultrashort laser pulses, as for example the laser-induced damage to optical components [2]. MPA provides a fundamental limitation in the transparency of optical window materials.

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 [6–9]. Elaborated mathematical models, both analytically and numerically, have been developed for describing the heat flow under a large number of simplifying approximations and assumptions regarding the laser beam and the sample.

The rapid development of the high-power cw and pulsed lasers is limited much more by such considerations as the susceptibility of windows and mirrors to damage. Ideal high-power laser window material would have low absorption coefficient at the laser wavelength. In this paper we consider the heating of a solid sample by a powerful laser pulse. In order to understand deeply of the physical absorption process, it is necessary to take into account multi-photon processes.

We carry out three-dimensional model calculations in which full account is taken not only of the three-photon absorption but also of the time characteristic of the laser beam as the heating source. We discuss the influence of the three-photon absorption coefficient in establishing the thermal profiles.

## 2. THE ANALYTICAL MODEL

In the present paper, the macroscopic heat equation is employed to investigate the temperature field in a semiconductor exposed to a laser with 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 centered to the probe is fully described by the partial differential equation:

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

where  $k$  is the thermal conductivity of the sample;  $\gamma$  represents the thermal diffusivity of the sample ( $\gamma = \frac{k}{c \cdot \rho}$ );  $c$  is the heat capacity of the sample;  $\rho$  is the mass density of 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 [7]), we have:

$$\begin{aligned} k \left. \frac{\partial T(r, z, t)}{\partial r} \right|_{r=b} + hT(b, z, t) &= 0, \\ k \left. \frac{\partial T(r, z, t)}{\partial r} \right|_{z=0} - hT(r, 0, t) &= 0, \\ k \left. \frac{\partial T(r, z, t)}{\partial r} \right|_{z=a} + hT(r, a, t) &= 0 \end{aligned} \quad (2)$$

where  $h$  is the heat transfer coefficient of the sample surface  $a$  and  $b$  are the thickness and the radius, respectively, of the sample. In the presence of one-photon and two-photon absorption and three photon absorption which are described by coefficients  $\alpha$ ,  $\beta$  and  $\gamma$  respectively, the change in the intensity of the light as it passes through the sample is given by [2]:  $\frac{dI}{dx} = -\alpha I - \beta I^2 - \gamma I^3$  when free carrier absorption is negligible.

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

$$A(r, z, t) = (\alpha \cdot I_{00}(r, z) + \beta I_{00}^2(r, z) + \gamma I_{00}^3) \cdot (h(t) - h(t - t_0))$$

where  $t_0$  is the pulse duration and  $h(t)$  is the step function.

The solution of the heat equation is (see Appendix A):

$$\begin{aligned} T_{\alpha\beta\gamma}(r, z, t) = \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} \left[ \frac{1}{\mu_i^2 + \lambda_j^2} \cdot f_{\alpha,\beta,\gamma}(\mu_i, \lambda_j) \cdot (1 - e^{-\theta_i^2 t} - (1 - e^{-\theta_j^2(t-t_0)}) \cdot h(t - t_0)) \right] \times \\ \times K_r(\mu_i, r) \cdot K_z(\lambda_j, z) \end{aligned} \quad (3)$$

where  $\theta_{ij}^2 = \gamma(\mu_i^2 + \lambda_j^2)$  and

$$f_{\alpha,\beta,\gamma}(\mu_i, \lambda_j) = \frac{1}{kC_i C_j} \int_0^a \int_0^b (\alpha I_{00} + \beta I_{00}^2 + \gamma I_{00}^3) r \cdot K_r(\mu_i, r) \cdot K_z(\lambda_j, z) dr dz.$$

The coefficients  $C_i$  and  $C_j$  are normalizing coefficients. The eigenvalues  $\mu_i$  and  $\lambda_j$  correspond to the eigenfunctions  $K_r(r, \mu_i)$  and  $K_z(z, \lambda_j)$ . The integral operators corresponding to the eigenfunctions  $K_r(r, \mu_i) = J_0(\mu_i \cdot r)$  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:

$$C_i = \int_0^b r K_r^2(r, \mu_i) dr = \frac{b^2}{2\mu_i^2} \left( \frac{h^2}{k^2} + \mu_i^2 \right) J_0^2(\mu_i b) \quad \text{and}$$

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

The eigenvalues  $\mu_i$  and  $\lambda_j$  are determined from the boundary conditions by the following equations [6–7]:

$$\frac{h}{k} J_0(\mu_i b) - \mu_i J_1(\mu_i b) = 0 \quad \text{and} \quad 2 \cdot \cot(\lambda_j a) = \frac{\lambda_j \cdot k}{h} - \frac{h}{\lambda_j \cdot k}.$$

### 3. RESULTS AND DISCUSSION

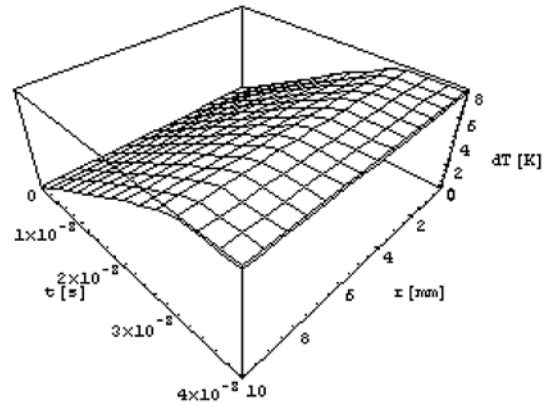
In the previous part of our paper, the heat diffusion equation was analytically solved in order to determine the temperature field inside a semiconductor sample.

One cylindrical sample was considered and its dimensions are the radius  $b = 10$  mm and the thickness  $a = 4$  mm. The sample was supposed to be irradiated by a 20 ns TEM<sub>00</sub> ( $\lambda = 0,694$   $\mu\text{m}$ ) laser beam with the power density  $10^6$  W/cm<sup>2</sup> and constant spatial power density profile.

The sample has the characteristics like in reference 2 and three-photon absorption coefficient  $\gamma = 0.1$  cm<sup>3</sup>/GW<sup>2</sup>. The temperature fields plotted in Fig. 1 correspond to  $z = 0$ ,  $h = 6 \times 10^{-7}$  Wmm<sup>-2</sup>K and a null surface absorption.

A typical temperature distribution versus time and radial coordinate for 20 ns TEM<sub>00</sub> laser beam is shown in Fig. 1. The temperature distribution reaches its

Fig. 1 – The temperature *versus* time and radial coordinate for 20 ns TEM<sub>00</sub> irradiation.



maximum after 20 ns and is constant in space. It is supposed that the radius of the laser beam is equal to the sample radius.

From our simulations, one can notice that the temperature field is greater by 0.2 K in comparison with the case when  $\gamma = 0$ . The model applied in this paper is one which assumes that the laser beam interacts with the sample *via* one-, two- and three-photon absorption. Our study concludes that the heat equation has a rigorous analytical expression for solid materials, and the three-photon absorption produces a detectable temperature field variation in comparison with the field produced by the one- and two-photon absorption.

The temperature profile model could be applied to any other solid sample introducing the specific constants of the material.

## REFERENCES

1. Maurizio Dabbicco and Ida Maria Catalano, Measurement of the anisotropy of the two-photon absorption coefficient in ZnSe near half the band gap, *Optics Communications*, **178**, (2000) 117–121.
2. Vaidya Nathan, A. H. Guenther and S. S. Mitra, Review of multiphoton absorption in crystalline solids, *J. Opt. Soc. Am. B*, Vol. **2**, No. 2, p. 294 (1985).
3. D. C. Hutchings and B. S. Wherrett, Theory of anisotropy of two-photon absorption in zinc-blende semiconductors, *Physical Review B*, Volume **49**, number 4, p. 2418, January (1994).
4. Mark D. Dvorak, W. Andreas Schroeder, David R. Andersen, Arthur L. Smirl and Brian S. Wherrett, Measurement of the Anisotropy of Two-Photon Absorption Coefficients in Zincblende Semiconductors, *IEEE Journal of Quantum Electronics*, VOL. **30**, NO. 2, p. 256, February (1994).
5. A. Vaidyanathan, A. H. Guenther and S. S. Mitra, Band-structure calculations of the two-photon absorption coefficients of GaAs, InP, CdTe and ZnSe, *Physical Review B*, Volume **24**, number 4, p. 2259, 15 August (1981).
6. T. Visan, D. Sporea and G. Dumitru, Computing method for evaluating the absorption coefficient of infrared optical elements, *Infrared Physics & Technology*, **39** (1998) 335–346.
7. M. Oane and D. Sporea, Temperature profiles modeling in IR optical components during high power laser irradiation, *Infrared Physics & Technology*, **42** (2001) 31–40.

- 
8. M. Oane and D. Sporea, Heat transfer in IR optical components during CO<sub>2</sub> laser irradiation, *SPIE Proc.*, Vol. **4430** (2001), pp. 898–904.
  9. N. S. Koshlyakov, M. N. Smirnov and E. B. Gliner, *Differential equation of mathematical physics*, (North-Holland Publishing, Amsterdam, 1964).