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Nanosized Thin Films Surface Effects in Standard Calorimetry Using Low Laser Intensity: Experiment Versus Theory

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In the present work we investigated the thermal field distributions obtained by low intensity laser irradiation of layers with variable sizes: from nano to bulk size. We selected Fe as bulk materials and Au for the underlying material with variable size at nano-scale. We find out a method in order to decrease the sample temperature: to put between the Fe bulk surface and incident laser beam a nanoscale Au thin film. On the other hand, we can choose to irradiate the face of Fe bulk, especially when we have a much unpolished Fe surface, which leads us to an increase of the total bulk thermal field. The mathematical models based on the Green function and integral transform technique where employed. The main conclusion is that at low laser intensity in the ultraviolet (UV) range using the proposed method we can decrease the thermal fields with about 10 to 15%. We choose the scale of study to be 20 to 500 nm because 20 nm is the lower limit of Fourier equation validity and 500 nm is the upper limit like theoretical definition of nanoscale materials. The experiment data are in concordance with our theoretical predictions and other data from literature. From theoretical point of view, we obtain an analogue equation for the temperature variation of the nanosized thin film–bulk metal system in 'comparison' with the simple bulk metal situation. It is important to mention from the very beginning that we are dealing with nanosized materials and not with nanomaterials. Only the scale of the materials changes in an important way the heat conduction.

Keywords: Ultraviolet (UV) laser, low intensity irradiation, nanoscale, thermal field distributions, Green function method, integral transform technique

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1 INTRODUCTION

It is of great importance for spatial satellites or other devices which are exposed to considerable radiation conditions to be protected. It is understood today that the cosmic space is full of “violent” radiation. Also, the electronic devices in very important experiments (like nuclear fusion) should be resistant to radiation [1-3].

In this work we evaluated from the experimental point of view the thermal fields obtained by irradiation of a bulk layer of Fe in contact with an Au layer with thicknesses of 100 and 500 nm. The reasons for this selection was that 20 nm is the lower limit of viability of the Fourier heat equation. One assumes that the entire system is irradiated by a flat laser beam. The thin films of nanosized dimensions are obtained from experimental point of view by magnetron sputtering in radio frequency (12.35 MHz). The size of the nanosized thin film is measured with an accuracy of 3 nm using a quartz oscillator. Another experiment is to choose an unpolished Fe surface without any nanosized thin film cover. We also evaluate the thermal fields qualitatively using theoretical models. For the first experiments (with Au thin films) we use the Green function method, while for the second experiments (with bulk Fe target) we use the results of integral transform technique.

2 THE GREEN FUNCTION METHOD

For solving the heat equation we used the Green function method. We define the effective perpendicular thermal conductivity for this multilayer medium as [4, 5]:

$$K_{\perp} = \frac{l_1 + l_2}{\frac{l_1}{k_1} + \frac{l_2}{k_2}} = \frac{l}{\frac{l_1}{k_1} + \frac{l_2}{k_2}} \tag{1}$$

where l is the total thickness of this medium structure, and l_i and k_i are the thickness and thermal conductivity of layer i.

Similarly, the effective thermal conductivity for the heat that flows in a parallel direction to the layers planes is:

$$K_{\|} = \frac{k_1 A_1 + k_2 A_2}{A_1 + A_2} = \frac{k_1 A_1 + k_2 A_2}{A} \tag{2}$$

where A is the area of the near and far faces and A_i and k_i are area of layer i and thermal conductivity of layer i, respectively.

Since for most cases of interest, the heat flux is unidirectional due to the temperature gradient, the heat equation in steady state can be written as

$$\left\{\frac{\partial}{\partial x}\left[K_x \cdot \frac{\partial T}{\partial X}\right] + \frac{\partial}{\partial y}\left[K_y \cdot \frac{\partial T}{\partial y}\right] + \frac{\partial}{\partial z}\left[K_z \cdot \frac{\partial T}{\partial z}\right]\right\} = -A(x, y, z) \qquad (3)$$

where K_x, K_y, K_z, are thermal conductivities along the x-, y- and z-directions, while $A(x, y, z)$ is the source term of the heat equation. Because we consider here a solid without melting, we can neglect the blackbody radiation of the sample.

We introduce further a linear temperature by the expression

$$\theta(T) = \theta(T_0) + \left(1 / K(T_0)\right)\int_{T_0}^{T} K(T')\,dT' \qquad (4)$$

where $\theta(T)$ and $\theta(T_0)$ are the linearized temperatures for the system at a temperature T and T_0 substrate temperature, respectively. One may see that if $K(T)$ is constant, we have from Equation (4): $\theta(T) - \theta(T_0) = T - T_0$

One may observe that the variation of the linear temperature is equal to the variation of the real temperature when K is independent of T, an assumption which is reasonable for a weak interaction. The perpendicular linear temperature then for the heat equation is given by [4, 5]

$$\theta_\perp = \frac{P\left[1 - R_\perp(T)\right]}{(g)^{1/2}(\pi)^{3/2} K(T_0)} \int_0^\infty f\perp(\xi)d\xi \qquad (5)$$

where $g = {K_\perp}/{K_{||}}$ and $K_{||} - K(T)$ with $P = {}^{1}/{}_{w}$ being the normalized incident power, where I and w are the laser beam intensity and beam waist, respectively; $R_\perp$ is the surface reflectivity for an incident beam which is perpendicular to the layer structure of the substrate; and T_0 is the original substrate temperature before the laser irradiation. The function $f\perp(\xi)$ is defined by

$$f_\perp(\xi) = \frac{exp - \left[\left[X^2\left(\xi^2 + 1\right)\right] + \left[Y^2 / \left(\xi^2 + 1\right)\right] + \left(Z^2 / g\xi^2\right)\right]}{\left\{\left(\xi^2 + 1\right)\right\}} \qquad (6)$$

where $X = {x'}/{w'}$, $Y = {y'}/{w'}$, and $Z = {z'}/{w'}$ (w' is the waist of the incident laser beam and $w' \to \infty$ for a flat laser beam) are the normalized coordinates of the system.

From the literature [6], for the Au bulk sample we have: K=500 W/mK. For a 500 nm thickness of Au thin film we have K=450 W/mK, while for 100 nm we have K=350 W/mK. Finally, for 50 nm we have K=250 W/mK. The Fe bulk geometry is a cube with l=4 mm. If we introduce these values into Equation (1) we obtain

$$K_{\perp 1} = \frac{4}{\frac{4}{55} + \frac{500}{450}} \approx\sim 55W / mK \tag{7}$$

Similarly, we obtain for the 100 nm and 50 nm particles:

$$K_{\perp 2} \approx\sim 55W / mK \tag{8}$$

and

$$K_{\perp 3} \approx\sim 55W / mK \tag{9}$$

For the parallel cases we obtain

$$K_{||1} = 252.5\ W / mK \tag{10}$$

$$K_{||2} = 202.5\ {}^{W}\!/_{mK} \tag{11}$$

and

$$K_{||3} = 152.5\ {}^{W}\!/_{mK} \tag{12}$$

We calculate the temperature at points (2 mm, 0 mm, 2 mm). Using arbitrary units and combining Equation (5) with Equation (6) with the values from relations given in Equations (7) to (12), we obtain

$$\theta_{\perp} = \frac{C}{(K_{\perp} K_{||})^{1/2}} \tag{13}$$

where C is a constant; that is, considering that the system is in steady state regime where the thermal conductivity does not depend on temperature.

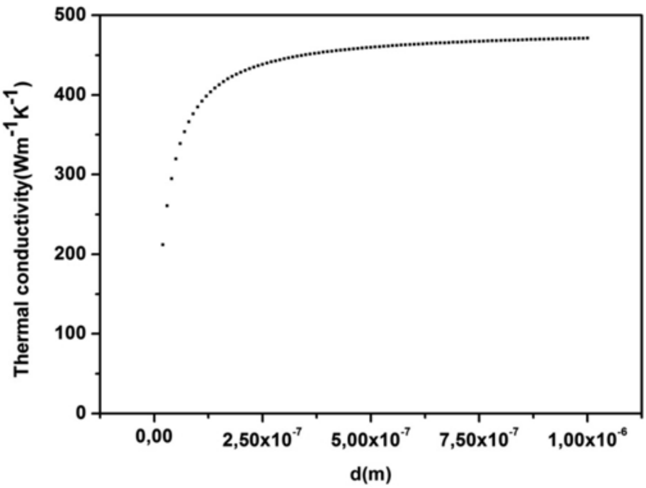

FIGURE 1
The plot of thermal conductivity *versus* the nanosized of Au thin films according to the theory developed by Holmes-Siedle and Adams [3].

We are now able to say that:

(i) The lower the dimension of the thin film in the limits 20 nm<Au layer thickness<500 nm has the lower the thermal conductivity. The thermal conductivity *versus* the thermal conductivity nanosized of thin films according to the theory developed in previous work [6] is also presented in Figure 1 and confirms our predictions; and
(ii) The lower the thermal conductivity the higher the Fe bulk temperature *T*. In a simple model [6] the nanosized thin film is consisted from electrons which fulfil the Schrödinger equation with periodic boundary conditions of the wave function, were is satisfied the Bloch equation. The lattice is formally defined as a quantized lattice vibration also known as phonons.

One may think at Equation (13) like the "nanosized" version of the well-known equation for the bulk thermal conductivity, which is equal to $\frac{C}{k}$, where $C = \frac{Qd}{At}$, with Q represented the amount of heat transferred in time t, on a cross section A in a material of thickness d.

3 THE INTEGRAL TRANSFORM TECHNIQUE

The integral transform technique is a theoretical method which allows us to solve the heat equation (*cf.* Equation (3)) using the Eigen functions and values [7-13]. We have the following relationship:

$$T(x,y,z,t) \sim A(x,y,z,t) = I(x,y) \times \left(\alpha \cdot e^{-\alpha z}(1-r_s) + r_s \cdot \delta(z)\right) \cdot \left(h(t) - h(t-t_0)\right) \quad (14)$$

where t_0 is the exposure time and *h(t)* is the step function. Here

$$\alpha(z) = \alpha + r_s.\delta(z) \quad (15)$$

is the total absorption coefficient, α is the bulk absorption coefficient and r_S is the surface absorption.
Our simulations have showed that [12] if

$$r_{S1} < r_{S2} \quad (16)$$

then we have

$$T(x,y,z,t,r_{S1}) < T(x,y,z,t,r_{S2}) \quad (17)$$

The meaning of Equation (17) is that the more rogues is a bulk surface the higher is the sample temperature (for the same incident laser beam).

4 EXPERIMENTAL DETAILS

We have made experiments of standard laser calorimetry using low intensity laser beam. The radiation source, that we have used in our set-up, was a pulsed Nd:YAG laser (Surelite II; Continuum) working at 355 nm and having a 6 ns pulse length at 10 Hz frequency repetition rate and a domain of energy at this wavelength (0.6/130 mJ). We have used permanently a monitoring system for the emitted laser beam energy. The laser beam has a top-hat spatial distribution of intensity. The beam was focused with a lens of 240 mm focal length on the surface of the probe.

In Figure 2 we present the schematic arrangement of our calorimetric experiments (the Fe bulk is a cube with edge of 4 mm). In Figure 3 we show the experimental results, during laser irradiation of the target.

5 CONCLUSIONS

It was observed that in case of Au particles ranging from 20 nm to 500 nm, the smaller is the thermal conductivity the larger is the temperature variation. This is an important point, because it opens up the possibility to design ther-

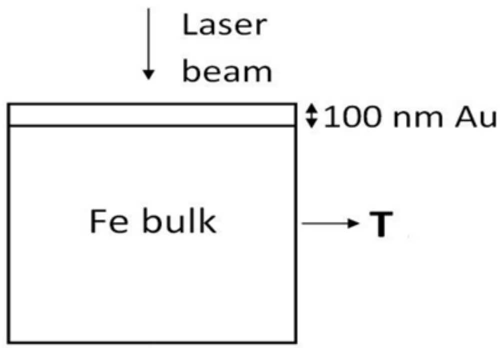

FIGURE 2
Schematic representation of the experimental arrangement for the laser–Fe bulk (a cube of 4 mm dimension) interaction, when on the Fe bulk is a 100 nm Au thin film.

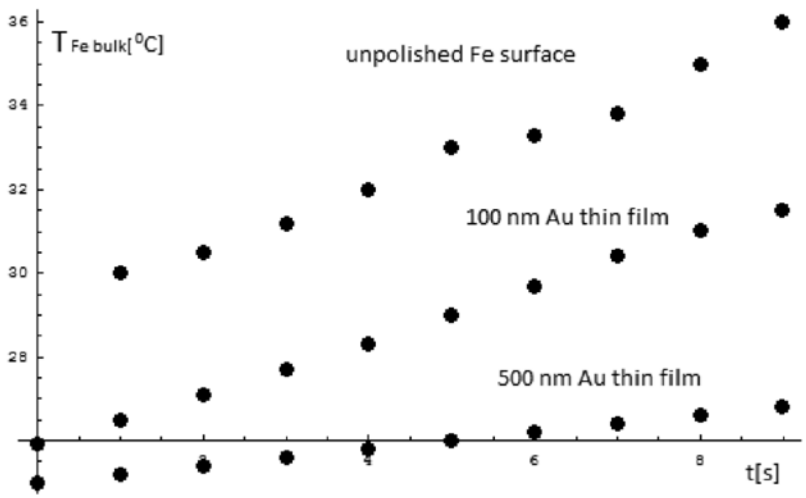

FIGURE 3
The experimental data for the three situations: (i) unpolished Fe surface; (ii) 100 nm Au thin film coating; and (iii) 500 nm Au thin film coating.

mal shields in case of solids covered with nanosized thin films. This behavior is essential when passing from bulk to nano - size objects in practical application. For the electron beams with low incident power the results are identical, due to the fact that the absorption properties are similar to the case of low power laser beam interactions [9]. The theory of the integral transform technique and Green function method can be found in the literature [8, 9]. More powerful models of the subjects treated in the present paper can be found elsewhere [14, 15].

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NOMENCLATURE

i	Index (1; 2)
x, x', X	Spatial direction (m)
y, y', Y	Spatial direction (m)
z, z', Z	Spatial direction (m)
A	Area of the target (m^2)
A_i	Area of the Layer *i* (m^2)
A_1	Area of the Layer 1 (m^2)
A_2	Area of the Layer 2 (m^2)
$A(x, y, z)$	Source term of the heat equation (J/m^3s)
$A(x, y, z, t)$	Source term of the heat equation (J/m^3s)
C	Constant
d	Distance (m)
$f\perp$	Function defined by Equation (6)
g	The ratio between $K\perp$ and $K_{\|\|}$
$h(t)$	Step function
I	Laser beam intensity (J/m^2s)
$K\perp$	Thermal conductivity on the perpendicular direction (W/mK)
$K_{\|\|}$	Thermal conductivity on the parallel direction (W/mK)
K_x	Thermal conductivity on the *x*-direction (W/mK)
K_y	Thermal conductivity on the *y*-direction (W/mK)
K_z	Thermal conductivity on the *z*-direction (W/mK)
ki	Thermal conductivity of the Layer *i* (W/mK)
k_1	Thermal conductivity of the Layer 1 (W/mK)
k_2	Thermal conductivity of the Layer 2 (W/mK)
l	Total thickness of this medium structure (m)
l_1	Thickness of the Layer 1 (m)
l_2	Thickness of the Layer 2 (m)
P	Normalized incident power (W)
Q	Heat transfer (J/m^2s)
R	Surface reflectivity (%)
$R\perp$	Surface reflectivity on the perpendicular direction (%)
r_S	Surface absorption coefficient
t	Time (seconds)
T	Temperature (K)

T_0	Substrate temperature (K)
w	Laser beam waist (m)
w'	Incident laser beam waist (m)

Greek symbols

θ	Linearized temperature (K)
$\theta\perp$	Linearized temperature on the perpendicular direction (K)
ξ	Real variable of integration
α	Bulk absorption coefficient (m^{-1})
$\alpha(z)$	Total absorption coefficient (m^{-1})

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