Analele Universităţii de Vest din Timişoara
Vol. XLVI, 2005
Seria Fizică

LASER-MATTER INTERACTIONS ON FS LEVEL: ROLE OF DEMBER EFFECT[1]

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Abstract:
In the following, we give a theoretical analysis of the interaction of ultrashort (fs) high intensity laser pulses with solids and present some of our experimental results.. The treatment is done by considering an electronic subsystem configuration with both linear and nonlinear interactions. We analyzed the charged particle separation (electrons and holes) due to their different mobilities in the subsurface layer (laser beam penetration width). We calculated the value of the Dember[(1)] field in this region and found that the electric gradient strongly depends on the pulse duration. We demonstrate the appearence of a very high Dember field at moderate laser intensities in material like GaAs and V_2O_5[(2)]. Using femtosecond pump and probe technique, we demonstrate ultrashort optical characteristic changes, such as reflectivity, due to the rearrangement of the internal electron-hole subsystem under the influence of ultrashort laser pulses. The work was done with the cooperative support of the National Science Foundation and Hungarian Academy of Sciences (MTA-NSF/44073-MTA/86).

1. Introduction

However the interest to the femtosecond time regime has a colossal growth since the last decades, the ultrashort laser – material interactions are not yet completely understood. In our investigations we had focused on the femtosecond pulse laser induced lattice dynamics to study the processes of subsurface alternations. To analyze the interaction of matter and ultrashort laser pulses it is necessary to know all the properties of the laser pulses, and moreover the optical parameters of the solid target. One of the most informative parameter of the solid is the dielectric function ($\varepsilon(\omega)$) of the material. This parameter consists of a real and of an imaginary part, where the real part means the refractive index (n) of the matter. Changes in the optical properies of a material (such as reflectivity) along and after the laser irradiation

[1] Invited lecture presented to TIM-05 conference, 24-25 November, 2005, Timisoara

can provide an insight to the electron and lattice dynamics of the material. Optical properties depend on the structure, the symmetry and the electron configuration of the matter. Changes of the electron configuration can be described by measuring the changes of the optical properties of the material. The dielectric function of the material is determined by the electron configuration[3,4]. Therefore by measuring the changes of ε(ω), induced by the ultrashort laser pulse, the linear and nonlinear interactions of laser and matter can be described. To study the interaction process of ultrashort laser pulses with semiconductors we used the Two Temperature Model (TTM)[5].

Using this model two subsystems can be described by two different temperatures one for the electrons and and one for the lattice, respectively. The time scale of the energy relaxation within the electron subsystem is much shorter than the time necessary to transfer this energy to the lattice. Thus, the thermodynamical temperature of the electrons is different from the lattice temperature. Thermal diffusion of the hot electrons from the irradiated region into the bulk takes place simultaneously with the transfer of energy to the lattice (phonons) by collisions. Heat diffusion of the electron subsystem is much faster than the lattice-mediated diffusion. The thermal conductivity of the lattice can therefore be neglected on the time scale of electron-phonon coupling The coupling coefficient characterizes the strength of the interaction between the electron and phonon subsystems. We use commonly accepted notations. The interaction processes between the electron and phonon subsystems are described by the following differential equation.

$$\frac{\partial U_e}{\partial t} + \nabla(-k_e \nabla T_e) = -g_{eo}(T_e - T_o) + \left[(1-R)(\alpha + \Omega n) I(z,t) + (1-R)^2 \beta^2 I^2(z,t)\right]$$

$$\frac{\partial U_o}{\partial t} = g_{eo}(T_e - T_o) - g_{ol}(T_o - T_a)$$

$$\frac{\partial U_a}{\partial t} + \nabla(-k_a \nabla T_a) = g_{oa}(T_o - T_a)$$

where$U_o=C_oT_o$ and $U_a=C_aT_a$ are the optical and acoustic phonon energy, g is the coupling coefficient, Ω is the free carrier absorption coefficient.

The heat transport effects usually appear 1 ps after the laser pulse reaches the material. After the first ps the thermal relaxation appears and electron subsystem gets cold for the tenth ps. Therefore, it is easier to understand the laser-matter interaction on short time scale, using a fs pulse, because the heat transport effect does not have to be taken into consideration. The temperature (T) at a depth (x), below the surface of a material hit by an ultrashort laser pulse, is governed by the Quantum Heat Transport equation:

$$\frac{1}{v^2}\frac{\partial^2 T}{\partial t^2}+\frac{m}{\hbar}\frac{\partial T}{\partial t}=\frac{\partial^2 T}{\partial x^2}$$

where *v* is the thermal pulse propagation speed and *m* is the heat carrier mass. The two different solutions of the equation are the ballistic transport of non-thermalized electrons and the diffusive transport of thermalized electrons.

We can calculate the density changes of charged carriers versus time using the following equation:

$$\frac{\partial \rho}{\partial t}=D\frac{\partial^2 \rho}{\partial x^2}+I_0 e^{-\frac{4\ln 2\left(x-\frac{c}{n}t\right)^2}{\tau_p{}^2}}\alpha(1-R)e^{-\alpha x}-\frac{\rho}{\tau_l}$$

where I_0 is the peak laser intensity (adjusted to get max carrier density of 10^{18} 1/cm^3), c denotes the speed of light, n is the index of refraction, t_p is the FWHM pulse duration (Gaussian pulse), a is the absorption constant, R is the reflectivity, D is the diffusivity, τ_l is the electron-hole pair lifetime.

The diffusivity of the electrons is about 20 times larger than for the holes. The electrons diffuse faster into the material than the holes causing that the net charge density is positive near the surface and negative deeper into the material. This effect produces a strong „Dember" electric field that can be calculated as follows:

$$E(x)=\frac{e}{\varepsilon_r \varepsilon_0}\int_0^x\left[\rho_h(x')-\rho_e(x')\right]dx'$$

where e is the elementary charge, ε_0 is the permittivity of free space, ε_r is the dielectric constant, ρ_h is the density of holes, and ρ_e is the density of electrons.

2. Computational and Experimental Results

In our treatments we adopted different model calculations and we have estimated the ballistic and diffusive solution of the quantum heat transport equation.

The following figure (1) shows the summation of the two solutions for a 20 fs and for a 50 as pulse.

The density of holes and electrons induced by an intense laser pulse can be calculated for both cases by using the equation of the evolution of carrier densities. The results of the calculation for a 50 as pulse can be seen on figure 2

The difference in the densities caused by the different diffusivity can be seen on figure (2). Fig. 3 shows the evolution of the calculated density of holes and electrons in time. The value of the Dember electric field is calculated for 700 as, 5000 as, 20 fs and 500 fs pulse duration at different depth under the surface. Fig. 4 shows that the gradient of the electric field strongly depends ont he pulse duration, longer pulses induce larger electric gradient. It can be also seen on fig.4 that the shorter the pulses the maximum of the generated electric gradient is closer to the surface.

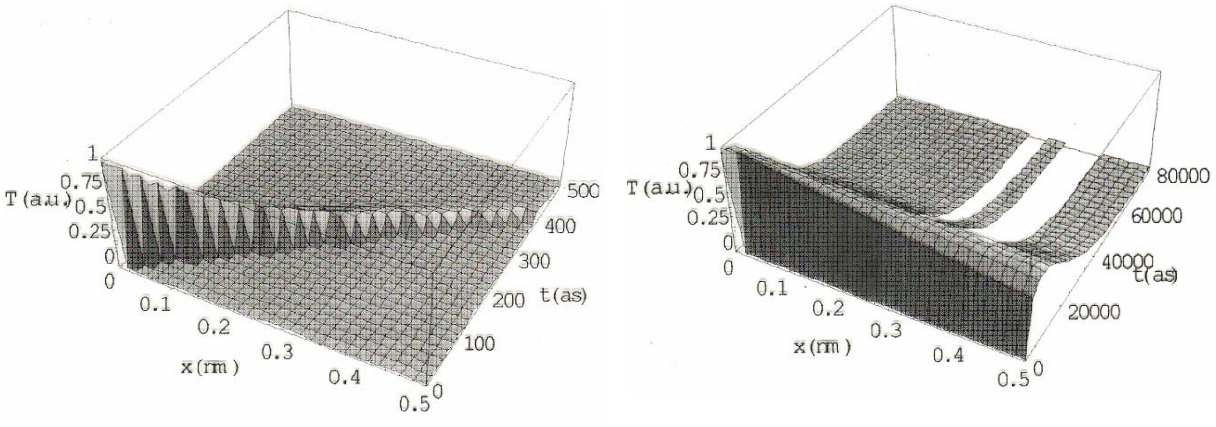

Fig.1. Sum of the ballistic and diffusive solutions as function of time and distance from the surface for a 50 as (left) and for a 20 fs (rigt) pulse

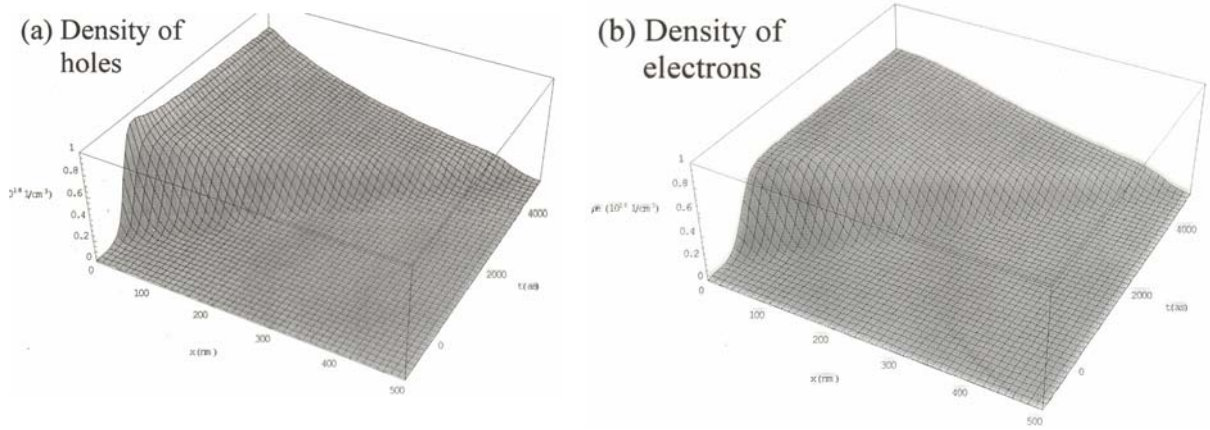

Fig.2. Density of charge carriers versus time and depth under the surface

We have studied the ultrafast lattice dynamics of V_2O_5 using a femtosecond pump and probe system. The central wavelength of the Ti:sapphire laser was 820 nm, the pump power was 25 mW, and the probe power was 2.5 mW. The pulse duration was 25 fs (FWHM).

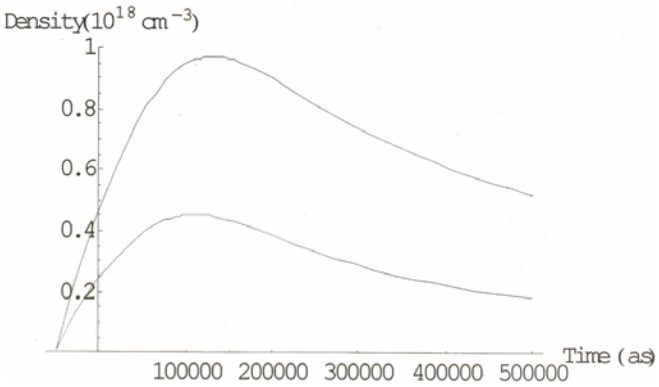

Fig.3. Charge cerrier densities as function of time at the surface. Dark line is the density of holes, the wan line is the density of electrons

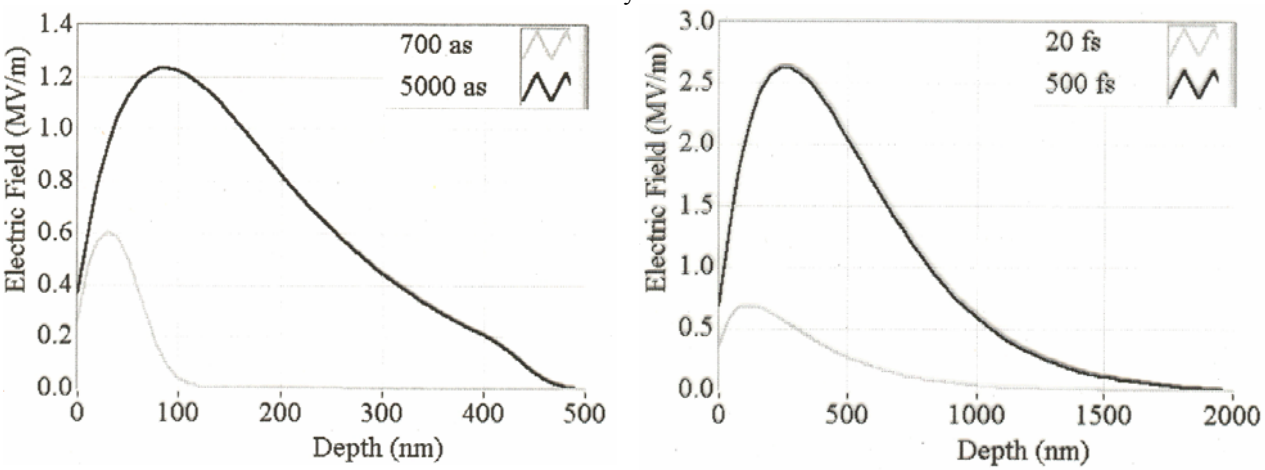

Fig.4. Charge cerrier densities as function of depth under the surface, induced by different pulse duration. Dark line is the density of holes, the wan line is the density of electrons

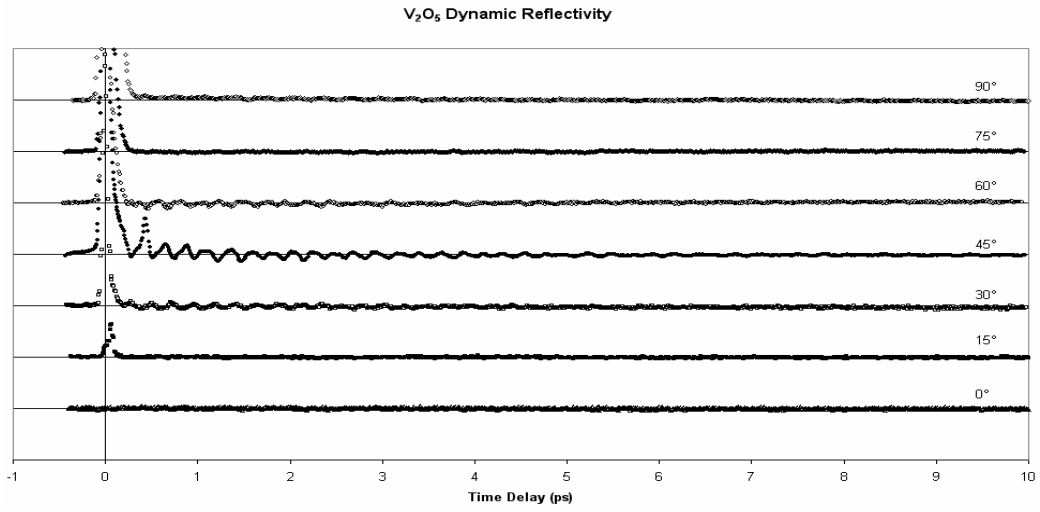

Fig. 5 Dynamic reflectivity changes in vanadium-pentoxid at different sample orientations versus time.

Our pump and probe measurements was made at different sample orientations, where the orientation means the angle between the pump beam polarization and the „a" axis of the V_2O_5. The sample was rotated along the „b"axis, besides that the incident angle of the pump

and probe beams was fixed. Fig. 5 shows the reflectivity changes of the sample at different orientations.We have found the largest peak at 90 degrees, and the largest amplitude oscillations at 45 degrees sample orientation. Making the Fourier transformation we have found that the oscillation of the reflectivity after the irradiation is the sum of two different wavenumbers (145 cm^{-1}, 104 cm^{-1}) oscillations. Each oscillation dominates in different time window: in the first five ps and from 5 ps to 10 ps, respectively.

Conclusions

As the summary of the temperature evolution it can be concluded that for a 50 as pulse the ballistic solution dominates and a short thermal pulse propagates into the medium. For a pulse of 20 fs duration the diffusive solution dominates and the temperature of the surface is high for a period of time approximately equal to the laser pulse duration. We have found that the gradient of the electric field strongly depends on the pulse duration, longer pulses induce larger electric gradient and as shorter the pulses are, as the maximum of the generated electric gradient is closer to the surface. We have studied the ultrafast lattice dynamics of V_2O_5 using a femtosecond pump and probe system and we have found a significant peak in reflectivity changes. At 45 degree sample orientation the largest amplitude reflectivity oscillation was observed.

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