International Journal of Thermophysics, Vol. 16, No. 6, 1995

Picosecond Thermal Pulses in Thin Gold Films

J. Marciak-Kozłowska,[1,2] Z. Mucha,[3] and M. Kozłowski[4]

Received March 1, 1995

In this paper it has been shown that, with the advent of lasers with a very short pulse duration, the effect of thermal wave propagation becomes important. To consider this effect, hyperbolic heat conduction in thin gold films was studied. It was shown that for heat fluxes of the order 10^8 W·cm^{-2}, a thermal wave is generated in thin gold films. The consideration of the hyperbolicity of heat transfer enables one to describe the temperature profile with one value of fluence.

KEY WORDS: hyperbolic heat diffusion; heat waves; pulse heating; thin metal films.

1. INTRODUCTION

Heat transport during fast laser heating of solids has become a very active research area due to the significant applications of short pulse lasers in the fabrication of sophisticated microstructures, syntheses of advanced materials, and measurements of thin-film properties. Laser heating of metals involves the deposition of radiation energy on electrons, the energy exchange between electrons and the lattice and the propagation of energy through media.

Ultrafast dynamics of hot electrons in metals has become an area of active investigation. The theoretical predictions showed that under ultrafast excitation conditions the electrons in a metal can exist out of equilibrium with the lattice for times of the order of the electron energy relaxation time [1]. Model calculations suggest that it should be possible to heat the

[1] Institute of Electron Technology, Al. Lotników 32/46, 02-668 Warsaw, Poland.
[2] To whom correspondence should be addressed.
[3] Institute of Fundamental Technological Research, Polish Academy of Sciences, Świętokrzyska 21, 00-049 Warsaw, Poland.
[4] Institute of Experimental Physics, Warsaw University, Hoża 69, 00-681 Warsaw, Poland.

electron gas to a temperature T_e of up to several thousand K for a few picoseconds, while keeping the lattice temperature T_l relatively cold. Observing the subsequent equilibration of the electronic system with the lattice allows one to study directly electron–phonon coupling under various conditions.

Several groups have undertaken investigations relating dynamics changes in the optical constants (reflectivity, transmissivity) to relative changes in electronic temperature. But only recently, the direct measurement of electron temperature has been reported.

In an earlier investigation [1], the temperature of hot electron gas in thin gold film ($l = 300$ Å) was measured, and a reproducible and systematic deviation from a simple Fermi-Dirac (FD) distribution for short time $\Delta t \sim 0.4$ ps were obtained. As stated in Ref. 1, this deviation arises due to the finite time required for the nascent electrons to equilibrate to a FD distribution. The nascent electrons are the electrons created by the direct absorption of the photons prior to any scattering.

In this paper, we investigate the relaxation dynamics of the electron temperature with the hyperbolic heat conduction equation (HHC) [2]. Conventional laser heating processes which involve a relatively low energy flux and long laser pulse have been succesfully modeled in metal processing and in measuring thermal diffusivity of thin films [3]. However, applicability of these models to short-pulse laser heating is questionable [1]. As is well-known, the Anisimov model [3] does not properly take into account the finite time for the nascent electrons to relax to the FD distribution. In the Anisimov model, the Fourier law for heat diffusion in the electron gas is assumed. However, the diffusion equation is valid only when the relaxation time is zero, $\tau = 0$, and the velocity of the thermalization is infinite $v \to \infty$.

In this paper, we use the HHC in which the relaxation time is finite and not equal to zero. For metals $\tau \leqslant 1$ ps. The heat propagation velocity is equal to the sound velocity in the electron gas and is of the order $\sim 1\ \mu\text{m} \cdot \text{ps}^{-1}$. In our study the HHC is applied to the investigation of heat transfer in low dimensional structures (LDS), i.e., the structures for which the characteristic dimension, l, is of the order of the mean free path for charge carriers (e.g., electrons). As it is shown in Ref. 2, for short delay times the heat is transferred in the form of heat waves which propagate in LDS with the sound velocity. In this paper it is shown that in LDS the nonequilibrium temperature pulses (temperature waves) generated by ultrafast (<1-ps) laser heating can be reflected from edges of LDS. Comparison of the calculated and observed in LDS temperature waves offers new possibility for the study of the dynamics of hot electrons in low-dimensional structures.

2. NONEQUILIBRIUM TRANSPORT OF HOT ELECTRON IN THIN METAL FILMS

The effects of ultrafast heat transport can be observed in the results of front-pump back-probe measurments [1]. The results of these type experiments can be summarized as follows. First, the measured delays are much shorter than would be expected if the heat were carried by the diffusion of electrons in equilibrium with the lattice (tens of picoseconds). This suggests that the heat is transported via the electron gas alone and that the electrons are out of equilibrium with the lattice on this time scale. Second, since the delay increases approximately linearly with the sample thickness, the heat transport velocity can be extracted $v_h \simeq 10^8\ \mathrm{cm \cdot s^{-1}} = 1\ \mu\mathrm{m \cdot ps^{-1}}$. This is of the same order of magnitude as the Fermi velocity of electrons in Au, $1.4\ \mu\mathrm{m \cdot ps^{-1}}$.

Since the heat moves at a velocity comparable to v_F, the Fermi velocity of the electron gas, it is natural to question exactly how the transport takes place. Since those electrons which lie close to the Fermi surface are the principal contributors to transport, the heat-carrying electrons move at v_F. In the limit of lengths longer than the momentum relaxation length, λ, the random walk behavior is averaged and the electron motion is subject to a diffusion equation. Conversely, on a length scale shorter than λ, the electron move ballistically with velocity close to v_F.

The importance of the ballistic motion may be appreciated by considering the different hot-electron scattering lengths reported in the literature. The electron–electron scattering length in Au, λ_{ee} has been calculated in Ref. 4. They find that $\lambda_{ee} \sim (E - E_F)^2$ for electrons close to the Fermi level. For 2-eV electrons $\lambda_{ee} \approx 35$ nm, increasing to 80 nm for 1 eV. The electron–phonon scattering length λ_{ep} is usually inferred from conductivity data. Using Drude relaxation times [5], λ_{ep} can be computed, $\lambda_{ep} \approx 42$ nm at 273 K. This is shorter than λ_{ee}, but of the same order of magnitude. Thus, we would expect that both electron–electron and electron–phonon scattering are important on this length scale. However, since conductivity experiments are steady-state measurements, the contribution of phonon scattering in a femtosecond regime experiment, such as pump-probe ultrafast lasers, is uncertain.

In the usual electron–phonon coupling model [3], one describes the metal as two coupled subsystems, one for electrons and one for phonons. Each subsystem is in local equilibrium so the electrons are characterized by a FD distribution at temperature T_e and the phonon distribution is characterized by a Bose–Einstein distribution at the lattice temperature T_l. The coupling between the two systems occurs via the electron–phonon interaction. The time evolution of the energies in the two subsystems is given by the coupled parabolic differential equations (Fourier law).

For ultrafast lasers the duration of pump pulse is of the order of relaxation time in metals [1]. In that case the parabolic heat conduction equation is not valid and a new equation for hyperbolic heat conduction must be used [2]:

$$\frac{1}{v_S^2}\frac{\partial^2 T}{\partial t^2}+\frac{1}{D_T}\frac{\partial T}{\partial t}=\nabla^2 T, \qquad D_T=\tau v_S^2 \tag{1}$$

In Eq. (1), v_S is the thermal wave speed, τ is the relaxation time, and D_T denotes the thermal diffusivity. In the following, Eq. (1) is used to describe the heat transfer in the thin gold films.

To that aim we define T_e is the electron gas temperature and T_l is the lattice temperature. The govering equations for nonstationary heat transfer are

$$\frac{\partial T_e}{\partial t}=D_T\nabla^2 T-\frac{D_T}{v_S^2}\frac{\partial^2 T_e}{\partial t^2}-G(T_e-T_l), \qquad \frac{\partial T_l}{\partial t}=G(T_e-T_l) \tag{2}$$

where D_T is the thermal diffusivity, T_e is the electron temperature, T_e is the lattice temperature, and G is the electron–phonon coupling constant. In the following, we assume that on a subpicosecond scale, the coupling between electron and lattice is weak [1] and Eq. (2) can be replaced by the following equations:

$$\frac{\partial T_e}{\partial t}=D_T\,\nabla^2 T-\frac{D_T}{v_S^2}\frac{\partial^2 T_e}{\partial t^2}, \qquad T_l=\text{constant} \tag{3}$$

Equation (3) describes nearly ballistic heat transport in a thin gold film irradiated by ultrafast ($\Delta t<1$ ps) laser beam.

The solution of Eq. (3) for 1D is given by

$$\begin{aligned}T(x,t)=\frac{1}{v_s}\int dx'\,T(x',0)\Bigg[&e^{-t/2\tau}\frac{1}{t_0}\,\Theta(t-t_0)\\&+e^{-t/2\tau}\frac{1}{2\tau}\left\{I_0\left(\frac{(t^2-t_0^2)^{1/2}}{2\tau}\right)\right.\\&\left.+\frac{t}{(t^2-t_0^2)^{1/2}}I_1\left(\frac{(t^2-t_0^2)^{1/2}}{2\tau}\right)\right\}\Theta(t-t_0)\Bigg]\end{aligned} \tag{4}$$

where v_s is the velocity of second sound, $t_0=(x-x')/v_s$, and I_0 and I_1 are modified Bessel functions and $\Theta(t-t_0)$ denotes the Heaviside function. We are concerned with the solution to Eq. (4) for a nearly delta function temperature pulse generated by laser irradiation of the metal surface. The pulse transferred to the surface has the shape:

$$\begin{aligned} \Delta T_0 &= \frac{\beta\rho_E}{C_V v_s \,\Delta t} && \text{for} \quad 0 \leqslant x < v_s\,\Delta t \\ \Delta T_0 &= 0 && \text{for} \quad x \geqslant v_s\,\Delta t \end{aligned} \tag{5}$$

In Eq. (5), ρ_E denotes the heating pulse fluence, β is the efficiency of the asborption of energy in the solid, $C_V(T_e)$ is the electronic heat capacity, and Δt is the duration of the pulse.

With $t = 0$ the temperature profile described by Eq. (5) yields

$$\begin{aligned} T(l, t) = {} & \frac{1}{2}\Delta T_0 e^{-t/2\tau}\Theta(t - t_0)\,\Theta(t_0 + \Delta t - t) \\ & + \frac{\Delta t}{4\tau}\Delta T_0\, e^{-\tau/2\tau}\left\{I_0(z) + \frac{t}{2\tau}\frac{1}{z}I_1(z)\right\}\Theta(t - t_0) \end{aligned} \tag{6}$$

where $z = (t^2 - t_0^2)^{1/2}/2\tau$ and $t = l/v_s$.

The solution to Eq. (3) when there are reflecting boundaries is the superposition of the temperature at l from the original temperature and from the image heat source at $\pm 2nl$. This solution is

$$\begin{aligned} T(l, t) = {} & \sum_{i=0}^{\infty} \Delta T_0 e^{-t/2\tau}\Theta(t - t_i)\,\Theta(t_i + \Delta t - t) \\ & + \Delta T_0 \frac{\Delta t}{2\tau} e^{-t/2\tau}\left\{I_0(z_i) + \frac{t}{2\tau}\frac{1}{z_i}I_1(z_i)\right\}\Theta(t - t_i) \end{aligned} \tag{7}$$

where $t_i = t_0$, $3t_0$, $5t_0$, $t_0 = l/v_0$.

For gold, $C_V(T_e) = C_e(T_e) = \gamma T_e$, $\gamma = 71.5\ \text{J}\cdot\text{m}^{-3}\cdot\text{K}^{-2}$, and Eq. (5) yields

$$\begin{aligned} \Delta T_0 &= \frac{1.4 \times 10^5\,\rho_E\beta}{v_s\,\Delta t\,T_e} && \text{for} \quad 0 \leqslant x < v_s\,\Delta t \\ \Delta T_0 &= 0 && \text{for} \quad x \geqslant v_s\,\Delta t \end{aligned} \tag{8}$$

where ρ_E is measured in $\text{mJ}\cdot\text{cm}^{-2}$, v_s in $\mu\text{m}\cdot\text{ps}^{-1}$, and Δt in ps. For $T_e = 300$ K

$$\begin{aligned} \Delta T_0 &= \frac{4.67 \times 10^2\,\beta\rho_E}{v_s\,\Delta t} && \text{for} \quad 0 \leqslant x \leqslant v_s\,\Delta t \\ \Delta T_0 &= 0 && \text{for} \quad x \geqslant v_s\,\Delta t \end{aligned} \tag{9}$$

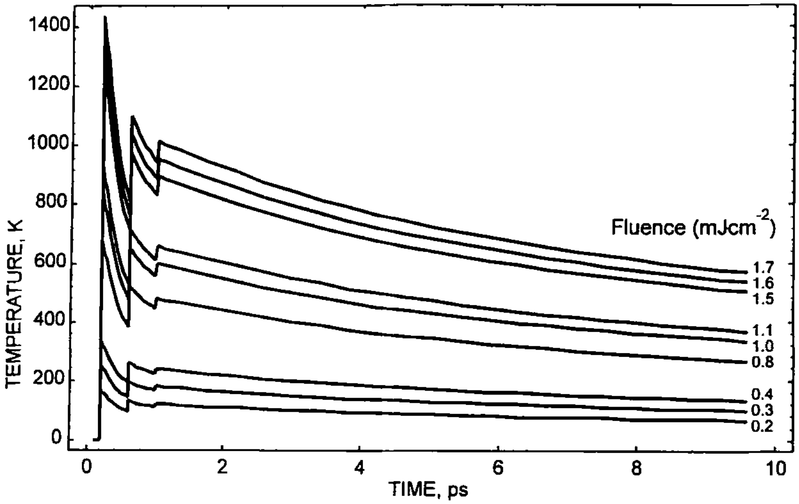

Fig. 1. Solutions of hyperbolic heat conduction (HHC) equation, Eq. (3).

In Fig. 1, the solution of Eq. (3) with the boundary condition given by Eq. (9) for $\rho_E = 0.2$, 0.3, 0.4, 0.8, 1, 1.1, 1.5, 1.6, and $1.7\ \mathrm{mJ \cdot cm^{-2}}$ is presented. The curves were calculated for $v_s = 0.15\ \mu\mathrm{m \cdot ps^{-1}}$ and $\tau = 0.12$ ps. As can be easily seen for $\rho_E > 0.2\ \mathrm{mJ \cdot cm^{-2}}$, the temperature shows the oscillations. Moreover, the amplitude of the oscillations grows with the fluence of the laser beam.

The solution of hyperbolic heat conduction equation, Eq. (7), contains two components:

$$T_1(l, t) = \sum_{i=0}^{\infty} \Delta T_0 e^{-t/2\tau} \Theta(t - t_i)\, \Theta(t_i + \Delta t - t) \tag{10}$$

$$T_2(l, t) = \sum_{i=0}^{\infty} \Delta T_0 \frac{\Delta t}{2\tau} e^{-t/2\tau} \left\{ I_0(z_i) + \frac{t}{2\tau} \frac{1}{z_i} I_1(z_i) \right\} \Theta(t - t_i) \tag{11}$$

The first component describes the thermal waves, which propagates with velocity v_s and can be reflected from the edges of the irradiated film. These thermal waves are damped due to e–e collisions. The second component, expressed by Eq. (11), is the diffusion component which for $t \to \infty$ tends to the solution of the parabolic diffusion equation (PHC; Fourier law).

$$\frac{1}{D_T} \frac{\partial T}{\partial t} = \nabla^2 T \tag{12}$$

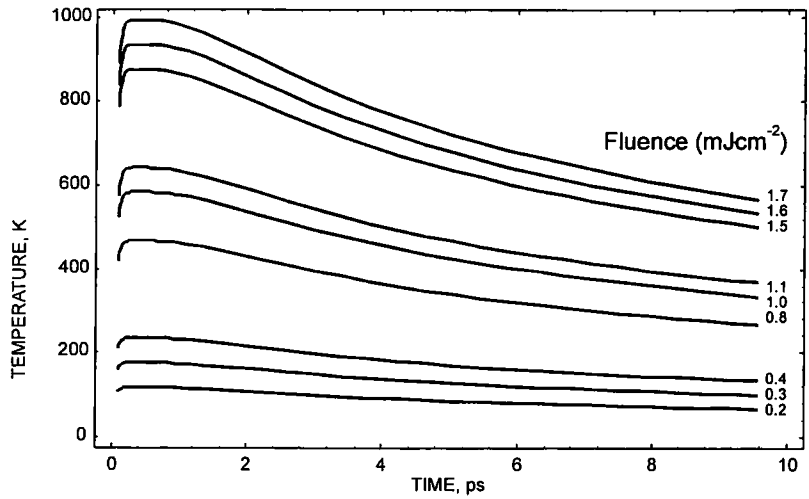

Fig. 2. Solution of parabolic heat conduction (PHC) equation, Eq. (12).

In Fig. 2, the solution of Eq. (12) is presented for $\rho_E = 0.2$, 0.3, 0.4, 0.8, 1, 1.1, 1.5, 1.6, and $1.7\ \mathrm{mJ \cdot cm^{-2}}$. The structure in temperature profile observed for $t < 2$ ps (Fig. 1) is the result of the reflection of the thermal waves from the edges of the gold film. The peaks in temperature profiles are observed for $t_1 = l/v_s$, $t_2 = 3l/v_s$, and $t_3 = 5l/v_s$. The temperature profiles

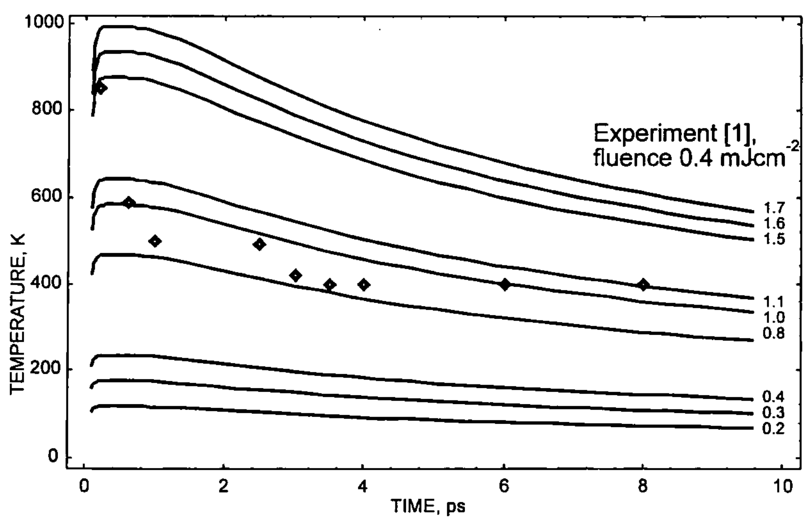

Fig. 3. Comparison of the solutions of PHC and experimental data for a 30-nm Au film [1].

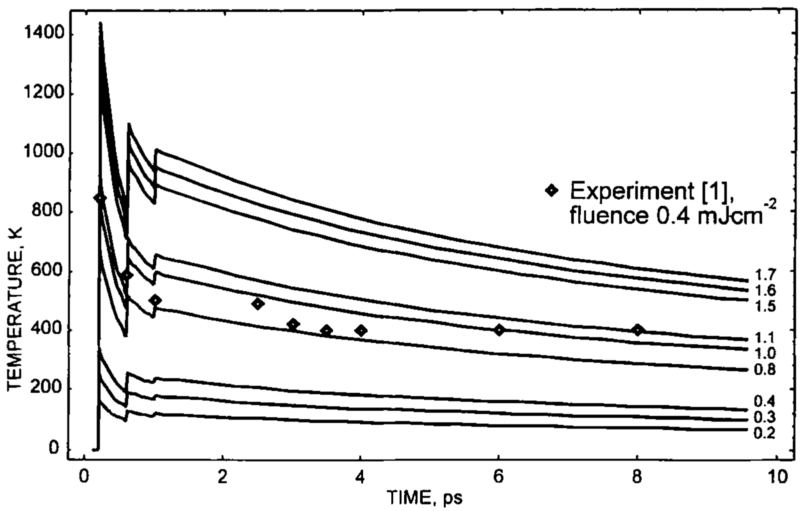

Fig. 4. Comparison of the solutions of HHC and experimental data for a 30-nm Au film [1].

obtained as the solution of parabolic equation (Fig. 2) do not show any oscillations for short times.

In Fig. 3, the comparison of the calculated parabolic temperature profiles and the measured one [1] is presented. With the help of the standard diffusion theory, the experimental data cannot be described with the one value of fluence. In order to obtain the fairly good description, one needs two value of fluence $\rho_E \sim 1.5$ and $\rho_E \sim 0.8\ \mathrm{mJ \cdot cm^{-2}}$.

In Fig. 4, the comparison of the calculated hyperbolic temperature profiles and the measured one [1] is presented. With the help the hyperbolic heat conduction equation, the experimental data can be satisfactorily described with the one value of fluence $\rho_E \sim 1\ \mathrm{mJ \cdot cm^{-2}}$. The fluence $1\ \mathrm{mJ \cdot cm^{-2}}$ for a 400-fs duration pump pulse give heat flux of the order $0.25 \times 10^{10}\mathrm{W \cdot cm^{-2}}$. Assuming, as in Ref. 1, that 15% of the light was absorbed, one obtains for heat flux the value $\sim 4 \times 10^8\mathrm{W \cdot cm^{-2}}$. Considering the above, we can conclude that for the heat fluxes of the order $10^8\mathrm{W \cdot cm^{-2}}$ the effect of hyperbolic heat conduction is significant.

3. CONCLUSION

We have shown that with the advent of lasers with a very short pulse duration, the effect of thermal wave propagation velocity becomes important. To consider this effect, hyperbolic heat conduction in thin gold films is studied. It was shown that for heat fluxes of the order $10^8\mathrm{W \cdot cm^{-2}}$ in thin gold film, the thermal waves are generated. The consideration of the hyperbolicity of heat transfer enables one to describe the temperature profile with one value of fluence.

ACKNOWLEDGMENT

This study was made possible by the financial support from the Polish Committee for Science Research under Grants 3 P408 004 05 and 7S 102 014 103.

REFERENCES

1. W. S. Fann, R. Storz, H. W. K. Tom, and J. Bokor, *Phys. Rev. Lett.* **68**:2834 (1992).
2. J. Marciak-Kozłowska, *Int. J. Thermophys.* **14**:593 (1993).
3. S. J. Anisimov, B. L. Kapeliovich, and T. L. Perelman, *Sov. Phys. JETP* **39**:375 (1975).
4. W. F. Królikowski and W. E. Spicer, *Phys. Rev. BI* 478 (1970).
5. N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rinehart and Winston, New York, 1976).