International Journal of Thermophysics, Vol. 14, No. 3, 1993

Wave Characteristic of Femtosecond Heat Conduction in Thin Films[1]

J. Marciak-Kozłowska[2]

The hyperbolic heat conduction equation (HHC) is solved for submicrometer gold film irradiated with a short-pulse laser. The transient temperature profiles are calculated. It is shown that the solutions of HHC and standard heat diffusion equation are significantly different for submicrometer films.

KEY WORDS: fermions; hyperbolic heat conduction; laser; transient temperature profile.

1. INTRODUCTION

New technologies based on fast laser heating are developing rapidly due to increasing availability of high-power, short-pulse lasers. The short-pulse energy deposition reduces heat-affected areas by minimizing heat diffusion and realizes ultrafast heating, melting and solidification. In addition, short-pulse laser heating is becoming an important tool in studying thermal properties of thin films.

Conventional laser heating processes, which involve a relatively low energy flux and long laser pulse, have been successfully modeled in metal processing and in measuring thermal diffusivity of thin films. However, the appropriateness of applying these models to short-pulse laser heating is subject to question [1]. In the present paper, we applied the hyperbolic heat conduction equation (HHC) to the heat propagation in thin metal films. The concept of HHC in a single-phase material dates back to Maxwell [2] and has since been derived by several approaches. In the

[1] Paper presented at the Third Workshop on Subsecond Thermophysics, September 17–18, 1992, Graz, Austria.

[2] Institute of Electron Technology, Al. Lotników 32/46, 02-668 Warsaw, Poland.

0195-928X/93/0500-0593$07.00/0

present paper, the solution of HHC is obtained for the one-dimensional problem. The calculated nonequilibrium temperature pulses propagate in the film as the thermal wave which is damped by collisions.

The propagation speed $v_s = \sqrt{\frac{1}{3}}\, v_F$ of the thermal wave is finite and gives rise to finite delay time of temperature pulse.

2. FORMULATION

The merit of the wave model, in contrast to the classical heat diffusion theory, lies in its unique way of describing a thermal signal propagating in solids with a finite wave speed. The finite wave speed has intrinsic influences on the mechanism of heat transfer. Mathematically, it renders an energy equation hyperbolic in nature. While the thermal wave speed resides in the wave term, the thermal diffusivity plays a role of damping in the thermal wave propagation. The weighted effect between the two is defined as the relaxation time [3, 4] in the wave theory. Under this frame, the classical diffusion theory is a special case of immediate response and the relaxation time is zero. The so-called hyperbolic theory of heat conduction describes the relaxation behavior in the history of thermal wave propagation. For both temperature and flux waves, a general feature is that a sharp wave front exists when penetrating through a solid medium. The thermal shock formation is a physical phenomenon pertinent to the wave theory [5].

According to the constitutive relation in the thermal wave model, heat flux, $\vec{q}$ obeys the relation [5, 6]:

$$\vec{q}(\vec{r}, t+\tau) = -k\nabla T(\vec{r}, t) \tag{1}$$

where τ is the relaxation time (a phase lag) and k is the thermal conductivity. The temperature gradient established in the material at time t results in a heat flux that occurred at a later time $t+\tau$ due to the insufficient time of response. For combining with the energy equation, however, all the physical quantities involved must correspond to the same instant of time. The Taylor's series expansion is thus applied to the heat flux $\vec{q}$ in Eq. (1) to give

$$\vec{q}(\vec{r}, t) + \frac{\partial \vec{q}(\vec{r}, t)}{\partial t}\tau + \frac{\partial^2 \vec{q}(\vec{r}, t)}{\partial t^2}\frac{\tau^2}{2} + \cdots = -k\nabla T(\vec{r}, t) \tag{2}$$

In the linearized thermal wave theory, the phase lag is assumed to be small and the higher-order terms in Eq. (2) are neglected. By retaining only the first-order term in τ, Eq. (2) becomes

$$\vec{q}(\vec{r}, t) + \tau \frac{\partial \vec{q}(\vec{r}, t)}{\partial t} = -k\nabla T(\vec{r}, t) \tag{3}$$

After combining Eq. (3) with the energy equation,

$$-\nabla \cdot \vec{q} + S = \rho C_p \frac{\partial T}{\partial t} \tag{4}$$

where S is the heat source per unit volume, ρ is the mass density, and C_p is the heat capacity, the result may have either a temperature T or a heat flux ($\vec{q}$) representation. The T representation is obtained by eliminating $\vec{q}$ from the two equations:

$$D_T \nabla^2 T + \left(\frac{1}{\rho C_p}\right)\left[S + \frac{D_T}{v_s^2}\frac{\partial S}{\partial t}\right] = \frac{D_T}{v_s^2}\frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} \tag{5}$$

For the medium without sources, one obtains instead of Eq. (5),

$$\frac{1}{v_s^2}\frac{\partial^2 T}{\partial t^2} + \frac{1}{D_T}\frac{\partial T}{\partial t} = \nabla^2 T, \qquad D_T = \tau v_s^2 \tag{6}$$

with τ being replaced by D_T/v_s^2 [3], where D_T is the thermal diffusivity and v_s the thermal wave speed. Equation (6) is the hyperbolic heat conduction equation (HHC).

3. NONEQUILIBRIUM HEAT TRANSPORT IN SUBPICOSECOND LASER-HEATED GOLD FILMS

The fact that the electronic heat capacity of metals is one to two orders of magnitude smaller than the lattice heat capacity has led to many investigations of nonequilibrium phenomena in metals with subpicosecond lasers. Model calculations suggest that it should be possible to heat the electron gas to temperature T_e of up to several thousand degrees for a few picoseconds while keeping the lattice temperature T_l relatively cold [7, 8].

Electromagnetic radiation with wavelength ranging from UV to near-IR interacts with metals through electron excitation and electron–phonon interactions. Photons excite electrons into higher energy levels and then the excited electrons thermalize rapidly, giving rise to a hot free electron gas. The high-temperature electron gas diffuses inside the metal and heats up the metal lattice through electron–photon collisions. This phenomenon has been observed experimentally by femtosecond photoemission [9]. Unresolved, however, is the role of heat transport in the dynamics of the hot electron–lattice system. This is because most experiments performed cannot distinguish between relaxation due to heat flow out of the probed region and electronic relaxation via photon emission. Only recently, the ultrafast heat transport in thin gold films under femtosecond laser irradiation was observed [10]. Time of flight measurements indicate that the heat

transit time scales linearly with the sample thickness and that the heat transport is very rapid, occurring at a velocity close to the Fermi velocity of electrons in gold. The experiment described in Ref. 10 may be good for testing ideas about heat wave propagation in this metal films. At present, the theory is based on the two temperature diffusion model of Anisimov et al. [8], even thought the relaxation times involved are well within the range where wave propagation could be dominant.

The objective of the subsequent investigation is to analyze the non-equilibrium heat transport in the Fermi gas of electrons in thin gold films. Guided by the results reported in the literature [10], we assume that in nonequilibrium electron gas heat is transferred as the thermal wave, i.e., the heat transport in thin films is described by HHC, Eq. (6).

First, we realized that for the Fermi electron gas, the thermal wave speed is defined as [11]

$$v_s = \left(\frac{p_F^2}{3mm^*} (1 + F_o^s) \right)^{1/2} \tag{7}$$

where m is the mass of free electron, m^* is the efective mass of interacting Fermions, and F_0^s is a dimensionless measure of the interaction strength in Fermi system. In the limit of weak interaction, $m^* \to m$, $F_o \to 0$, and $v_s \to \sqrt{\frac{1}{3}}\, v_F$. Assuming for Fermi velocity $v_F = 1.4 \times 10^8\ \mathrm{cm \cdot s^{-1}}$, one obtains $v_s = 0.8 \times 10^8\ \mathrm{cm \cdot s^{-1}}$. The calculated thermal wave speed (second sound speed) is of the same order of magnitude as the heat transport speed extracted in the literature [10].

In this paper, the short-pulse laser heating of a metal film is analyzed on the basis of a one-dimensional model, since the beam diameter is typically much larger than the heat diffusion penetration depth in a very short time. The temporal shape of a laser pulse is assumed as follows:

$$T(x, t) = \begin{cases} \Delta T_o & \text{for } 0 < x < v_s \Delta t \\ 0 & \text{for } x > v_s \Delta t \end{cases} \tag{8}$$

With the temperature profile described by Eq. (8), the solution of Eq. (6) reads

$$T(l, t) = \frac{1}{2} \Delta T_o \exp\left(-\frac{t}{2\tau} \right) \Theta(t - t_o)\, \Theta(t_o + \Delta t - t) + \frac{1}{4\tau} \Delta t\, \Delta T_o \exp\left(-\frac{t}{2\tau} \right) \left\{ I_o(z) + \frac{t}{2\tau} \frac{1}{z} I_1(z) \right\} \Theta(t - t_o) \tag{9}$$

where $z = (t^2 - t_o^2)^{1/2}/2\tau$ and $t_o = x/v_s$. In Eq. (9) I_o and I_1 are the modified Bessel functions, and Θ denotes the Heaviside function. The first term in

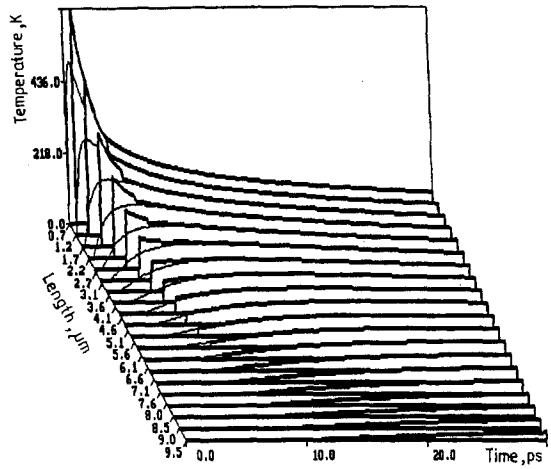

Fig. 1. Temperature pulses for gold film. Initial temperature $\Delta T_0 = 800$ K. Length is measured in micrometers and time in picoseconds. Heavy lines indicate the solution of the HHC equation; thin lines, the Fourier equation.

this solution corresponds to ballistic propagation of the temperature pulse damped by $\exp(-t/2\tau)$. The second term corresponds to the propagation of the energy scattered out of the ballistic pulse by diffusion. In the limit $\tau \to \infty$ the ballistic pulse alone arrives at the detector. In the limit $\tau \to 0$ the second term takes on asymptotic form which is solution to the conventional diffusion equation.

In Fig. 1 the calculated temperature profiles are plotted for $\tau = 0.6$ ps and $v_s = 10^8$ cm $\cdot$ s^{-1}. The input value of temperature, ΔT_0, is 800 K. The calculated curves represent the solution of the Fourier equation (parabolic heat diffusion equation; PHC) and the non-Fourier equation (HHC), Eq. (6). As can be seen in the thin films irradiated by a laser pulse, for a short time ($t \sim \tau$), the solutions of PHC and HHC are significantly different. For example, for a submicrometer ($l = 0.7$ μm) film, the temperature obtained from PHC is 440 K, while for the same film from HHC the temperature obtained is 655 K. For thick films, $l > 2$ μm, solutions of both equations, PHC and HHC, overlap.

4. CONCLUSIONS

In this paper, the solution of HHC for thin films is obtained. It is shown that for submicrometer films the solutions of standard heat diffusion

equation and HHC are significantly different. Considering that PHC is obtained when $v_s = \infty$ is assumed and, as shown in the paper, v_s is finite and $v_s \sim 10^8\ \mathrm{cm} \cdot \mathrm{s}^{-1}$, the applicability of PHC to the description of heat transport in this films is not based firmly. As discussed above, the HHC offers a plausible description of the thermal processes in the submicrometer films.

REFERENCES

1. D. E. Glass, M. N. Ozisik, and W. S. Kim, *Numer. Heat Transfer* A18:503 (1990).
2. J. C. Maxwell, *Philos. Trans. R. Soc. Lond.* **157**:49 (1967).
3. M. Chester, *Phys. Rev.* **131**:2013 (1963).
4. M. Chester, *Phys. Rev.* **145**:76 (1966).
5. D. Y. Tzou, in *Annual Review of Heat Transfer, Vol. IV*, L. T. Chang, ed. (Hemisphere, Washington, D.C, 1991), Chap. 3.
6. D. Y. Tzou, *ASME J. Heat Transfer* **114**:310 (1992).
7. M. Z. Kaganov, I. M. Lifshitz, and L. V. Tanatarov, *Sov. Phys. JETP* **4**:173 (1957).
8. S. I. Anisimov, B. L. Kapeliovich, and T. L. Perelman, *Sov. Phys. JETP* **39**:375 (1975).
9. J. G. Fujimoto, J. M. Liu, E. P. Ippen, and N. Bloembergen, *Phys. Rev. Lett.* **53**:1837 (1984).
10. S. D. Brorson, J. G. Fujimoto, and E. P. Ippen, *Phys. Rev. Lett.* **59**:1962 (1987).
11. D. Pines and P. Nozieres, *The Theory of Quantum Liquids* (Benjamin, New York, 1966), p. 23.