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# A Novel Method for Determination of the Thermal Diffusivity of Thin Films Using a Modulated $CO_2$ Laser[1]

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The thermal diffusivity of thin metal films has been measured by combining a fast infrared radiation thermometer with a mercury cadmium telluride (MCT) detector and a $CO_2$ laser modulated at a radio frequency up to 2 MHz. The laser output beam modulated by an acousto-optic modulator (AOM) is directed to the front surface of the blackened copper thin film (10 $\mu$m thick, 9.5 mm in diameter). The thermal radiation from the back surface of the sample is detected. From the observed phase delay in the detected signal of 0.68 radian to the input laser beam, the thermal diffusivity is determined to be $1.11 \times 10^{-4}\,\mathrm{m^2{\cdot}s^{-1}}$, which agrees well with the value of $0.99 \times 10^{-4}\,\mathrm{m^2{\cdot}s^{-1}}$ calculated from literature results. The method is generally applicable for measurements of thermal properties of nano/micro materials.

**KEY WORDS:** $CO_2$ laser; fast infrared radiation thermometer; mercury cadmium telluride (MCT) detector; periodic heating; thermal diffusivity; thin film.

## 1. INTRODUCTION

Recent rapid progress in the semiconductor industry and in materials science requires a measurement technique capable of detecting a fast temperature change in a very small area of the sample surface. A noncontact method is also important, so as to be free from contamination of

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the sample. The conventional method with a thermocouple or a resistance thermometer may not be applicable for this purpose. A high-speed thermo-reflectance method may be employed in the measurements [1]. However, this is applicable only for flat surfaces with specular reflection. Infrared radiation thermometry is a useful technique for the above measurement, that is, the detection technique of thermal radiation from the sample specimen irradiated by a laser flash or a periodically modulated laser beam can be used for rough sample surfaces. In this case, it is not necessary to polish the surface of the sample [2–6].

A mid-infrared radiation thermometer with laser flash excitation has been previously employed in our group [7]. The dynamic range, however, was limited from dc to 10 kHz. In the present research, we have developed a radiation thermometer which has a higher speed time response and higher spatial resolution. A radiation thermometer in the 10 $\mu$m range with a high-speed photovoltaic-type mercury cadmium telluride (MCT) detector has been developed. A wider dynamic range up to 2 MHz has been achieved. The thermometer is operated without a mechanical chopper, which is frequently used in infrared radiation thermometry. By combining this thermometer and high frequency modulated $CO_2$ laser, we have developed the proposed measurement system. This method allows us to measure the thermal properties of a variety of small thin specimens, which are transparent in the visible region and opaque in the infrared region such as glass, sapphire, and some semiconductors.

## 2. EXPERIMENTAL APPARATUS AND PROCEDURES

The present radiation measurement system can detect a fast temperature change in the radiating substance at the microsecond level. The required specifications of a radiation thermometer, which is applied for thin film thermal diffusivity measurements, are as follows: the temperature ranges from room temperature to over 500°C, the dynamic range is from dc to 1 MHz, the temperature resolution must be less than 0.1°C, and the spatial resolution on the sample surface is less than 1 mm. To achieve these specifications, we have employed a highly sensitive infrared detector, sophisticated infrared optics, and a radiation shield. The actually developed radiation thermometer has a dynamic range of 1 kHz to 2 MHz in dc-operation mode without a chopper, a temperature resolution of 50 mK, and a spatial resolution of 500 $\mu$m. The MCT detector, cooled with liquid nitrogen, has its peak sensitivity at 10 $\mu$m. The cryogenic MCT detector has better temperature resolution and time response than uncooled thermal detectors. Gold-coated parabolic mirrors are used as an imaging system of the thermometer. The focal lengths of the primary and secondary parabolic mirrors are 127 and 254 mm, respectively. The effective diameter of the mirror is 50 mm. The detector aperture diameter is 1 mm, and the detector is mounted in a vacuum Dewar. The radiation shield placed in front of the detector prevents background radiation from entering the detector. The radiation shield is made of aluminum, and the inside surface is treated with a diffusive low reflectance black coating (Hino-black). The temperature of the radiation shield is kept at 77 K. A high-speed operational amplifier with a feedback resistance of 1 MΩ has been developed. Detailed descriptions of similar concepts [8] of the design and construction of the radiation thermometer are presented. The radiation thermometer is shown in Fig. 1, which is calibrated against a blackbody from 0 to 900°C. The output signals from this radiation thermometer range from 0 to 3 V for blackbody temperatures from 0 to 900°C.

A block diagram of the periodic heating measurement system is shown in Fig. 2. The system is composed of two main parts, the pump (heating) part and the probe (detection) part. The pump part consists of

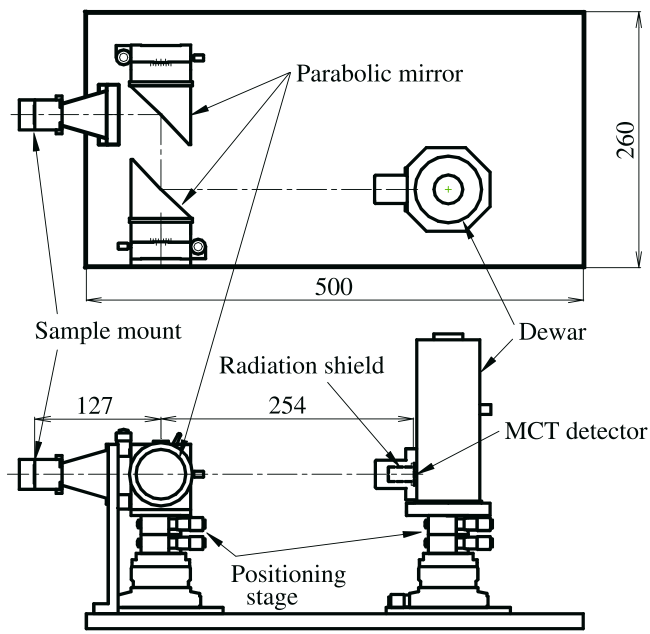


**Fig. 1.** Top and side views of the radiation thermometer. Sizes of components are shown in mm.

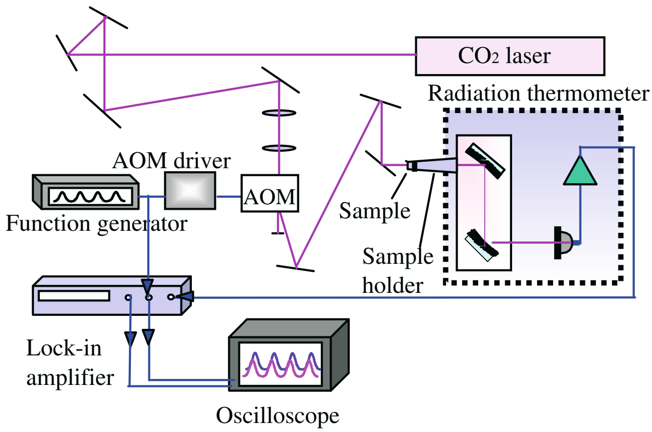


**Fig. 2.** Schematic diagram of experimental setup.

the high frequency modulated laser system, and the probe part consists of the fast radiation thermometer. The power and the frequency of the 10.6 μm cw $CO_2$ laser are stabilized by a feedback system with a piezoelectric transducer (PZT). The laser maximum output power is 4 W which is attenuated by using an aperture. The laser radiation is modulated by an acousto-optic modulator (AOM) over the frequency range from 1 kHz to 2 MHz. The function generator provides the modulation signal to the AOM, the reference signal to the lock-in amplifier, and the reference time scale on the oscilloscope. The time response of the infrared thermometer is evaluated by detecting the laser radiation modulated up to 2 MHz. The result at 1 MHz is shown in Fig. 3. The output signal from the thermometer responds well to the high frequency modulation from 1 kHz to 2 MHz. The time delays in the detection system and AOM are evaluated and are found to be negligible. The thermal diffusivity is obtained by measuring the phase delay of the signal through the sample. The signal without a sample gives the zero-delay signal on the cathode ray tube (CRT) display. The modulated laser light irradiates the front surface of the sample set on a taper-shaped sample mount. The components of the sample mount are shown in Fig. 4. The sample mount is made of aluminum and treated with a low reflectance black coating (Hino-black). The thin film is set on the sample holder and covered with an insulator ring. The sample holder with a screw cap is set on the sample mount. The radiation from the back surface of the sample is focused on the detector aperture of 1 mm diameter by two parabolic mirrors. The modulated laser beam irradiates a small area ($1.77 \times 10^{-6}$ m$^2$) of the sample.

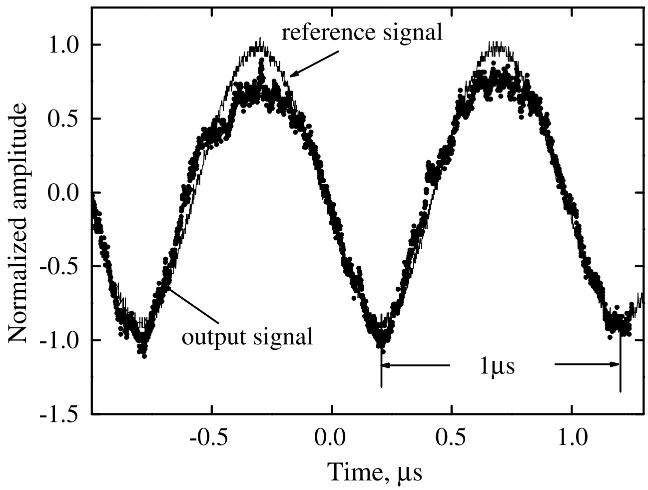


**Fig. 3.** Output signal from the radiation thermometer and the reference signal are showing good time response at a modulation frequency of 1 MHz.

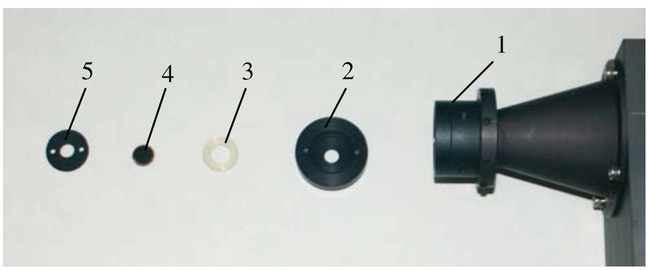


**Fig. 4.** Components of the sample mount. 1. sample mount; 2. sample holder, 3. insulator ring, 4. sample; 5. screw cap.

## 3. METHOD OF ANALYSIS

Since the sample thickness is much smaller than the sample diameter, the one-dimensional model of thermal conduction is employed (Fig. 5). The differential equation of thermal conduction is given by

$$\alpha\left(\frac{\partial^2 T(x,t)}{\partial x^2}\right)=\frac{\partial T(x,t)}{\partial t}, \tag{1}$$

where $T(x,t)$ is the temperature as a function of the distance $x$ from the sample surface and the time $t$, and $\alpha$ is the thermal diffusivity.

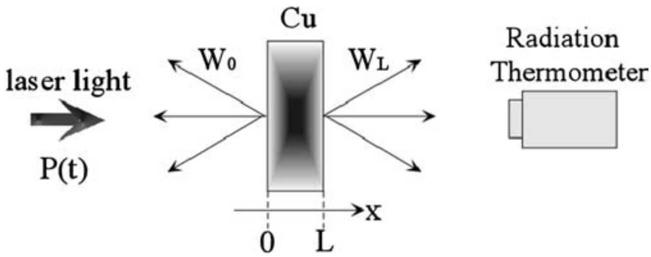


**Fig. 5.** One-dimensional model of thermal conduction. The thin film is irradiated by the modulated laser beam, ($P(t)$), and becomes an equilibrium by emitting thermal radiation ($W_0$, $W_L$) from the front and back surfaces. The thermometer detects $W_L$.

Equation (1) satisfies the boundary conditions at the front ($x=0$) and back ($x=L$) surfaces,

$$P = W_0 + \left(k\frac{\partial T(x,t)}{\partial x}\right)_0 \tag{2}$$

$$\left(k\frac{\partial T}{\partial x}\right)_L = W_L \tag{3}$$

where $P$ is the injected laser power, $W_0$ is the radiation power from the front surface of the specimen, $k$ is the thermal conductivity, $x$ is the distance from the front surface, and $W_L$ is the radiation power from the back surface of the specimen. $T(x,t)$ can be approximated by a sinusoidal function with a phase delay of $\delta_x$ to the phase of input laser light. It is given by

$$T(x,t) = A\sin(\omega t + \delta_x) \tag{4}$$

where $A$ is an arbitrary amplitude. At the back surface, $x=L$, the phase delay is given by [9]

$$\tan\delta_L = \frac{b(\tan B - \tanh B) + 2aB\tan B\tanh B + 2B^2(\tan B + \tanh B)}{b(\tan B + \tanh B) + 2aB - 2B^2(\tan B - \tanh B)} \tag{5}$$

where $B, a$, and $b$ are dimensionless parameters. These are given by

$$\begin{aligned} B &= L(\omega/2\alpha)^{1/2} \\ a &= L(c_L + c_0) \approx 2Lc_L \\ b &= L^2 c_L c_0 \approx L^2 c_L^2 = a^2/4 \end{aligned} \tag{6}$$

where

$$c_x = \left( \frac{\partial W}{\partial T} \Big/ k \right)_x \tag{7}$$

The value of the parameters, $a$ and $b$, are determined to be $a = 7.4 \times 10^{-5}$ and $b = 1.4 \times 10^{-9}$ at the present experimental conditions, $L = 10\,\mu$m, and $\omega = 2\pi \times 10^6$ Hz. Equation (5) can be approximated by

$$\tan \delta_L \approx -\frac{\tan B + \tanh B}{\tan B - \tanh B} \tag{8}$$

From the observed value of the phase delay $\delta_L$, the parameter $B$ and then the thermal diffusivity $\alpha$ can be obtained.

## 4. EXPERIMENTAL RESULTS AND DISCUSSION

The system is applied to a copper thin film with 10 μm thickness and 9.5 mm diameter. Both surfaces of this specimen are blackened with a carbon coating whose thickness is about 50 nm (Quick Carbon Coater: Sanyu-Denshi) and become highly emissive at 10.6 μm. The specimen is placed on a thermally insulated holder. Since the time constant of thermal diffusion of the present specimen is on the order of 1 μs, the modulation frequency is set to 1 MHz. We measured the modulated radiation signal from the back surface of the specimen and the modulated signal without the specimen. By least-squares fitting of the sinusoidal curve to the observed signal, we obtained a phase delay of 0.68 radian at 1 MHz. The experimental results are shown in Fig. 6. From Eq. (8) the thermal diffusivity $\alpha$ is determined to be $\alpha = 1.11 \times 10^{-4}\ \mathrm{m^2 \cdot s^{-1}}$ .

We calculate the literature value [10] from $\alpha = k/(C \cdot \rho)$ where $k$ is the thermal conductivity, $C$ is the heat capacity, and $\rho$ is the density for the dc part of the temperature $T$. $T$ is estimated by balancing the input laser power with the radiation power from both surfaces of the sample. It is given by

$$T^4 = \frac{P}{4\pi \varepsilon \sigma S} \tag{9}$$

where $P$ is the injected laser power, $\varepsilon$ is an emissivity of 1, $\sigma$ is the Stefan–Boltzmann constant, and $S$ is the heating area. The temperature $T = 480°\mathrm{C}$ is calculated by using a laser power of 400 mW and a heating area of $1.77 \times 10^{-6}\ \mathrm{m^2}$. The result is $\alpha = 0.99 \times 10^{-4}\ \mathrm{m^2 \cdot s^{-1}}$ at 480°C which agrees well with the experimental value from this study of $\alpha = 1.11 \times 10^{-4} \mathrm{m^2 \cdot s^{-1}}$.

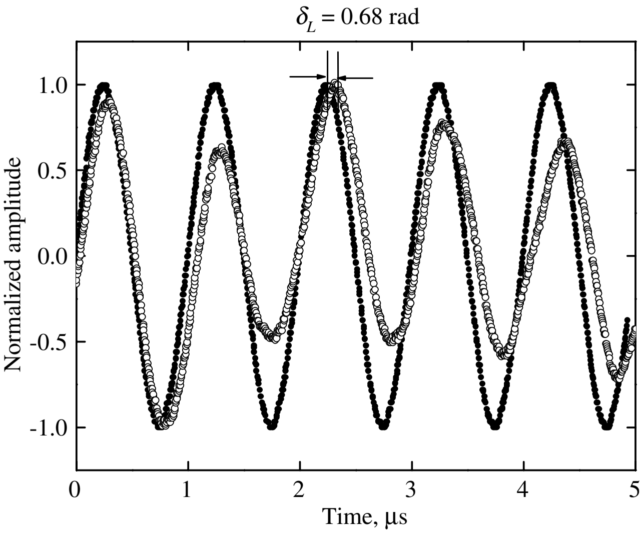


**Fig. 6.** Sinusoidal signals of the thermal radiation and the laser beam. Open circles show the signal from the back surface of the copper metal film and solid circles show the reference signal of input laser beam. A phase delay of 0.68 radian is observed.

We have confirmed the validity of this measurement system by obtaining a reliable value of the thermal diffusivity of a thin film. The uncertainty of this method depends on the estimation of the sample temperature, because there is difficulty in the measurement of the heating area. The thermal diffusivity is weakly dependent on the temperature in the present temperature range.

## 5. CONCLUSION

We have developed a novel method for determination of the thermal properties of thin films using a fast radiation thermometer and a modulated $CO_2$ laser beam. The characteristic feature of this method is that the phase delay in Eq. (5) changes sensitively as a function of $B$, which is inversely proportional to the square root of the thermal diffusivity. Therefore, we can determine the thermal diffusivity with high accuracy. We need to accurately measure only the phase delay in the case of the periodic heating method.

The novel point of this system is that we are using an infrared laser as an excitation source. Therefore, we can investigate materials such as glass, sapphire, and some semiconductors, which have absorption bands at 10 $\mu$m but are transparent in the visible region. The blackening, which may introduce uncertainty in the measurement, is not necessary if the specimen is opaque in the infrared region.

To improve and extend the present experimental system, we are trying to reduce the sample temperature by attenuating the dc part of the heating and by attaching a radiator to the sample holder.

We are also going to measure the thermal properties of a layer sample deposited on glass substrates. The substrates absorb infrared laser radiation and prevent deterioration of the layer sample. This measurement system can be applied to any sample deposited on glass substrates.

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