Study on thermal diffusivity of materials by laser photothermal reflection technique

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A method of measuring thermal diffusivity of materials at room temperature by photothermal reflection technique is described. An intensity-modulated Ar^+ laser beam is used as incident light. The beam is focused to about 1 mm diameter spot and illuminates the sample surface. HgCdTe infrared detector is used to receive photothermal signal. Using this technique, the photothermal signals are experimentally measured as the function of different frequencies. The thermal diffusivities can be obtained by fitting the experimental data. On the other hand, the thermal diffusivities of one-way composite and orthogonal symmetric arranged composites Al_2O_3/Al are measured in transverse, longitudinal and arbitrary directions. The results show that the diffusivity of one-way material decreases with the increase of the measurement angle; the diffusivity of orthogonally arranged material almost keeps the same when measurement angle changes.

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Laser photothermal technique has been recently used to measure material properties and analyze material defects[1,2]. And photothermal transmission technique is used in these applications. In this technique, the light source and the detector are laid bilaterally to the sample, therefore, the photothermal signal of the sample is weak when the sample is thick. For this reason, this technique is only suitable for thin sample[3,4]. However, in photothermal reflection technique, the light source and the detector are laid in the same side of the sample, consequently there is no special requirement to the sample thickness. In this letter, the principle and technique of measuring diffusivity by photothermal reflection technique are studied, the thermal diffusivities of composite Al_2O_3/Al are measured and the experimental results are discussed and analyzed.

A laser beam modulated by frequency f is introduced to the sample. The sample absorbs laser energy and produces a periodic temperature fluctuation and infrared radiation. Using appropriate infrared detector, photothermal signal can be detected. For single-layer sample, the incident beam intensity can be written as

$$I = I_0[1 + \exp(i\omega t)], \tag{1}$$

where I_0 is the amplitude of the incident beam, $\omega = 2\pi f$ is the angular frequency. According to Stefan-Boltzmann radiation law, the change of the heat radiation from the sample is

$$\delta W = 4\varepsilon\sigma T^3 \delta T, \tag{2}$$

where σ is Stefan-Boltmann constant, ε is the emissivity of the sample, T is the temperature on the sample surface, W is the radiation intensity.

Photothermal signal detected by the detector is proportional to the change of δW. Under the condition of strong absorption, the amplitude and phase of photothermal signal are given by using model of one-dimensional thermal conduction equation[5]

$$A = \frac{\sqrt{2}I_0}{2kx}\left[\frac{\exp(2lx) + \exp(-2lx) + \cos(2lx)}{\exp(2lx) + \exp(-2lx) - \cos(2lx)}\right]^{\frac{1}{2}}, \tag{3}$$

$$\begin{aligned}\Phi = {} & \arctan\left[\frac{\exp(lx) - \exp(-lx)}{\exp(lx) + \exp(-lx)}\tan(lx)\right] \\ & - \arctan\left[\frac{\exp(lx) + \exp(-lx)}{\exp(lx) - \exp(-lx)}\tan(lx)\right] - \frac{\pi}{4},\end{aligned} \tag{4}$$

where k is the thermal conductivity of the sample, $x = \sqrt{\frac{\pi f}{\lambda}}$ (λ is the thermal diffusivity of the sample), l is the thickness of the sample.

The amplitude and phase of photothermal signal can be detected at different frequency. According to Eqs. (3) and (4), the thermal diffusivity of the material can be obtained by nonlinear fitting.

The laser photothermal reflection measuring system is schematically shown in Fig. 1. An Ar^+ laser with power up to 1.5 W provides the laser beam. The beam modulated by the chopper (PAR192) is directed onto the sample surface. The radiation emitted by the sample surface is collected and focused onto the HgCdTe detector using two concave spherical mirrors (CSM). The detector is placed in the focal plane of one of the CSM, and the heated area on the sample surface is centred at the focal point of the other mirror. An argon-coated Ge window is mounted in front of the detector to block the visible radiation from the laser. A preamplifier amplifies the thermal signal before being sent to the lock-in-amplifier (PAR 5204 model). Finally, the data are processed by computer. In order to ensure the sample to be of single-layer, it is mounted on a hollow platform that is fixed on a stepping motor, which is used for moving the platform. The stepping motor is controlled by the computer.

The sample is Al_2O_3/Al composite. The matrix is aluminium. There are the same two layers of Al_2O_3 fiber. The density of the fiber is 3.2 g/cm^3, its volume fraction

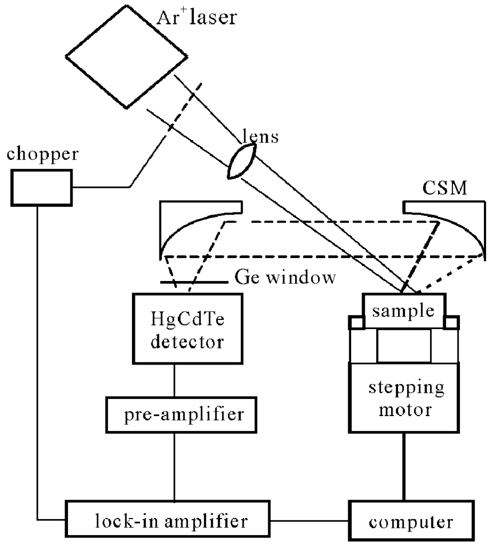

Fig. 1. Schematic of the laser photothermal reflection technique for measuring the diffusivity.

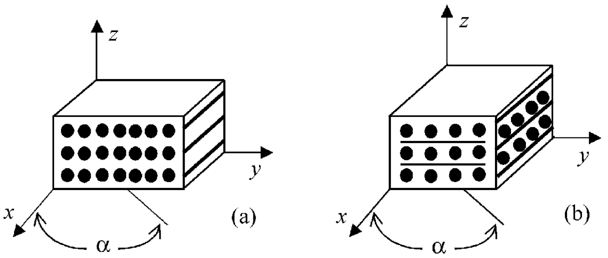

Fig. 2. Structure diagram of sample arrangement. (a) Fiber one-way arrangement; (b) fiber orthogonal symmetric arrangement.

to Al_2O_3/Al composite is 35%. The Al_2O_3/Al materials are made by hot-pressing method. The fiber is arranged in two form: single direction and orthogonal symmetric. The sizes of the sample are $15 \times 10 \times 1.08$ mm^3 and $15 \times 10 \times 1.16$ mm^3. The surfaces of the sample are polished.

When operating the stepping-motor the incident laser beam scans the sample along a straight line. Five points, which separate by 1 mm, are measured in each direction. For each point the amplitude and the phase are measured at different modulated frequency. The photothermal signal is the average value of three sampling. Using Eqs. (3) and (4), the thermal diffusivity at each point can be obtained by nonlinear fitting. The thermal diffusivity along a line can be obtained by calculating the average value of the five points. Meanwhile, the thermal diffusivities of Al_2O_3 fiber are measured at arbitrary direction. Figure 3 is the fitting curve of the amplitude and the phase at coordinate point (8, 5) mm in x-y plane of Fig. 2 for orthogonal symmetric material. The experimental results for Al_2O_3/Al composite and Al_2O_3 fiber are given in Table 1.

The experimental result shows that the thermal diffusivity of single direction arranged Al_2O_3/Al composite decreases with increasing angle α. On the contrary, the thermal diffusivity of orthogonal symmetric arranged Al_2O_3/Al composite is almost unchanged. For single direction material, the boundary area of phonon and electron increases with the increase of α. This increases the moving resistance of the phonon and electron, consequently, the thermal conductivity of the composite along the direction with α.

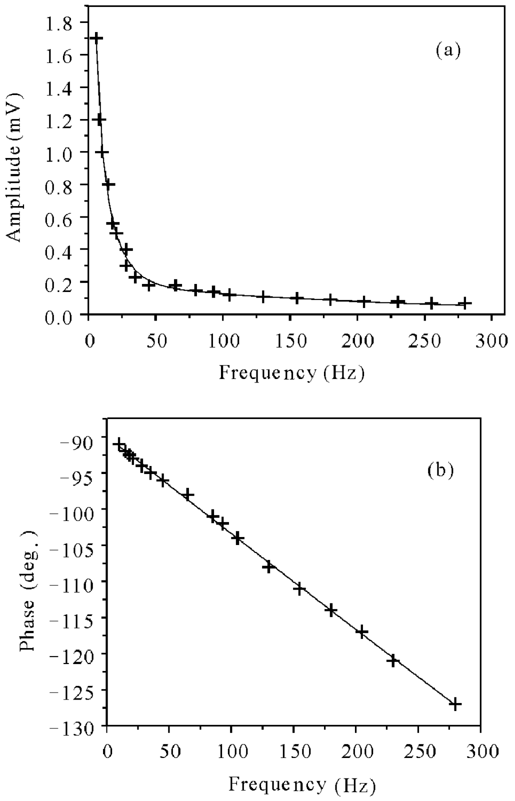

Fig. 3. Fitting curves of amplitude and phase with frequency for orthogonal symmetric material at (8, 5) mm point for fiber direction. (a) Amplitude; (b) phase.

For orthogonal symmetric arranged material, the volumes of the fiber along x and y are the same, $V_{fx} = V_{fy} = V_f/2$, the matrix volumes are equal too, $V_{mx} = V_{my} = V_m$. Assuming the thermal conductivities of the composite along α direction are $k_{x\alpha}$, $k_{y\alpha}$, where $k_{x\alpha}$ is made of the x direction fiber and $k_{y\alpha}$ is made of the y direction fiber. The thermal conductivity of the whole composite along α $k_\alpha$can be written as

$$k_\alpha = k_{x\alpha} + k_{y\alpha}. \qquad (5)$$

According to Ref. [6],

$$k_\alpha = k_0 - (\sin\alpha)^2 \left[V_f(1 - V_f)(k_m - k_f)^2\right]/(2k_0), \qquad (6)$$

where $k_0 = V_m k_m + V_f k_f$, k_α is the thermal conductivity of the composite in along α direction, k_m is the thermal conductivity of the matrix, k_f is the thermal conductivity of the fiber, V_f is the volume fraction of the fiber, V_m is the volume fraction of the matrix. According to Eq. (6), $k_{x\alpha}$, $k_{y\alpha}$ can be expressed as

$$\begin{aligned} k_{x\alpha} &= k_0 - (\sin\alpha)^2 \\ &\quad \cdot \left[V_{fx}(1 - V_{fx})(k_m - k_f)^2\right]/(2k_0), \qquad (7) \\ k_{y\alpha} &= k_0 - [\sin(90 - \alpha)]^2 \\ &\quad \cdot \left[V_{fy}(1 - V_{fy})(k_m - k_f)^2\right]/(2k_0). \qquad (8) \end{aligned}$$

Table 1. Experiment Results for Al_2O_3/Al Composites

Thickness (mm)	Fiber Arrange Model	Test Direction α (deg.)	Fitting* Model	Diffusivity (cm^2s^{-1})	Fitting Errors (Q%)	Diffusivity of Fiber (cm^2s^{-1})
1.08	One-Way	0	AF	0.54	3	0.23
			PF	0.53	2	
		30	AF	0.42	3	0.18
			PF	0.41	3	
		45	AF	0.31	3	0.15
			PF	0.32	3	
		60	AF	0.20	3	0.11
			PF	0.21	3	
		90	AF	0.11	2	0.06
			PF	0.09	2	
1.16	Orthogonal Symmetric	0	AF	0.45	3	
			PF	0.46	3	
		30	AF	0.47	3	
			PF	0.46	3	
		45	AF	0.47	2	
			PF	0.45	3	
		60	AF	0.46	2	
			PF	0.47	2	
		90	AF	0.47	3	
			PF	0.45	3	

*Fitting model: AF is amplitude fitting, PF is phase fitting.

Using Eqs. (5), (7) and (8), we get

$$k_\alpha = 2k_0 - \left[V_f(1 - V_f/2)(k_m - k_f)^2\right]/(4k_0). \qquad (9)$$

In Eq. (9) the thermal conductivity is not related to angle α, therefore, the thermal diffusivity of orthogonal symmetric is equal in any material direction.

We measure the thermal diffusivity of composite by using laser photothermal reflection technique. The amplitude and the phase of the photothermal signal can be detected simultaneously, and the thermal diffusivities are obtained by nonlinear fitting of photothermal signal to frequency. Accordingly, the results are more dependable and accurate than other methods. The thermal diffusivity single direction arranged Al_2O_3/Al material obviously changes with direction at the x-y plane and thermal diffusivity of orthogonal symmetric arranged Al_2O_3/Al material keeps unchanged.

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References

1. A. Salazar, A. Sanchez-Lavega, and J. Fernadez, J. Appl. Phys. **69,** 1216 (1991).
2. A. C. Boccara, D. Fournier, and J. Badoz, Appl. Phys. Lett. **36,** 130 (1980).
3. P. Z. Li and G. Y. Zhou, Appl. Opt. **31,** 3781 (1992).
4. P. Z. Li and Q. H. Wang, Chin. J. Scientific Instrument (in Chinese) **15,** 265 (1994).
5. G. X. Guan, X. M. Zheng, and P. Z. Li, Chin. J. Infrared Res. (in Chinese) **7A,** 201 (1988).
6. P. G. Klemens, International J. Thermophysics **11,** 971 (1990).