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Measurement of Thermal Diffusivity at Low Temperatures Using an Optical Reflectivity Technique[1]

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An experimental arrangement has been developed for measuring the transient temperature responses and the thermal diffusivities of foil materials in the range of 10 to 300 K by using the optical reflectivity technique. The cryogenic system with optical windows is designed to provide temperatures from 10 to 300 K. The front surface of a foil specimen is heated by a pulsed Nd:YAG laser. *In situ* measurement of the reflectivity of a continuous-wave He–Ne laser at the rear surface is conducted on the microsecond time scale. Using the temperature dependence of reflectivity, the transient temperature response is deduced. The thermal diffusivity is obtained by fitting Parker's formulae to the experimental data on temperature rise. Stainless-steel foils are chosen as samples and are studied in the region from 10 to 300 K. The accuracy is examined by comparing the present results with the theoretical temperature responses and thermal diffusivity data from the literature. Good agreement is observed.

KEY WORDS: low temperature; optical reflectivity technique; stainless-steel foil; thermal diffusivity; transient temperature response.

1. INTRODUCTION

The extensive applications of cryogenic technologies to technical and industrial areas, such as microelectronic device cooling and cryogenic vacuum systems, require thermophysical properties at low temperatures for successful design of facilities and production of materials. The transient temperature measurement technique at low temperatures can provide not only a method for measuring thermal diffusivity [1] but also an opportunity for observing

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some new phenomena in heat conduction [2, 3]. In the low-temperature region, however, since the conventional infrared radiation technique is no longer valid, a new measuring technique is needed.

A change in temperature affects the complex refractive index [4], which in turn influences the optical reflectivity of the material. Accordingly, the time-resolved reflectivity measurement will reveal the transient temperature response. The use of the optical reflectivity technique offers a possibility for measuring the high-speed temperature response in the temperature region exceeding the waveband limits of infrared detectors, for example, at low temperatures. There are two techniques, in general, for time-resolved reflectivity measurements: the modulated heating–pulse probing technique [5–7] and the nonmodulated heating–continuous-wave probing technique [8–11]. The first method can overcome the response limits of photodetectors and can measure small signals of temperature changes using a modulation technique but cannot be used to measure a temperature profile with a slowly decreasing speed. The second method can be used for a temperature profile of any shape but has difficulty measuring a temperature change faster than the response of the photoelectronic sensor. The pulse probing technique has been used successfully to measure the transient reflectivity changes caused by electron temperature changes in metals irradiated with ultrashort laser pulses [6, 11]. The continuous-wave probing technique has been used for transient temperature measurements of semiconductors in the range above room temperature. Regarding the measurement of the transient temperature response and the thermal diffusivity at low temperatures using this technique, however, few studies are found in the literature.

This work develops an experimental arrangement for measuring the transient temperature response in the temperature range 10 to 300 K using the continuous-wave probing technique. The front surface of a foil specimen is heated by laser-pulse irradiation. *In situ* measurement of the reflectance signal of a continuous-wave laser at the rear surface is conducted and used to deduce the temperature response on the microsecond time scale. The thermal diffusivity is obtained by using the measured temperature rise. Stainless-steel foils are used as samples. The accuracy of the method is examined by comparing the present results with theoretical temperature responses and thermal diffusivity data from the literature.

2. EXPERIMENTAL

2.1. Principle of Measurement

Transient temperature measurement by the optical reflectivity technique is based on the variation of the material complex refractive index [4],

$N = n(T, \lambda) + ik(T, \lambda)$, with temperature. The complex refractive index is related to the reflectivity by the Fresnel formula [12], which is given by

$$R(T, \lambda) = \frac{(n(T, \lambda) - 1)^2 + k^2(T, \lambda)}{(n(T, \lambda) + 1)^2 + k^2(T, \lambda)} \tag{1}$$

at normal incidence of light, where λ is the wavelength of incident light and T is the temperature of the surface. For incident light at a fixed wavelength, the reflectivity changes simply with temperature. If the reflectivity–temperature relation is known, the time–resolved reflectivity measurement will reveal the transient temperature response. For a large temperature change, the temperature dependence of the reflectivity of the sample is basically necessary for deducing the temperature from the reflectivity. This is usually determined by ellipsometric measurements [13] or quantum mechanical calculations with the band structure [14]. Some results in the literature show that, for metals [14], semiconductors [15], and superconductors [16], the reflectivity is approximately linearly proportional to the temperature.

For a small temperature change and a fixed wavelength, the reflectivity change as a function of the temperature on the surface of the sample can be written in the form of a Taylor series expansion as

$$\begin{aligned} \Delta R(T, \lambda) &= R(T_0 + \Delta T, \lambda) - R(T_0, \lambda) \\ &= R'(T_0, \lambda)\, \Delta T + \tfrac{1}{2} R''(T_0, \lambda)(\Delta T)^2 + \cdots \approx R'(T_0, \lambda)\, \Delta T \end{aligned} \tag{2}$$

where T_0 is the initial temperature and ΔT is the temperature change. According to Eq. (2), when the *in situ* reflectivity measurement is used to obtain only the temperature change history, without the need to know the temperature values, the temperature–time curve can be deduced directly from the normalized reflectivity–time curve.

2.2. Experimental Setup

The experimental setup for *in situ* optical reflectivity measurement at low temperatures is shown in Fig. 1. The cryostat with optical windows is designed to provide temperatures from 10 to 300 K. The temperature at the sample holder is set by a microprocessor-based digital temperature indicator/controller with a silicon diode sensor. A pulsed Nd:YAG laser with a wavelength of 1064 nm is used as the heating source. The pulse duration of the laser beam is measured to be 17 ns, using a silicon PIN photodiode with rise and fall times of 0.2 ns and a digitizing oscilloscope with a 500-MHz sampling speed. The output energy of a single pulse is

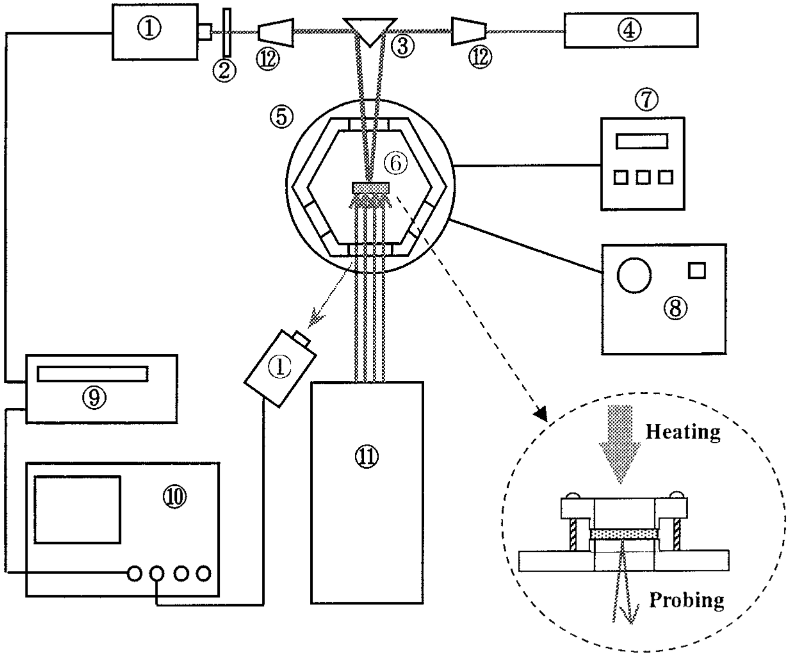

Fig. 1. Experimental setup for transient optical reflectivity measurement during pulse-laser heating at low temperatures. (1) Photodetector; (2) optical filter; (3) prism; (4) He–Ne laser; (5) cryostat; (6) sample; (7) temperature controller; (8) compressor; (9) preamplifier; (10) oscilloscope; (11) Nd:YAG laser; (12) beam expander.

measured in the range of 250 to 300 mJ by a power meter. A laser beam spot with a diameter of 8 mm can cover the surface of the sample through the front optical window on the cryostat. In this situation, a temperature rise of less than 2 K in the sample is measured by a thermocouple.

A continuous-wave He–Ne laser is employed as the probing light source for the reflectance measurement. Because previous work [9] did not show an obvious advantage of p-polarized light compared with unpolarized light, the unpolarized laser is used here. The power of the probing laser is 2 mW, which could be weak enough that the temperature rise caused by it can be ignored. The He–Ne laser beam is expanded to 1.8 mm by a beam expander and led to the center of the rear surface of the sample by a prism and the rear optical window on the cryostat. The angle of incidence is approximately 0°. The normal incidence and the expansion of the probing laser beam can decrease the error caused by thermoelastic displacement of the sample. The reflected light is collected by a beam expander set in the opposite direction, passes through a narrow-band interference filter, and then goes into a fast silicon PIN photodiode. A filter is used to avoid sensing lights not from the probing laser, such as the heating laser, thermal radiation from the sample, and other light sources. Since the change rate of the reflectivity with temperature is very low, the photoelectronic circuit is designed to output only signals indicating changes in reflectivity. The output signals are amplified by a low-noise preamplifier, with a gain of 1 to 10,000 and a frequency band of dc–1 MHz, and recorded on a SONY Tektronix TDS520 two-channel digital storage oscilloscope with a highest sampling speed of $1\ \mathrm{GS} \cdot \mathrm{s}^{-1}$. A silicon PIN photodiode is placed at the front of the optical window for the incidence of heating laser, which gives a trigger signal to determine the initial point of the reflectance response.

3. RESULTS AND DISCUSSION

3.1. Temperature Responses

An SUS304 (Cr, 18–20%; Ni, 8–11%; Mn, $<2\%$; Si, $<1\%$; Fe, balance) stainless-steel foil with a thickness of 90 μm is taken as the sample. The front surface is heated by a Nd:YAG laser pulse, and the transient optical reflectivity change at the rear surface is probed by a continuous-wave He–Ne laser. According to the measuring principle [Eq. (2)], because the temperature rise is small, the normalized temperature response can be deduced directly from the normalized transient reflectivity change.

Figure 2 shows the normalized transient reflectivity changes, as the normalized temperature responses, measured at 10, 50, 150, 250, and 290 K. At low temperatures, a larger fluctuation noise can be observed in the response curves, which is caused mainly by the thermoelastic displacement but not electric noise. This was confirmed by changing the holder and the thickness of the sample. With a decrease in temperature, the reflectivity change rate may become low and the fixture of the sample may become loose, resulting in an increase in the fluctuation noise. This noise is decreased by expanding the probing beam and improving the fixture of the sample, but it is still the main error source in the measurements.

In Fig. 2, the theoretical fitting curves, which are obtained using Parker's formula of normalized temperature response [1],

$$\Delta T(t) = 1 + 2 \sum_{m=1}^{\infty} (-1)^m \exp(-m^2 \pi^2 a t / L^2) \tag{3}$$

are also plotted for comparison, where a, t, and L are the thermal diffusivity, time, and thickness, respectively. The good agreement between the experimental results and the theoretical calculations shows that the transient

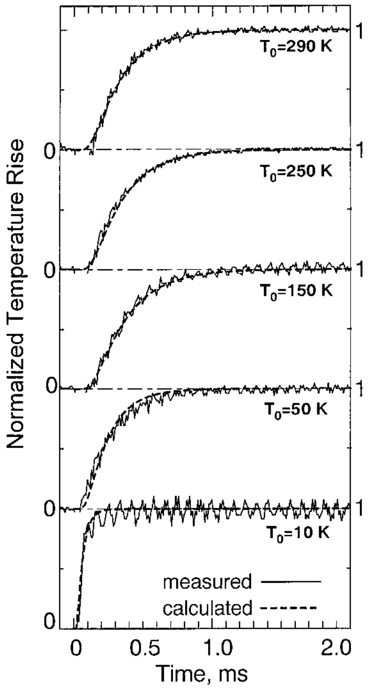

Fig. 2. Normalized transient optical reflectivity changes, as normalized temperature responses, of 90-μm SUS304 stainless-steel foils under laser-pulse heating at various temperatures.

reflectivity changes reveal the temperature responses accurately. By comparing the temperature responses at different temperatures, it can be seen that a decrease in the initial temperature increases the rise speed of the temperature at the rear surface. This is because of the increase in thermal diffusivity with decreasing temperature.

3.2. Thermal Diffusivities

Utilizing Eq. (3), thermal diffusivities can be estimated by fitting the measured temperature responses. From the estimated temperature data and the measurement results, the standard errors of the measurement are obtained as 0.011 (10 K) and 0.059 (290 K). Beside the main error source (thermal vibration), the temperature dependence of the thermal diffusivity may become another error source at low temperatures. Figure 3 shows the

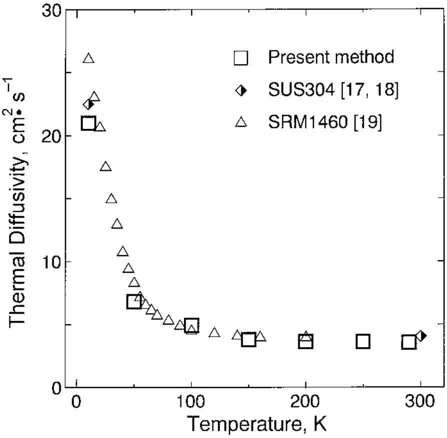

Fig. 3. Thermal diffusivities of SUS304 stainless-steel foils in the temperature range from 10 to 290 K obtained from transient reflectivity measurements of temperature responses.

thermal diffusivities of SUS304 stainless-steel foils in the temperature range from 10 to 300 K. Each plotted result, which is an average value obtained from four measurements, has a relative deviation below 5%. According to the results, the thermal diffusivity increases slightly with decreasing temperature from 300 to 100 K. At low temperatures (below 50 K), a steep increase in thermal diffusivity with decreasing temperature is observed.

Some literature results are plotted in Fig. 3 for comparison. Unfortunately only two reference data on thermal diffusivities, at 300 K [17] and 10 K [18], could be found for SUS304 stainless steel. The thermal diffusivities of SMR1460 stainless steel are also plotted as reference data [19]. As shown in Fig. 3, the present results agree well with the literature data at all temperatures.

4. CONCLUDING REMARKS

The transient temperature responses of SUS304 stainless-steel foils under pulsed laser heating have been measured in the temperature range 10 to 300 K using the *in situ* optical reflectivity technique. The thermal diffusivities were obtained by fitting Parker's formula with the measured temperature rise. The good agreement between the experimental results and literature data demonstrates that the present method is reliable for measuring thermal diffusivity at low temperatures. Extending the time scale of measurement from the present microsecond to the nanosecond range may provide an opportunity for observing some new phenomena in heat conduction.

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