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Development of a low-temperature laser interferometric dilatometer using a cryogenic refrigerator

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Abstract. A low-temperature laser interferometric dilatometer using a liquid-helium-free cryogenic refrigerator was developed for the absolute measurement of a linear thermal expansion coefficient with high accuracy. To evaluate the performance of the laser interferometric dilatometer, measure-ments were carried out on a high-purity single crystal of silicon over the temperature range 10 – 320 K. The measured value of the expansion coefficient was also compared with the recommended value by CODATA. The present result was in good agreement with the recommended value below room temperature, and the deviation from the recommended value was less than $\pm 0.01 \times 10^{-6}$ K^{-1}.

1 Introduction

The linear thermal expansion coefficient, α, is one of the most important properties to evaluate in solid materials because it determines the thermal stress that occurs between different materials with changing temperature. In particular, when a cryogenic apparatus composed of many kinds of materials is cooled for use, a large thermal stress occurs inside the apparatus. Therefore, accurate estimates of α are very important in cryogenic engineering to design apparatus. High sensitivity is required to measure α in solids at cryogenic temperatures because the α values in solids are generally small at low temperatures.

We have developed a laser interferometric dilatometer which has high sensitivity and an ability to carry out absolute measurements of the changes in specimen length in the temperature range 4.2 – 320 K (Okaji et al 1995, 1997). The laser interferometric dilatometer consists of an optical interferometer and an optical cryostat of the liquid-helium continuous-flow type. However, the use of liquid helium is troublesome and has obstructed the improvement of the efficiency in measurements.

In this work, we have developed a new low-temperature laser interferometric dilatometer which is a liquid-helium-free system. The new apparatus is composed of an optical interferometer and a cryogenic refrigerator. The value of α of a single crystal of silicon was measured over the temperature range 10 – 320 K to evaluate the performance of our new system, and the measured results were compared with the value recommended by the Committee on Data for Science and Technology (CODATA) (White and Minges 1985).

2 Experimental apparatus and specimen

A schematic diagram of the new laser interferometric dilatometer is shown in figure 1. The cryogenic refrigerator (V204SC, Daikin Industries Ltd, 4-12 Nakazaki-Nishi 2-chome, Kita-ku, Osaka, Japan) is fixed on a support frame and is inserted into a cryostat as shown in figure 1. Such cryogenic refrigerators usually generate vibrations with amplitude of several 100 μm due to the reciprocating motion of the piston in the cylinder. To suppress the influence of the vibration in the measurements, we adopted the cryostat with a vibration-free refrigeration transfer interface (202 series, Nagase & Co. Ltd, 5-1 Nihonbashi Kobunacho, Chuo-ku, Tokyo, Japan). The cryogenic refrigerator and the cryostat are connected to each other by a vacuum bellows to prevent the propagation of vibration from the cryogenic refrigerator to the cold head in the cryostat. The nominal

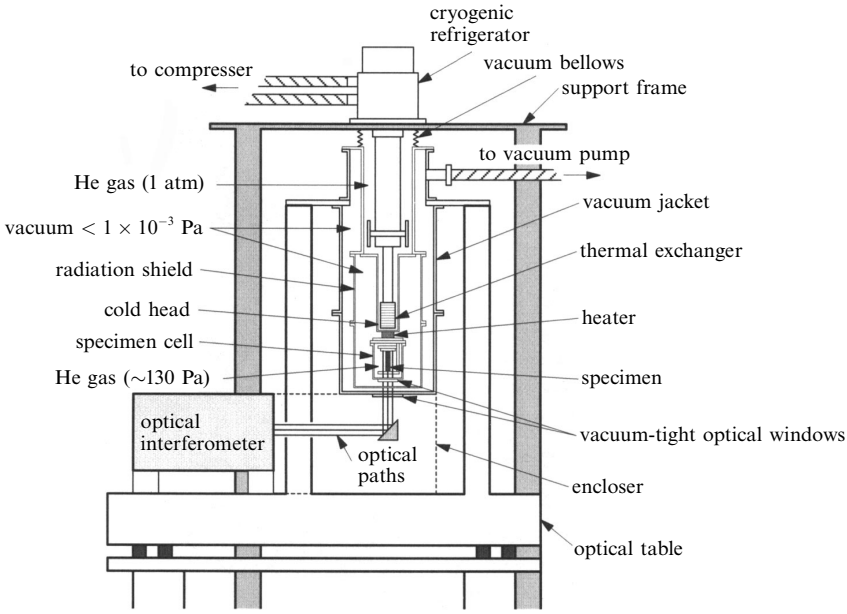

Figure 1. Schematic diagram of the low-temperature laser interferometric dilatometer with a cryogenic refrigerator.

amplitude of the vibration at the cold head of the cryostat is less than 1 μm. The lowest temperature of the cold head achievable with a specimen cell is about 10 K, and the cooling time required from room temperature to 10 K is about 5 h.

In the α measurements, the temperature of a specimen was raised in increments, ΔT of 10 K to 320 K and kept constant at each step for 1 h. The α value was determined from the changes in length, ΔL, with temperature, ΔT, with reference to the initial specimen length at room temperature, L_0 ($=20.002$ mm), according to the equation $\alpha = 1/L_0(\Delta L/\Delta T)$.

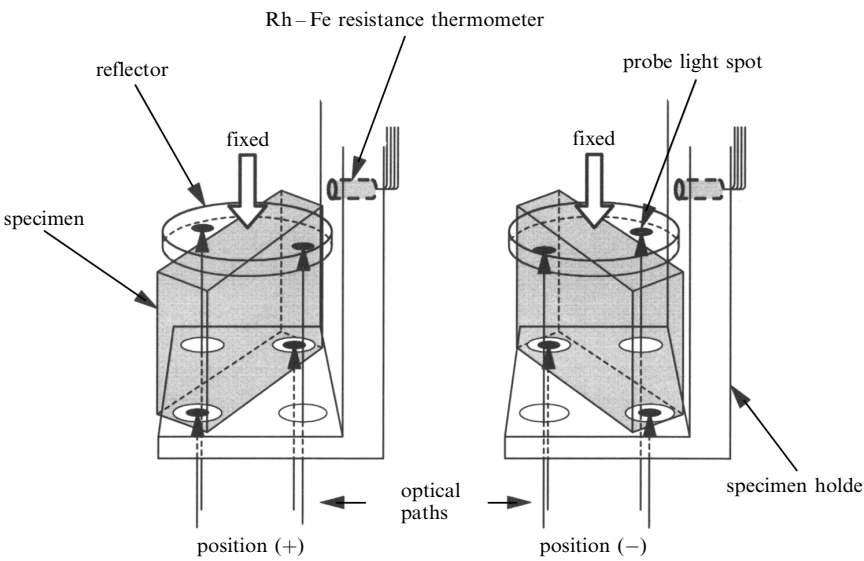

Figure 2. Arrangements of the specimen in the specimen cell.

The changes in length, ΔL, were measured with a double path type optical interferometer utilising an optical heterodyne method. The details of the optical interferometer have been reported in previous papers (Okaji and Yamada 1997; Okaji et al 1997). In figure 2, the arrangements of the specimen and specimen holder are shown. The probe lights from the optical interferometer were reflected by the front surface of the specimen and the reflector on the rear surface of the specimen. The reflector was fixed to the specimen by a parallel spring. The sign of the detected ΔL depended on the position of the specimen. The ΔL were detected as changes in the phase difference between the reference signal and the probe signal by a digital lock-in amplifier (model SR 850, Stanford Research Systems, 1290-D Reamwood avenue, Sunnyvale, CA 94089, USA). The sensitivity of the digital lock-in amplifier was 0.001 deg (which corresponds to 4.4×10^{-3} nm), but the actual resolution of the optical interferometer was about 0.1 nm because of air disturbances in the laboratory and the instability of room temperature.

The temperature change, ΔT_m, was measured by means of calibrated Rh – Fe resistance thermometer (RF-100, Lake Shore Cryotronics, Inc., 64 East Walnut Street, Westerville, OH 43081-2399, USA) which was put on the specimen holder, as shown in figure 2. The sensitivity of the thermometer is about 1 mK over the measured temperature range. Helium gas at about 130 Pa was used as a thermal exchange gas and was sealed in the specimen cell at room temperature to improve the uniformity of temperature in the specimen cell. The changes in pressure of the thermal exchange gas with temperature change did not influence the measurements of displacement by the optical interferometer.

A high-purity single crystal of silicon made by the floating zone method was prepared for the evaluation of performance in the present measurement system. The shape of the specimen was a rectangular prism with a base 8 mm $\times$ 20 mm and a height of 20 mm. Both base surfaces were parallel and were polished flat and smooth. We measured α in the height direction of the specimen corresponding to crystallographic axis (001). The α of silicon is isotropic because the symmetry of its crystal structure is cubic.

3 Measurement results and discussion

The expected vibration amplitude in the specimen cell was about 1 μm, whereas the resolution of our optical interferometer was 0.1 nm. We examined the possibility of carrying out measurements of a very small displacement in the presence of a large amplitude vibration. Under these circumstances, our measurement system had two advantages. First, our optical interferometer was sensitive only to the change of distance between both base surfaces of the specimen and was insensitive to the parallel movement of the specimen with a reflector. Therefore, we expected that the influence of vibration on the specimen would be decreased, because the reflector was held on the rear surface of the specimen by a parallel spring. The detected amplitude of vibration of the phase of the probe signal from a vibrating specimen observed by an oscilloscope was 0.1 – 0.2 fringe (corresponding to 15.8 – 31.6 nm). The second advantage was that it was possible to remove the frequency components of vibration higher than 2 Hz by choosing a suitable value for the time constant on the lock-in amplifier, because the frequency of the periodic motion of our cryogenic refrigerator was 2 Hz. The standard deviation of the scatter of the output signal as a function of time constant is shown in figure 3. The result shows that it was possible to measure the changes in length in the presence of a vibration with a scatter similar to a vibration-free condition when the value of the time constant is more than 30 ms. Accordingly, we set the time constant at 3 s for the present measurements. Thus, we are able to measure the displacement in the presence of vibration with sufficient sensitivity to determine α.

Typical measured results for α for silicon at position (+) and position (−) (see figure 2) are shown in figure 4. The solid line in figure 4 is the result of least squares regression of a

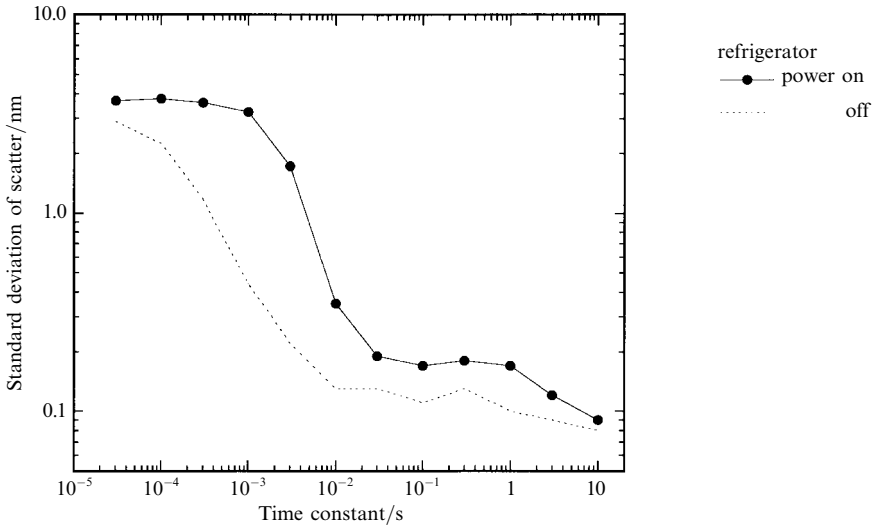

Figure 3. Standard deviation of the scatter of an output signal in a displacement measurement as a function of the lock-in amplifier time constant.

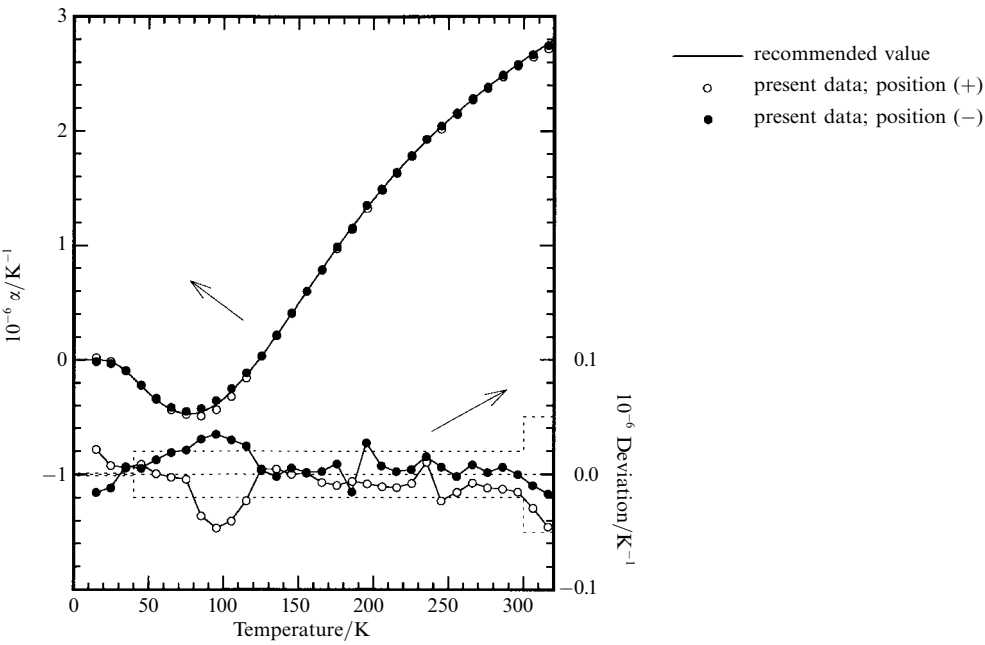

Figure 4. Typical measured α values at position (+) and position (−), and the deviation from the recommended values. The dashed lines represent the maximum probable error of the recommended value (0.001×10^{-6} K^{-1} below 40 K, 0.02×10^{-6} K^{-1} at 40 – 300 K, and 0.05×10^{-6} K^{-1} at 300 – 800 K) and the zero level.

fifth-order polynomial onto the recommended α values for silicon from CODATA (White and Minges 1985) over the temperature intervals 10 – 40, 40 – 100, and 100 – 350 K. This curve was in good agreement with the recommended values and we compared our measured results with it. The maximum deviation of the measured α value from the recommended value was about 0.05×10^{-6} K^{-1}. It is noted that the deviations of the measured values at position (+) and position (−) are approximately symmetric with respect to the recommended value. The total change in length detected by the optical interferometer, ΔL, is

expressed as $\Delta L = \Delta L_s + L_{pd} + L_{pi}$, where ΔL_s is the actual change in specimen length, ΔL_{pd} is an offset term which depends on the position of the specimen, and ΔL_{pi} is an offset term which is independent of the position of the specimen. It was considered that ΔL_{pd} was mainly caused by the deformation of the cryostat with temperature, and the sign of ΔL_{pd} was reversed for the specimen position. Therefore, ΔL_{pd} is cancelled by averaging values of ΔL measured at position (+) and position (−). ΔL_{pi} can be removed by measuring specimens of different length. In the present work, we carried out measurements at each specimen position three times alternately to remove the ΔL_{pd} term. The scatter of $(\alpha_+ + \alpha_-)/2$ from an α_{ave}, where α_{ave} was determined by regressing a fifth-order polynomial onto all measured results over the temperature intervals 10 – 40, 40 – 100, and 100 – 350 K, α_+ and α_- are values of α at position (+) and position (−) of the specimen, respectively, as is shown in figure 5. As shown in figure 5, the scatter of $(\alpha_+ + \alpha_-)/2$ from α_{ave} values is independent of temperature over the measured temperature range and the standard deviation of the scatter is 0.0068×10^{-6} K^{-1}. The reproducibility of the measured α in the present system is equal to or higher than that in our previous system using liquid helium (Okaji et al 1995, 1997). The total α_{ave} and the deviation from the recommended value are shown in figure 6. The measured value was in agreement with the recommended value to within 0.01×10^{-6} K^{-1} in the temperature range 10 – 300 K. The deviation was less than the maximum probable error of the recommended value over 40 – 300 K. However, the deviation was larger than the maximum probable error of the recommended value below 40 K. This is due to insufficient resolution of the present dilatometer to measure α to within 0.001×10^{-6} K^{-1}. On the other hand, the deviation tended to increase above room temperature and, for example, the deviation was about -0.042×10^{-6} K^{-1} at 315 K.

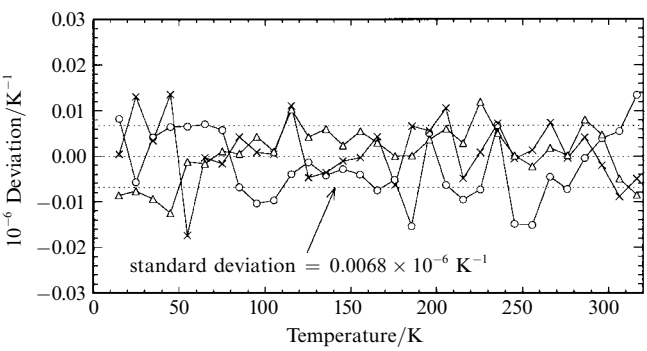

Figure 5. Reproducibility of $(\alpha_+ + \alpha_-)/2$ over three measurements.

In our system, the thermometer and the specimen were physically separated for two reasons. First, the thermometer was too large to put into the specimen and second, excessive stress of the electric wires of the thermometer with temperature should be prevented. We measured the temperature difference between the specimen and the thermometer, δT, by putting a dummy specimen with an extra small resistance thermometer at the specimen position. The temperatures of the extra thermometer and the usual Rh – Fe resistance thermometer were measured simultaneously under the usual step-by-step heating pattern. The temperature dependence of δT at the end of a temperature step, where $\delta T = T_{sp} - T_{RF}$, where T_{sp} and T_{RF} denote the temperature of the specimen and the Rh – Fe resistance thermometer, respectively, is shown in figure 7. δT was very small (∼15 mK) and constant below 220 K, and increased with temperature [$d(\delta T)/dT = -0.0016$]. The maximum value of $d\alpha/dT$ below 220 K was 0.019×10^{-6} K^{-1} around 150 K. Thus, the maximum error of the measured α caused by δT was estimated at 0.0003×10^{-6} K^{-1} below 220 K. The error was much smaller than the scatter of α_{ave} shown in figure 5, and the error caused by δT could be ignored below 220 K. On the other hand,

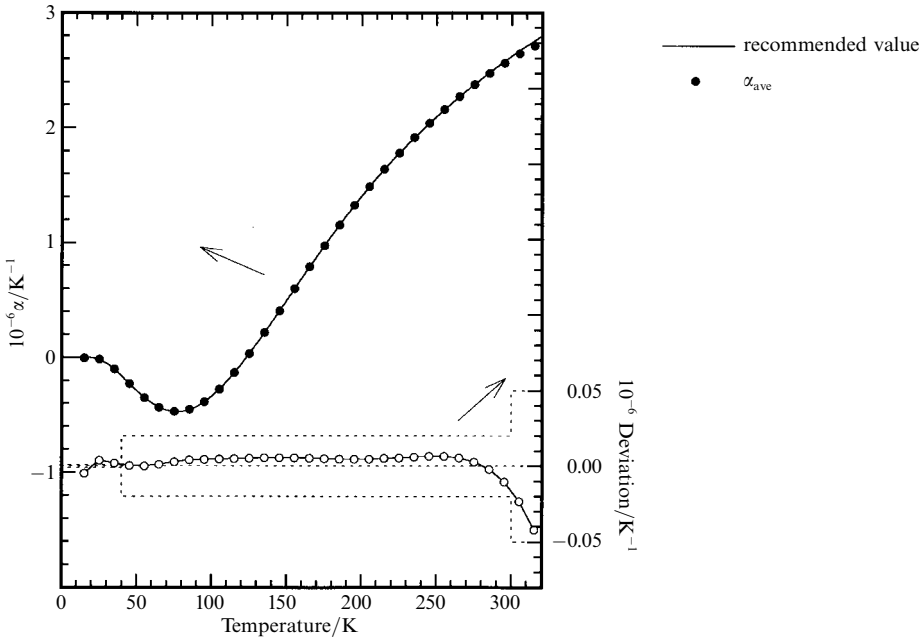

Figure 6. Comparison of the total α_{ave} with the recommended value. The open circles denote the deviation of the total α_{ave} from the recommended value.

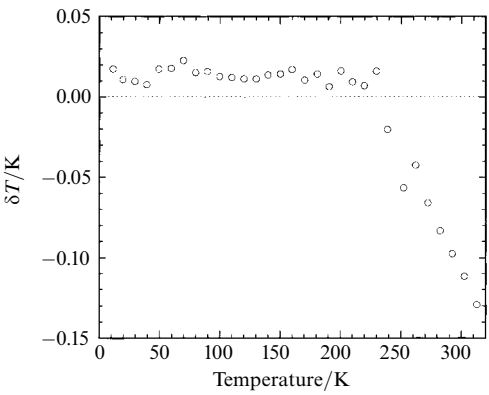

Figure 7. The difference in temperature between the specimen and the Rh – Fe resistance thermometer, δT, as a function of temperature.

above 220 K the maximum deviation of the measured α caused by δT became -0.0011×10^{-6} K^{-1}. In this temperature region, there was an additional error caused by the change of δT with temperature. The additional error was $[d(\delta T)/dT]\alpha$; for example, it was estimated to be -0.0044×10^{-6} K^{-1} at 315 K. Therefore, the maximum total error caused by δT was estimated to be -0.0055×10^{-6} K^{-1} above 220 K. However, the deviation of the total α_{ave} from the recommended value was larger than the estimated total error caused by δT above room temperature as shown in figure 6. We considered that the increased deviation above room temperature was caused by the position-independent offset term (ΔL_{pi}). It was necessary to carry out measurements with specimens of different length to estimate ΔL_{pi}.

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4 Summary

A laser interferometric dilatometer with a cryogenic refrigerator was developed for the measurement of the linear thermal expansion coefficient with high accuracy. Measurements of a high-purity single crystal of silicon were carried out over the temperature range 10 – 320 K to evaluate the performance of the laser interferometric dilatometer. The measured values were also compared with the values recommended by CODATA. The values of α_{ave} showed extremely good agreement with the value recommended by CODATA below room temperature (the deviation from the recommended value is less than $\pm 0.01 \times 10^{-6}$ K^{-1}). Above room temperature, the deviation increased with temperature (the maximum deviation was about -0.042×10^{-6} K^{-1} at 315 K). The deviation may be related to the offset term which is independent of the specimen positions at high temperature.

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