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Absolute Thermal Expansion Measurements of Single-Crystal Silicon in the Range 300–1300 K with an Interferometric Dilatometer[1]

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The thermal expansion coefficient of single-crystal silicon has been measured in the range 300–1300 K using an interferometric dilatometer. The measurement system consists of a double-path optical heterodyne interferometer and a radiant image furnace with a quartz vacuum tube, which provides both accuracy and rapidity of measurement. The uncertainties in length and temperature determination are within 4 nm and 0.4 K, respectively. A high-purity dislocation-free FZ silicon single crystal was used in the study. Thermal expansion coefficients of silicon oriented in the [111] direction have been determined over the temperature range from 300 to 1300 K. The standard deviation of the measurement data from the best fitting for the fifth-order polynomial in temperature is $2.1 \times 10^{-8}\,\mathrm{K}^{-1}$. The present value for the thermal expansion coefficient agrees within $9 \times 10^{-8}\,\mathrm{K}^{-1}$ with the interferometric measurement of polycrystalline pure silicon by Roberts (1981) between 300 and 800 K and within $1.2 \times 10^{-7}\,\mathrm{K}^{-1}$ with the single-crystal X-ray diffractometric measurement by Okada and Tokumaru (1984) between 300 and 1300 K.

KEY WORDS: high temperature; interferometry; single-crystal silicon; thermal expansion.

1. INTRODUCTION

Silicon has been considered to be a suitable standard material for thermal expansion at higher temperatures, because of its small expansion coefficient, high purity, isotropic expansion coefficient, and relatively high melting point [1].

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There exist many investigations on the thermal expansion of silicon at high temperatures, which are reviewed in Ref. 2. However, most of the references are rather old and the data are widely divergent and not reliable enough from the present technical point of view. Absolute measurements using interferometric techniques have been carried out by Ibach [3] in 1969 and Roberts [4] in 1981. The maximum temperature of their data is about 850 K. Okada and Tokumaru [2] in 1984 measured the thermal expansion coefficient of single-crystal silicon between room temperature and 1500 K using an X-ray diffractometer. The accuracy of their data is limited to about $2 \times 10^{-7}\,K^{-1}$, which is caused mainly by the error in temperature measurement.

In the conventional DC fringe detection method, such as Fizeau interferometry, the change in DC level and poor visibility of the interference fringes increase the uncertainty in the fringe determination and reduce the accuracy in length measurement. In contrast, the AC fringe detection method is not influenced by the DC-level variations. Therefore, optical heterodyne AC interferometry is considered to be a suitable technique for high-temperature absolute dilatometry.

In this paper, we describe a coefficient of the thermal expansion measurement system for higher temperature ranges, which has been developed at the National Research Laboratory of Metrology (NRLM) [5, 6], and the measurements on pure single-crystal silicon in the range 300–1300 K.

2. MEASUREMENTS

The silicon single-crystal specimen used in the present work was cut from a FZ single-crystal cylinder manufactured by Shin-etsu Chemical Co. Ltd. Resistivity of the material is greater than 1 k$\Omega \cdot$cm. The cylinder was originally prepared for X-ray interferometric measurements of lattice spacing and for precise density measurements at NRLM. The interferometer specimen was cut in the form of a rectangule 8.100 mm in length and 10 × 14 mm in cross section. Thermal expansion coefficients of the specimen in the [111] direction have been determined over 100 K temperature intervals in the temperature range from 300 to 1300 K.

A schematic diagram of the double-path optical heterodyne interferometer used in the present work is shown in Fig. 1. The system provides both high accuracy and versatility, with nanometer-order resolution in length measurement, and a wide tolerance in specimen shapes and dimensions [5].

In optical heterodyne interferometry, a frequency difference is introduced between the two beams in the interferometer. In this case, the

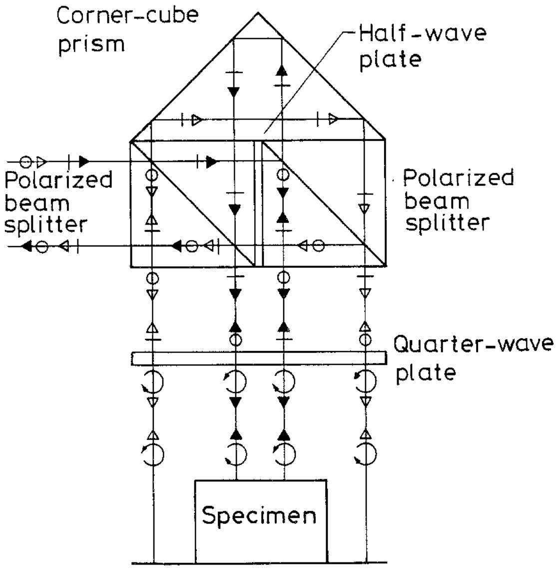

Fig. 1. The double-path interferometer.

difference is produced by Zeeman splitting of the stabilized Zeeman laser. The phase difference between the interfering optical beams which corresponds to the phase change of the beat signal can be measured accurately using a frequency counter. Resolution of 1 nm in length measurement can be obtained easily between room temperature and 1100 K [6].

For the present study, we have modified the previous system [5, 6] to obtain both a higher sensitivity and a better long-term stability in the length measurement. The mechanical arrangement of the dilatometer is shown in Fig. 2. The furnace is composed of a quartz vacuum tube and a radiant image furnace. The interferometer and the Zeeman-stabilized laser are set below the furnace. The interferometer is temperature controlled to within 0.05 K in order to obtain long-term stability. The whole system is set up on the vibration-proof iron base plate. The specimen was polished on both ends to be flat within one-tenth of a wavelength and parallel within 5 arc s. Pure polycrystalline silicon was used as a reflecting plate and was also polished to within one-tenth of a wavelength. The reflecting plate is set on the specimen and placed in a graphite enclosure. The laser beam from the interferometer is introduced from the bottom of the vacuum tube. This arrangement has the advantages of reducing temperature changes to the optics from the furnace and versatility of specimen shape and optical alignment.

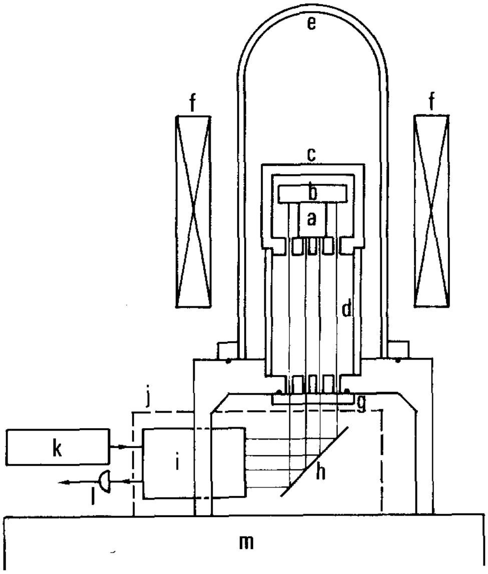

Fig. 2. The arrangement of the measurement system. a, Specimen; b, silicon plane reflector; c, graphite enclosure; d, alumina supporting tube; e, quartz vacuum tube; f, radiant image furnace; g, optical window; h, plane reflector; i, double-path interferometer; j, constant-temperature enclosure; k, Zeeman-stabilized laser; l, optical detector; m, vibration-proof base plate.

R-type thermocouples were used for temperature measurements. The thermocouples were calibrated in the testing and inspection section at the NRLM with an accuracy of 0.2 K below the Au point (1337 K).

3. RESULTS

The linear thermal expansion of the specimen was determined between equilibrium temperatures. The coefficients were calculated using

$$\alpha_m = \Delta L/(L_0 \Delta T) \qquad (1a)$$

where $\Delta L = L_2 - L_1$, L_0 is the specimen length at 293 K, and $\Delta T = T_2 - T_1$. The subscript 1/2 represents the initial/final state.

The measured expansion coefficients (in K^{-1}) are fitted by a fifth-order polynomial function of temperature using the usual least-squares method. The resulting polynomial coefficients in the temperature range 300 to 1300 K are given by the following equation:

$$\alpha_m = (a + bT + cT^2 + dT^3 + eT^4 + fT^5) \times 10^{-6} \tag{1b}$$

$$a = -3.7321326$$

$$b = 3.9818886 \times 10^{-2}$$

$$c = -8.6755433 \times 10^{-5}$$

$$d = 9.6666003 \times 10^{-8}$$

$$e = -5.3256454 \times 10^{-11}$$

$$f = 1.1548206 \times 10^{-14}$$

where T is in K. The deviations of the measured values for several runs from the calculated values as defined by Eq. (1b) are shown in Fig. 3. The standard deviation of the 80 data points from the best least-squares fit is $2.1 \times 10^{-8}\ \mathrm{K}^{-1}$ (about 0.55 % of the average expansion coefficient). Corrections for finite value of ΔT's [7] were applied to obtain the limiting value of the coefficients defined by

$$\begin{aligned}\alpha &= dL/(L_0\, dT) \\ &= (A + BT + CT^2 + DT^3 + ET^4 + FT^5) \times 10^{-6}\end{aligned} \tag{2}$$

$$A = -3.6599918$$

$$B = 3.9577389 \times 10^{-2}$$

$$C = -8.6489150 \times 10^{-5}$$

$$D = 9.6569768 \times 10^{-8}$$

$$E = -5.3256454 \times 10^{-11}$$

$$F = 1.1548206 \times 10^{-14}$$

The linear thermal expansion coefficients calculated by Eq. (1b) and Eq. (2) are given in Table I, at intervals of 100 K. The maximum difference between the measured and the corrected coefficients is $2.1 \times 10^{-8}\ \mathrm{K}^{-1}$ at the low-temperature end.

4. ERROR ANALYSIS

The error sources in length measurements are (1) the resolution of fringe determination, within 1 nm; (2) the long-term stability of the interferometer, 3 nm; and (3) the linearity of the AC fringe determination, 2 nm. These give the random uncertainty in length measurement as $\delta L = 4$ nm $[= (1^2 + 3^2 + 2^2)^{1/2}]$, because all of these errors are considered to be ran-

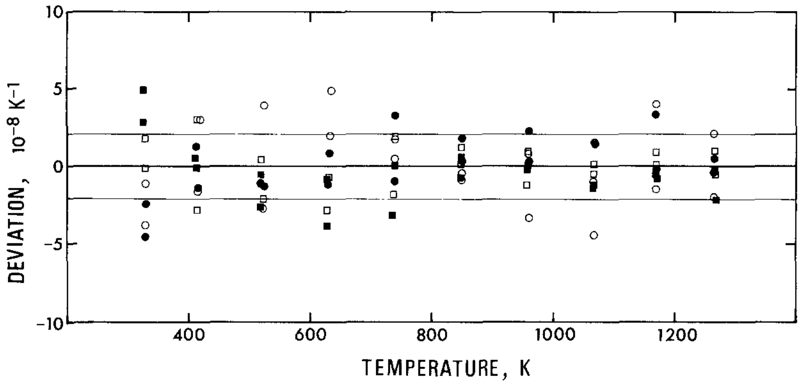

Fig. 3. Deviation of measured values for several runs (○, ●, □, ■: first–fourth runs) from the calculated values as defined by Eq. (1b).

Table I. Generated Smooth Values of the Thermal Expansion Coefficient from Polynomial Fits, Eqs. (1b) and (2)[a]

T (K)	$10^6\,\alpha_m$ (K^{-1})	$10^6\,\alpha$ (K^{-1})
300	2.612	2.633
400	3.256	3.268
500	3.604	3.610
600	3.803	3.805
700	3.941	3.942
800	4.063	4.063
900	4.180	4.180
1000	4.289	4.290
1100	4.383	4.383
1200	4.465	4.464
1300	4.563	4.560

[a] The maximum amount of the correction of the finite temperature interval is about $2\times10^{-8}\,K^{-1}$ at the lowest-temperature end.

dom errors. The error sources in temperature measurement are (1) the calibration of the thermocouples, $\delta T_c = 0.2$ K, which is considered to be a systematic error; (2) the temperature resolution, 0.01 K; and (3) the temperature difference between the specimen and the thermocouples or the temperature distribution near the specimen, 0.3 K. These give the random error δT_r as 0.3 K. Thus, the total uncertainty in the determination of expansivity (in T^{-1}) is given as follows:

$$\begin{aligned}\alpha &= [2 \times (\delta L/L_0 \, \Delta T)^2 + 2 \times (\alpha_a \delta T_r/\Delta T)^2]^{1/2} + 2 \times \alpha_a \delta T_c/\Delta T \\ &= [0.49 \times 10^{-17} + 2.74 \times 10^{-16}]^{1/2} + 1.4 \times 10^{-8} \\ &= 3.2 \times 10^{-8} \qquad (3)\end{aligned}$$

where $\delta L = 4 \times 10^{-9}$ m, $L_0 = 8.1 \times 10^{-3}$ m, $\Delta T = 100$ K, and $\alpha_a = 3.9 \times 10^{-6}$ K^{-1}; the average thermal expansion coefficient of the silicon in the measured temperature range, $\delta T_r = 0.3$ K, and $\delta T_c = 0.2$ K. In the present case, the square of the uncertainty in temperature measurement is about five times larger than that in length measurement. Thus, the major error source of the measurement is attributed to the temperature measurement. The first square-root term in Eq. (3) represents the estimated value of the total random uncertainty, the amount of which is almost the same as the measured standard deviation of each data point from the polynomial fit, 2.1×10^{-8} K^{-1} (Fig. 3).

5. DISCUSSION

The present results are compared with the expansion coefficients of some selected references, as shown in Fig. 4 and Table II. Figure 4 shows the deviation of the reported data (Lyon et al. [8], below 340 K; Roberts [4], 300–850 K; Bennett [9], 300–680 K; Ibach [3], 300–850 K; Okada and Tokumaru [2], 300–1300 K; CINDAS [10], 300–1300 K) from the present polynomial fit calculated from Eq. (2). The present data agrees well with the very precise data of Lyon et al. [8] near room temperature and with the interferometric measurement by Roberts [4] below 600 K, but the difference increases to 9×10^{-8} K^{-1} at higher temperatures. Bennett's interferometric data [9] are larger by about 10×10^{-8} K^{-1} at the high-temperature end. Ibach's interferometric data [3] are below the present values by 8×10^{-9} K^{-1} near room temperature. The maximum temperatures of these interferometric measurements are limited to temperatures below 850 K.

There are not many reliable measurement results in the temperature region above 850 K. The thermal expansion coefficient of single-crystal silicon was given by Okada and Tokumaru [2] using X-ray diffractometry.

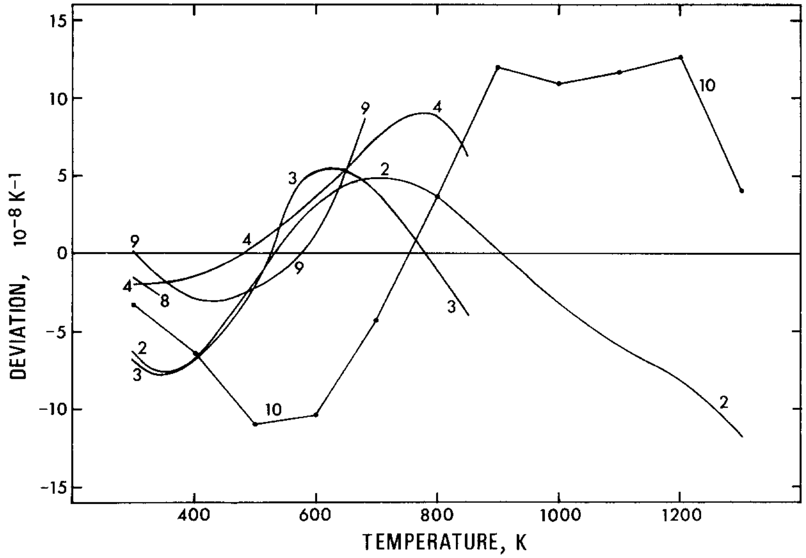

Fig. 4. Deviation of the reported data (Lyon et al. [8], below 340 K; Roberts [4], 300–850 K; Bennett [9], 300–680 K; Ibach [3], 300–850 K; Okada and Tokumaru [2], 300–1300 K; CINDAS [10], 300–1300 K) from the present polynomial fit calculated from Eq. (2).

Table II. Comparisons Among the Present Data of the Linear Thermal Expansion Coefficients Derived from Eq. (2) and Previously Reported Data

	$10^6\alpha$ (K^{-1})					
T (K)	Present work (1988)	Roberts [4] (1981)	Bennett [9] (1981)	Ibach [3] (1969)	Okada and Tokumaru [2] (1984)	CINDAS [10] recommended
300	2.633	2.614	2.635	2.56	2.573	2.6
350	2.999	2.980	2.977		2.923	
400	3.268	3.253	3.239	3.20	3.201	3.2
450	3.465	3.458	3.437		3.420	
500	3.610	3.614	3.588	3.59	3.593	3.5
550	3.719	3.738	3.709		3.729	
600	3.805	3.842	3.817	3.86	3.836	3.7
650	3.877	3.933	3.929		3.921	
700	3.942	4.016	4.062	3.98	3.991	3.9
750	4.003	4.090			4.049	
800	4.063	4.151		4.05	4.099	4.1
850	4.122	4.191		4.08	4.143	
900	4.180				4.184	4.3
1000	4.290				4.258	4.4
1100	4.383				4.325	4.5
1200	4.464				4.385	4.6
1300	4.560				4.440	4.6

The difference between the present data and the data of Ref. 2 is $1.2 \times 10^{-7}\,K^{-1}$ in the range 300 to 1300 K, which is not statistically significant because the scatter of the data of Ref. 2 is about $2 \times 10^{-7}\,K^{-1}$. The present data also agree with the recommended value of Ref. 10 over the temperature range, 300 to 1300 K, within $1.2 \times 10^{-7}\,K^{-1}$.

6. CONCLUSIONS

(1) The thermal expansion coefficient of FZ single-crystal silicon along the [111] axis was accurately measured by means of an optical heterodyne interferometric dilatometer in the temperature range between 300 and 1300 K.

(2) The fifth-order polynomial equation [Eq. (2)] for the limiting value of the linear thermal expansion coefficient is obtained.

(3) The standard deviation of the linear thermal expansion coefficient from the polynomial fit is $2.1 \times 10^{-8}\,K^{-1}$.

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