Title	Refractive index and extinction coefficient of Si at 400nm between 10 and 300K
Author(s)	Nagakubo, Akira; Ogi, Hirotsugu; Hirao, Masahiko
Citation	Japanese Journal of Applied Physics. 2015, 54(12), p. 128001-1-128001-3
Version Type	AM
URL	https://hdl.handle.net/11094/84532
rights	© 2015 The Japan Society of Applied Physics.
Note	

Refractive index and extinction coefficient of Si at 400 nm between 10 and 300 K

Akira NAGAKUBO[1]*, Hirotsugu OGI[1], and Masahiko HIRAO[1],

[1] *Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan*

Optical properties of Si are important and widely studied. However, temperature behaviors of refractive index for visible light at low temperatures have not been measured. In this study, we measured complex refractive index of Si at 400 nm between 10 and 300 K using picosecond ultrasound spectroscopy. Measured refractive index at room temperature well agrees with reported values, confirming the accuracy of our measurement, and we found that refractive index at 10 K is smaller than reported values.

KEYWORDS: Refractive index, cryogenic temperature, picosecond ultrasound spectroscopy

Silicon is one of the most important materials for a substrate of various thin films, solar cell, and so on, and its optical properties are important; many researchers have measured refractive index or dielectric constant for infrared regions[1–4] and visible light.[5–11] Their temperature behaviors are also studied in wide temperature and wavelength ranges. Although there are some reports for temperature dependence of the refractive index of Si in an infrared region,[12–15] continuous temperature behaviors of refractive index for visible light are not reported below room temperature; above room temperature, Vuye *et al.* measured refractive index and extinction coefficient of Si between 293 and 723 K for 264 ∼ 826 nm light,[16] and Jellison and Burke obtained refractive index and reflectivity as a function of wavelength and temperature.[17] Below room temperature, Dash and Newman measured extinction coefficient for 364 ∼ 1032 nm at 77 and 300 K.[18] Daunois and Aspnes reported complex dielectric constants 225 ∼ 413 nm at 10 and 293 K,[19] and Jellison and Modine measured them for 275 ∼ 729 nm at 10 K, 300 K, and several higher temperatures.[20–22] Lautenschlager *et al.* measured continuous temperature behaviors of the interband critical point energies between 30 and 820 K, but they reported dielectric constants only at 30, 243, 510, and 793 K.[23] However, continuous temperature behaviors of refractive index have not been reported, and their reported values do not agree with each other. In this study, we measure temperature behaviors of refractive index and extinction coefficient of Si at 400 nm every 20 K below room temperature, and found that refractive index at 10 K is smaller than reported values by 2.1 ∼ 3.6%.

We measure 400-nm-wavelength refractive index n_0 and extinction coefficient κ of a 0.5 mm-thick float-zone (FZ) silicon using picosecond ultrasonics between 10 and 300 K. We developed an optics for cryogenic-temperature picosecond ultrasonics, whose details appear elsewhere.[24,25] We use a titanium/sapphire pulse laser, whose wavelength, repetition rate, and band width are 800 nm, 80 MHz, and 7.0 nm, respectively. To improve signal-to-noise ratio, we modulate the pump light pulses as 100 kHz and extract the modulated component using an lock-in amplifier. The wavelength λ of probe light is converted into 400 nm through a second harmonic generator of 2-mm β-barium borate crystal, whose band width becomes 2.0 nm. Both of the pump and probe light pulses are normally incident to a specimen through a 20-times objective lens. We measure the spectra of pump and probe light pulses using a spectrometer (USB2000+VIS-NIR, Ocean Optics), whose accuracy for wavelength is corrected by a Hg-Ar calibration source (HG-1, Ocean Optics).

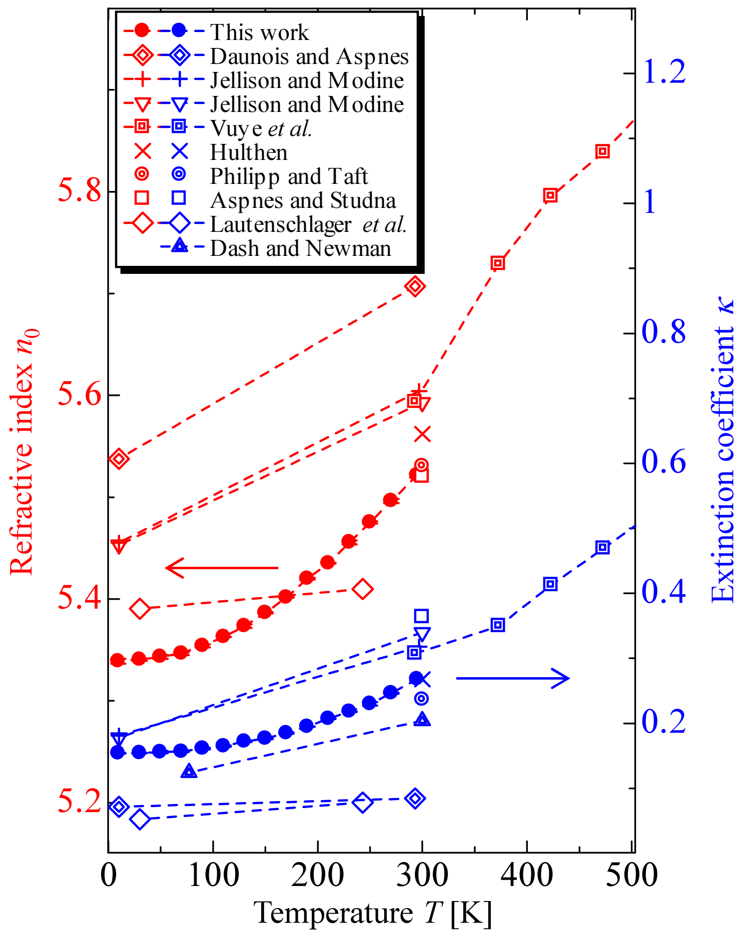

Fig. 1. (Color online) Measured and reported temperature dependence of n_0 and κ. Reference data are from Daunois and Aspnes,[19] Jellison and Modine,[21,22] Vuye *et al.*,[16] Hulthén,[9] Philipp and Taft,[5] Aspnes and Studna,[11] Lautenschlager *et al.*,[23] and Dash and Newman.[18]

We use a (100) single-crystal float-zone silicon, and deposited 10-nm Al thin film on the surface as a transducer. The pump light pulse is absorbed in it and excites an

*E-mail address: akira.nagakubo@abc.me.es.osaka-u.ac.jp

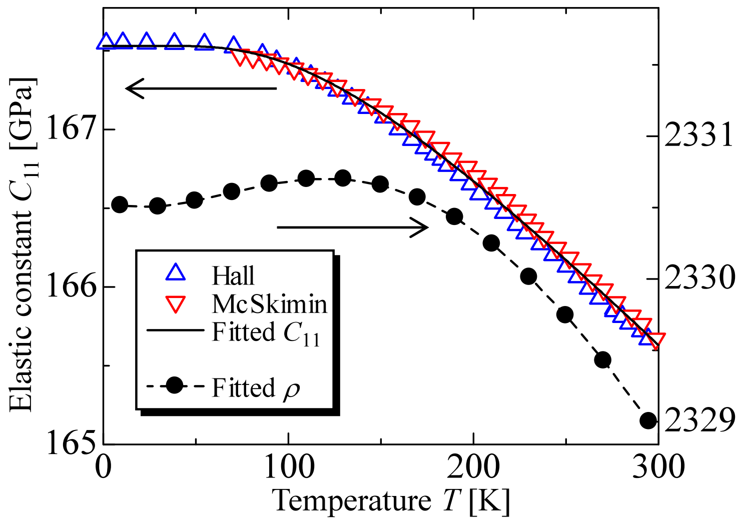

Fig. 2. (Color online) Temperature dependence of (slid line) elastic constants C_{11} and (dashed line with solid circle) mass density ρ. Reference data of C_{11} are from McSkimin[28] and Hall.[29] We calculated temperature dependence of mass density ρ from a reported temperature dependence of lattice constant.[32]

ultra-sharp strain pulse, which diffracts the time-delayed probe light backward. The diffracted light in the specimen and the reflected probe light at the surface interfere with each other, resulting in an oscillating signal, whose frequency f is written by Bragg's condition:[26,27]

$$f = \frac{2n_0 v}{\lambda} \tag{1}$$

where n_0 and v are the refractive index and longitudinal-wave sound velocity of Si, respectively. This is called Brillouin oscillation and we can obtain n_0 with known λ and v. From average values of reported temperature behaviors of the elastic constant C_{11} of Si,[28,29] we fitted Varshni's equation $C_{11}(T) = C_0 - s/[\exp(\theta/T) - 1]$,[30] where s and θ are fitting parameters relating to Grüneisen parameter and Debye characteristic temperature, respectively,[31] and T denotes temperature in Kelvin. Using $C_{11}(T)$ and temperature dependence of mass density $\rho(T)$, we calculate sound velocity $v(T) = \sqrt{C_{11}(T)/\rho(T)}$ at each temperature with reported temperature dependence of lattice constant[32] as shown in Fig. 2. Differences in C_{11} reported by McSkimin[28] and Hall[29] are less than 0.05% between 74 and 300 K.

Figure 3 (a) shows measured Brillouin oscillations of FZ-Si at each temperature, where the thermal background was subtracted. We observed exponentially decaying Brillouin oscillations at each temperature as shown in Fig. 3 (b), and its attenuation coefficient becomes smaller as the temperature decreases (Fig. 3 (c)), reflecting the decrease in extinction coefficient. We observed time-domain attenuation ($e^{-\alpha_t t}$), which can be converted into space-domain attenuation ($e^{-\alpha x}$) by $\alpha = \frac{\alpha_t}{v}$ since the amplitude of Brillouin oscillation reflects the amplitudes of probe light and ultrasound at the time t and the propagation distance $x = vt$. Because extinction coefficient of Si at 400-nm light is enough larger than acoustic attenuation, attenuation coefficient α of

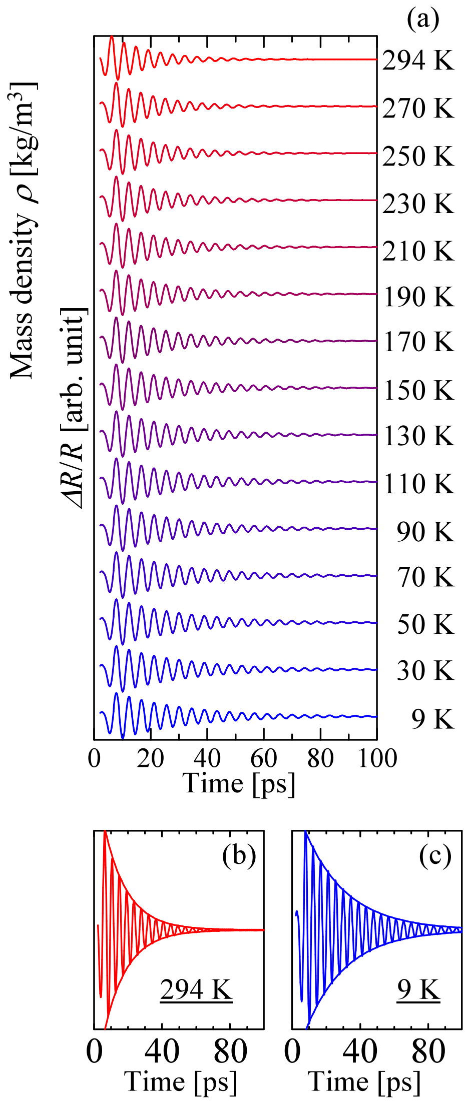

Fig. 3. (Color online) (a) Observed Brillouin oscillations at each temperature. (b and c) Brillouin oscillation with fitted envelopes at 294 and 9 K, respectively.

Brillouin-oscillation amplitude represents that of probe light: Acoustic attenuation of Si in a GHz-frequency range is estimated to be of the order of 10 $\sim$ 100 cm^{-1},[33,34] resulting in about 1% damping within 100 ps, which further decreases in low temperatures. On the other hand, measured α is 83800 $\pm$ 500 cm^{-1} at room temperature. Therefore, we neglect acoustic attenuation and calculate κ directly from the attenuation of Brillouin oscillation. When we write complex refractive index $n = n_0 - i\kappa$, the electric field of the probe light E in Si can be written by

$$E = E_0 e^{i\left[\omega t - \frac{\omega}{c/(n_0 - i\kappa)}x\right]} = E_0 e^{-\frac{2\pi\kappa}{\lambda}x} e^{i(\omega t - n_0 k x)} \tag{2}$$

where ω, c and k are angular frequency, velocity, and wave vector of the probe light in vacuum, and t and x denote time and propagation distance, respectively. The amplitude of a Brillouin oscillation is proportional to E^2, therefore, we can obtain the extinction coefficient as $\kappa = \frac{\alpha\lambda}{4\pi}$.

Measured refractive index and extinction coefficient are shown in Fig. 1 and Table I. We measured Brillouin oscillations at three different points on the specimen at each temperature, leading to a standard deviation less than 0.044% and 1.1% for n_0 and κ, respectively. At room temperature, we obtained $n_0 = 5.5215 \pm 0.0020$ and $\kappa = 0.2679 \pm 0.0020$ for $\lambda = 401.6$ nm, where n_0 agrees with averaged reported values by 0.8% difference ($n_0 = 5.567 \pm 0.035$ and $\kappa = 0.305 \pm 0.047$).[5,9,11,16,21,22] Our κ value is relatively smaller than reported values by 12%. Hara and Nishi reported that extinction coefficient of p-type Si increases with increases in the concentration of holes,[35] indicating that our FZ-Si has smaller carrier concentration.

Thermo-optic temperature coefficients dn_0/dT and $d\kappa/dT$ between 250 and 294 K are 1030 ± 43 and 843 ± 59 ppm/K, respectively. Vuye *et al.* measured n_0 and κ between 293 and 723 K for 264 $\sim$ 826 nm light, and from their data between 293 and 473 K, we estimated $dn_0/dT = 1381$ and $d\kappa/dT = 907$ ppm/K for $\lambda = 401.6$ nm, where $n_0 = 5.594$ and $\kappa = 0.311$ at 293 K.[16] n_0 and dn_0/dT measured by Vuye *et al.* are larger than our values and we consider that these discrepancies stem from following reasons: Vuye *et al.* measured the complex refractive index using an *in situ* ellipsometry, which makes it possible to reduce the native oxide layer. However, the measurements are more complicated and, nevertheless, the oxide layer cannot be neglected and affects the measured values; they estimated its thickness was 2.0 nm before the high-temperature measurements and it decreased to 1.6 nm after the measurements. They argued that the results were identical, but its effects are controversial. On the other hand, Brillouin-oscillation method is not affected by the oxide layer or surface thin film; to determine n_0, we use the frequency f of an observed signal, sound velocity v of Si, and wavelength λ of probe light. The measurement precision (standard deviation: SD) of f is 0.04% at most between 10 and 300 K, and inaccuracy or SD of v and λ is less than it: We used the average value of elastic constants independently measured by two groups. Their values well agree with each other, leading to consistent sound velocities within 0.02% difference between 74 and 300 K. As mentioned above, we performed wavelength correction for the spectrometer; we measured Ag and Hg characteristic spectra emitted by the calibration source between 405 and 922 nm, and determined a correction function. Using it, we obtained the 404-nm spectrum of Hg within 0.01% inaccuracy. Measurement precision and laser stability for wavelength are smaller than it by a factor 0.1. Therefore, we conclude that we succeeded in determining refractive index of Si within measurement precision.

Table I. Measured and reported refractive index n_0 and extinction coefficient κ of Si at room temperature (RT) and various temperature T for wavelength $\lambda = 401.6$ nm.

Reference	T (K)	n_0	κ
This work	295.0	5.5215 ± 0.0020	0.2679 ± 0.0020
	270.4	5.4962 ± 0.0020	0.2463 ± 0.0008
	250.0	5.4752 ± 0.0011	0.2301 ± 0.0025
	230.4	5.4557 ± 0.0019	0.2182 ± 0.0005
	210.3	5.4351 ± 0.0003	0.2071 ± 0.0005
	190.2	5.4199 ± 0.0008	0.1950 ± 0.0001
	170.1	5.4015 ± 0.0004	0.1849 ± 0.0010
	150.2	5.3865 ± 0.0006	0.1767 ± 0.0012
	130.0	5.3735 ± 0.0018	0.1720 ± 0.0010
	110.2	5.3627 ± 0.0002	0.1648 ± 0.0011
	90.1	5.3540 ± 0.0013	0.1611 ± 0.0013
	70.0	5.3466 ± 0.0014	0.1565 ± 0.0005
	49.7	5.3432 ± 0.0019	0.1557 ± 0.0016
	29.9	5.3404 ± 0.0011	0.1542 ± 0.0002
	9.4	5.3390 ± 0.0023	0.1541 ± 0.0004
[5]	R. T.	5.5307	0.2365
[7]	R. T.	5.7459	1.0799
[9]	R. T.	5.5622	0.2679
[11]	R. T.	5.5202	0.3635
[16]	293	5.5936	0.3073
[18]	300		0.2041
	77		0.1242
[20]	297		0.3321
	10		0.1867
[19]	293	5.7072	0.0844
	10	5.5377	0.0715
[21]	300	5.5929	0.3397
	10	5.4532	0.1777
[22]	297	5.6042	0.3177
	10	5.4552	0.1809
[23]	243	5.4098	0.0781
	30	5.3905	0.0524

n_0 and κ measured at 9.4 K are 5.3390 ± 0.0023 and 0.1541 ± 0.0004, respectively, which do not agree with reported values; there are two comparable values measured by Daunois and Aspnes[19] and Jellison and Modine,[21,22] and their n_0 at 10 K (5.5377 and 5.4542) are higher than our value by 3.7 and 2.2%, respectively. Although their room-temperature values (n_0 =5.7072 and 5.5986) are close to Vuye *et al.* ($n_0 = 5.5936$), they are also also higher than our room-temperature value (5.5215 ± 0.0020) and other reported values; Philipp and Taft[5] and Aspnes and Studna[11] reported $n_0 = 5.5307$ and 5.5202 at room temperature, respectively, which well agree with our value. Furthermore, ellipsometry measurements at low temperatures are also complicated and need many corrections:[21] Si surface has an oxide layer of unknown thickness and complex refractive index, and it is difficult to correctly establish the surface model. On the other hand, our low-temperature measurements need only well-known temperature behaviors of sound velocity. Therefore, we consider that we succeeded in determining refractive index even at low temperatures, and found that refractive index of Si at low temperatures are smaller than some reported values.

Summary, we have shown that Brillouin oscillation method is a powerful tool for refractive-index measurement. Using this method, we firstly reported complex refractive index of Si for 401.6 nm light between 10 and 300 K, and found that n_0 at 10 K is smaller than reported values. Our results will also contribute to many other optical measurements at cryogenic temperature using Si.

1) H. B. Briggs, Phys. Rev. **77**, 287 (1950).
2) C. D. Salzberg and J. J. Villa, J. Opt. Soc. Am. **47**, 244 (1957).
3) W. Primak, Appl. Opt. **10**, 759 (1971).
4) J. J. Villa, Appl. Opt. **11**, 2102 (1972).

5) H. R. Philipp and K. A. Taft, Phys. Rev. **120**, 37 (1960).
6) H. R. Philipp and H. Ehrenreich, Phys. Rev. **129**, 1550 (1963).
7) H. W. Verleur, J. Opt. Soc. Am. **58**, 1356 (1968).
8) H. R. Philipp, J. Appl. Phys. **43**, 2835 (1972).
9) R. Hulthén, Phys. Scr. **12**, 342 (1975).
10) G. K. M. Thutupalli and S. G. Tomlin, J. Phys. C **10**, 467 (1977).
11) D. E. Aspnes and A. A. Studna, Phys. Rev. B **27**, 985 (1983).
12) M. Cardona, W. Paul, and H. Brooks, J. Phys. Chem. Solids **8**, 204 (1959).
13) H. W. Icenogle, B. C. Platt, and W. L. Wolfe, Appl. Opt. **15**, 2348 (1976).
14) H. H. Li, J. Phys. Chem. Ref. Data **9**, 561 (1980).
15) J. Komma, C. Schwarz, G. Hofmann, D. Heinert, and R. Nawrodt , Appl. Phys. Lett. **101**, 041905 (2012).
16) G. Vuye, S. Fisson, V. N. Van, Y. Wang, J. Rivory, and F. Abelès, Thin Solid Films **233**, 166 (1993).
17) G. E. Jellison and H. H. Burke, J. Appl. Phys. **60**, 841 (1986).
18) W. C. Dash and R. Newman, Phys. Rev. **99**, 1151 (1955).
19) A. Daunois and D. E. Aspnes, Phys. Rev. B **18**, 1824 (1978).
20) G. E. Jellison and F. A. Modine, Appl. Phys. Lett. **41**, 180 (1982).
21) G. E. Jellison and F. A. Modine, J. Appl. Phys. **53**, 3745 (1982).
22) G. E. Jellison and F. A. Modine, Phys. Rev. B **27**, 7466 (1983).
23) P. Lautenschlager, M. Garriga, L. Viña, and M. Cardona, Phys. Rev. B **36**, 4821 (1987).
24) K. Tanigaki, T. Kusumoto, H. Ogi, N. Nakamura, and M. Hirao, Jpn. J. Appl. Phys. **49**, 07HB01 (2010).
25) A. Nagakubo, A. Yamamoto, K. Tanigaki, H. Ogi, N. Nakamura, and M. Hirao, Jpn. J. Appl. Phys. **51**, 07GA09 (2012).
26) A. Devos and R. Côte, Phys. Rev. B **70**, 125208 (2004).
27) H. Ogi, T. Shagawa, N. Nakamura, M. Hirao, H. Odaka, and N. Kihara, Phys. Rev. B **78**, 134204 (2008).
28) H. J. McSkimin, J. Appl. Phys. **24**, 988 (1953).
29) J. J. Hall, Phys. Rev. **161**, 756 (1967).
30) V. P. Varshni, Phys. Rev. B **2**, 3952 (1970).
31) H. Ledbetter, Phys. Status Solidi B **181**, 81 (1994).
32) D. N. Batchelder and R. O. Simmons, J. Chem. Phys. **41**, 2324 (1964).
33) J. Y. Duquesne and B. Perrin, Phys. Rev. B **68**, 134205 (2003).
34) B. C. Daly, K. Kang, Y. Wang, and D. G. Cahill, Phys. Rev. B **80**, 174112 (2009).
35) H. Hara and Y. Nishi, J. Phys. Soc. Jpn. **21**, 1222 (1966).