

On the lattice parameter and thermal expansion coefficient of silicon [Comments on the paper "Influence of thermal expansion on the lattice parameter of silicon," F. Liu, Powder Diffr. 9, 260–264 (1994)]

W. Paszkowicz

Institute of Physics, Al.Lotników 32/46, 02-668 Warsaw, Poland

(Received 8 March 1995; accepted for publication
10 April 1995)

The subject of the cited paper are the values of the lattice parameter and thermal expansion coefficient, α , of silicon. Silicon is one of most important reference materials for many applications; therefore, the idea of such review and concluding the "best" values of lattice constant and/or thermal expansion coefficient is very good. However, one can feel some deficiency after reading the paper due to the lack of recent references which are certainly more important than the historical ones. The only two cited recent results are those obtained with the participation of the author of the paper and published earlier in the same journal (Liu and Zheng, 1991; Liu, 1993); some other ones are referenced as unpublished; the choice of all others ends about 1980. We shall briefly justify below the need for a continuation of the cited paper which would include a discussion of the results reported after 1980.

The author indicates one of the lowest experimental α values among the cited ones ($2.45 \times 10^{-6} \text{ K}^{-1}$) as the best one. The reader would be interested in understanding for what physical reasons certain methods give better values than the other ones. The arguments for the given choice considered in the paper are rather of a numerical nature and therefore they are not very convincing. The temperature dependence of the thermal expansion coefficient for silicon is well known. Such dependence has been calculated by Matsuo Kagaya and Soma (1985). An empirical dependence $\alpha(T)$ has been presented by Okada and Tokumaru (1984) on the basis of experiment and numerous literature data. The α value $2.45 \times 10^{-6} \text{ K}^{-1}$ refers to the room temperature which is not well defined and no relation to the temperature dependence of α is discussed. Such an approach may be quite satisfactory in a small temperature range (room temperature range) but at least a comment on such dependence would be required. According to Okada and Tokumaru (1984) the thermal expansion coefficient at room temperature is somewhat higher, about $2.6 \times 10^{-6} \text{ K}^{-1}$, and it varies by about $0.01 \times 10^{-6} \text{ K}^{-1}$ with 1 K temperature change. Among the recent papers, Windisch and Becker (1990) and Fewster and

Andrew (1995) use the α value equal $2.56 \times 10^{-6} \text{ K}^{-1}$ for lattice-parameter correction, while Härtwig *et al.* (1994) use $2.51 \times 10^{-6} \text{ K}^{-1}$. Questions remain about which of all these values is most accurate and could be recommended; in my opinion the paper by Liu (1994) does not fully solve this problem.

What about the value of the lattice parameter of Si, there are some valuable recent works which should be discussed (e.g., Okada and Tokumaru, 1984; Berger, 1984; Windisch and Becker, 1990; Härtwig *et al.*, 1994, Fewster and Andrew, 1995). The fact that the discrepancies between the values of lattice parameters reported by different authors decrease with the progress of experimental techniques is illustrated in Fig. 1, which includes the results cited by Liu (1994) (except the oldest one) completed by several examples of lattice-parameter values from the recent papers cited above. Briefly speaking, the importance of the new achievements consists in:

- (1) working out new experimental methods for measurement of lattice constant of single crystals including mosaic and bent crystals (Berger, 1984; Fewster and Andrew, 1995) (the method of the latter reference has been shown to be applicable also for a powdered silicon specimen);
- (2) investigating the same material in many laboratories, carefully correcting systematic errors in the Bond method (Härtwig *et al.*, 1994), and taking account of the difference between the metric and nonmetric units (Windisch and Becker, 1990; Härtwig *et al.*, 1994);
- (3) investigating silicon single crystals of various impurity level (Okada and Tokumaru 1984; Windisch and Becker, 1990) and exhaustively relating the results to literature data (Okada and Tokumaru 1984); and
- (4) showing by an original method which employs a high resolution diffractometer, that the single crystalline and polycrystalline silicon both have practically the same value of lattice constant, $a_s = a_p$, the difference ($3 \times 10^{-5} \text{ \AA}$) being less than the (small) standard deviation of a_p (Fewster and Andrew, 1995). This observation seems to indicate the source of apparent discrepancy between the larger lattice parameter value of bulk and the smaller one for powdered silicon which could be concluded from experiments performed in various laboratories. (This discrepancy is clearly visible in Fig. 1). Contrary to some opinions (cited also by Liu, 1994), these are therefore most probably the systematic experimental errors in the powder method which cause silicon in the experiments to yield usually a_p lower than a_s .

To complete these remarks we indicate a small error which slipped into the references. Reference 42 cited twice on page 264 is undefined. In addition, we would like to comment on the title of the paper. At the first sight, if one assumes that the expression "thermal expansion" is given in its broad sense (a physical phenomenon), there is no problem. But for a crystallographer the thermal expansion of a crystal may mean a temperature change of lattice parameter, what leads to the following meaning of the title: "Influence of lattice-parameter temperature change on the lattice pa-

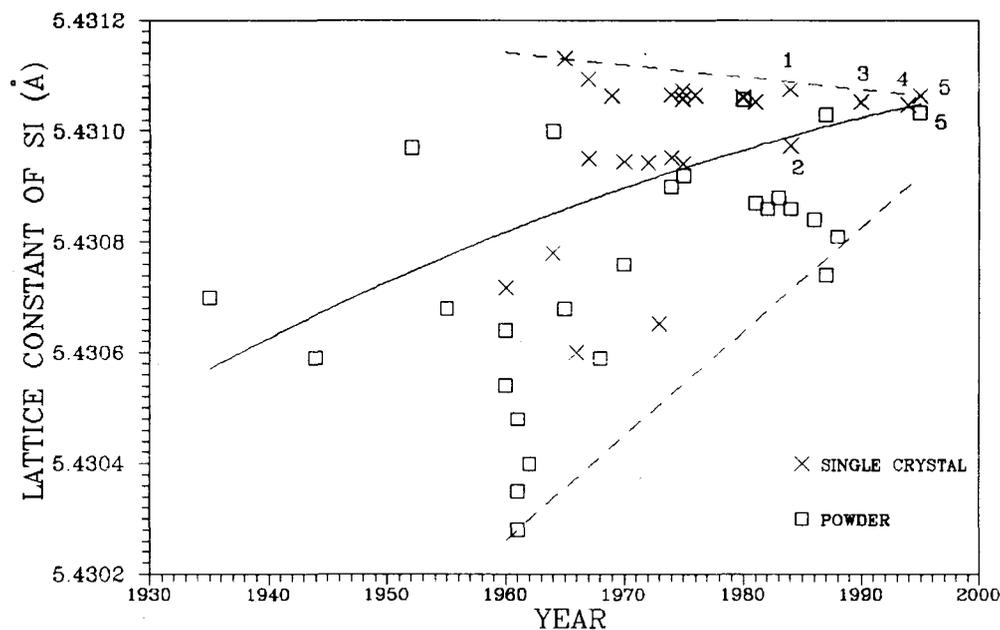


Figure 1. Historical perspective of the lattice parameter of silicon. The figure is founded on values at 25 °C collected in Table II of Liu (1994) completed by the following examples of recent results: (1) Okada and Tokumaru (1984), $\lambda = 1.540581 \text{ \AA}$, $T = 298.5 \text{ °K}$, $a = 5.431074(2) \text{ \AA}$; (2) Berger (1984), $\lambda = 1.540562 \text{ \AA}$, $T = 298.1 \text{ °K}$; $a = 5.430865(19) \text{ \AA}$ [$5.430974(19) \text{ \AA}$ in the metric system]; (3) Windisch and Becker (1990), $\lambda = 1.54059292(45) \text{ \AA}$, $T = 22.50 \text{ °C}$, $a = 5.43102032(34) \text{ \AA}$ (high purity silicon); (4) Härtwig *et al.* (1994), $\lambda = 1.54059292(45) \text{ \AA}$, $T = 20 \text{ °C}$, $a = 5.4309839 \text{ \AA}$ (weighted mean from six laboratories); and (5) Fewster and Andrew (1995), λ of monochromatized Cu $K\alpha_1$, $T = 24 \text{ °C}$, $a = 5.43102(5) \text{ \AA}$ for powder, $5.431051(6) \text{ \AA}$ for a single crystal. The given above original data of references 1–5 were corrected for thermal expansion before plotting. For (2) the metric value is plotted (the problem of conversion to the metric system has been discussed, e.g., by Windisch and Becker, 1990, and Härtwig *et al.*, 1994). The lines are drawn as a guide to the reader's eye.

parameter of silicon" being a tautology. A simpler title, e.g., "Influence of temperature on the lattice parameter of silicon," would be less controversial.

One can doubt whether a further meaningful improvement of the values of lattice constants and thermal expansion coefficients of standard substances can be achieved in a single laboratory. The sources of discrepancies can be identified with the help of tests in which the same material is investigated in many laboratories and/or using various methods (as in the paper by Härtwig *et al.*, 1994), or when various materials are studied by the same experimental technique (as, e.g., in the work by Fewster and Andrew, 1995). It would be particularly interesting if such studies could include determination of lattice parameter and thermal expansion coefficient in a common "room" temperature or in a small temperature interval for high purity silicon powder supplied by various manufacturers, in order to determine similarities and differences of potential standard materials and to better characterize and/or improve the applied experimental methods.

Berger, H. (1984). "A Method for Precision Lattice-Parameter Measurement of Single Crystals," *J. Appl. Cryst.* **17**, 451–455.

Fewster, P. F., and Andrew, N. I. (1995). "Accurate Lattice Parameter Measurement" (to be published).

Härtwig, J., Bąk-Misiuk, J., Berger, H., Brühl, H.-G., Okada, Y., Gross-wig, S., Wokulska, K., and Wolf, J. (1994). "Comparison of Lattice Parameters Obtained from an Internal Silicon Monocrystal Standard," *Phys. Stat. Sol. A* **142**, 19–26.

Liu, F., and Zheng, B. (1991). "The Linear Coefficient of Thermal Expansion of Silicon at Room Temperature," *Powder Diffr.* **6**, 147–152.

Liu, F. (1993). "Confirmation of the New Technique for Measuring the Linear Thermal Expansion of Silicon," *Powder Diffr.* **8**, 36–38.

Liu, F. (1994). "Influence of Thermal Expansion on the Lattice Parameter of Silicon," *Powder Diffr.* **9**, 260–264.

Matsuo Kagaya, H., and Soma, T. (1985). "Temperature Dependence of the Linear Thermal Expansion Coefficient for Si and Ge," *Phys. Stat. Sol. B* **129**, K5–K8.

Okada, Y., and Tokumaru, Y. (1984). "Precise Determination of Lattice Parameter and Thermal Expansion Coefficient of Silicon between 300 and 1500 K," *J. Appl. Phys.* **56** (2), 314–320.

Windisch, D., and Becker, P. (1990). "Silicon Lattice Parameters as an Absolute Scale of Length for High Precision Measurements of Fundamental Constants," *Phys. Stat. Sol. A* **118**, 379–388.