THERMAL EXPANSION OF ALPHA QUARTZ

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Abstract

Existing data for thermal expansion of alpha quartz (between -50° C and 150° C) have been critically analyzed through a program funded by ETDL. A recommended "best" set of values was received as were third-, fourth-, and fifth-order power series expansions for the coefficients of thermal linear expansion (CTE), referenced to 0° C. In order to fully utilize the results, relationships between the CTEs and the thermal expansion coefficients (α_{ij}) were derived and α_{ij} referenced to 25° C were obtained. Based on the results, an additional analysis to third order in α_{ij} was performed. The new α_{ij} values allow direct comparison with previously published thermal expansion coefficients. The influence of the new α_{ij} on determinations of quartz material temperature coefficients and on the calculation of temperature coefficients of frequency for the case of the AT-cut are discussed.

Introduction

There is an internationally recognized interest in obtaining more reliable alpha-quartz material constants [1,2]. This interest also is recognized, e.g., by ongoing projects sanctioned by the Electronic Industries Association (EIA) [3], which have highlighted the need to examine all of the quartz material constants.

The thermal expansion coefficients $\alpha_{ij}^{(n)}$, also known as the thermoelastic constants, are critical parameters in piezoelectric crystal resonator and filter calculations. For example, the first order temperature coefficient of frequency of the simple thickness modes of a crystal plate can be shown to be of the form [4]:

$$2T_f^{(1)} = T_{\bar{c}}^{(1)} - T_\rho^{(1)} - 2T_h^{(1)} \qquad (1)$$

where f= frequency, $\bar{c}$= piezoelectrically stiffened elastic constant, ρ = mass density, and h=thickness. The quantities $T_\rho^{(1)}$ and $T_h^{(1)}$ depend directly on thermal expansion coefficients and are given by:

$$T_\rho^{(1)} = -(\alpha_{11}^{(1)} + \alpha_{22}^{(1)} + \alpha_{33}^{(1)}) \qquad (2)$$

and

$$T_h^{(1)} = +\alpha_{22}^{\prime(1)} \qquad (3)$$

where the $\alpha_{ij}^{(1)}$ are thermal expansion constants and $\alpha_{22}^{\prime(1)}$ is the thermal expansion constant rotated to the crystallographic direction of the plate thickness, which is taken as the x_2-direction. The $\alpha_{ij}^{(n)}$ are components of tensors of the second rank.

At present there are three sets of thermal expansion coefficients in general use: the Bechmann, Ballato, and Lukaszek (BBL) set [4]; the American Institute of Physics (AIP) set [5] ; and the Mizan and Ballato (MB) set [6]. The MB set is based on a digitization of a graph published by Sosman [7]. Brice [8] and James [9] have also contributed alternative data sets for alpha quartz thermal expansion. In Fig. 1 it is shown that the thermal expansion coefficients in general use do not agree with one another over the temperature range -50° C to + 150° C.

Experimental Data

Funding and time constraints did not allow the gathering of new experimental data. Instead, a literature search and critical analysis of published data were performed. This phase was accomplished by the Purdue University - Center for Information and Numerical Data Analysis and Synthesis (CINDAS). CINDAS operates the High Temperature Materials - Mechanical, Electronic and Thermophysical Properties Information Analysis Center (HTMIAC) for the Department of Defense [10].

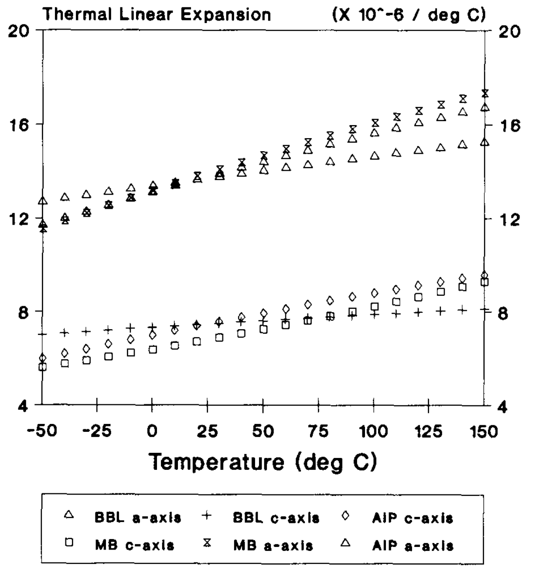

Fig. 1. Thermal expansion calculations using the thermal expansion coefficients of BBL [4], AIP [5], and MB [6]. There is little agreement for the temperature range -50° C to 150° C.

The HTMIAC analysis involved searching, compiling, and critically analyzing existing thermal expansion data. The HTMIAC recommended values for the coefficient of thermal linear expansion (CTE) along the a-axis were obtained by fitting to the data of White [11] and Barron [12], in conjunction with the data of Jay [13], Johnson and Parsons [14], and Lager et al. [15]. Data of Dorsey [16] and Buffington and Latimer [17] also were included. The recommended values for CTE along the c-axis were obtained from Refs. [11-15] and also included the data of Nix and McNair [18].

The recommended coefficient values for CTE were fitted to 3rd-, 4th-, and 5th-order power series of temperature referenced to 0° C and valid for the temperature range -50° C to +150° C. The CTE curve fit coefficients are listed in Table I. See Fig. 2 for a plot of the fit to a 5th-order power series.

As shown in Figs. 3-5, none of the thermal expansion coefficient sets in general use agree well with the HTMIAC recommended data.

ETDL Analysis

Unfortunately, the CTE used by HTMIAC are not equivalent to the thermal expansion coefficients, $\alpha_{ij}^{(n)}$, that are normally used in frequency control R&D. Specifically,

$$CTE \equiv 1/h(\theta_o)\ \partial h/\partial\theta \quad (4)$$

with reference temperature θ_o=0° C, whereas the more usual thermal expansion coefficients, $\alpha_{ij}^{(n)}$, are obtained from a Taylor series expansion of length or thickness (h) about a reference temperature θ_o=25° C :

$$h(\theta)=h(\theta_o)\sum_{n=0}^{m}1/n!\ 1/h(\theta_o)\ \partial^n h/\partial\theta^n\ (\theta-\theta_o)^n \quad (5)$$

whence

$$\alpha_{ij}^{(n)} = T_h^{(n)} \equiv 1/n!\ 1/h(\theta_o)\ [\partial^n h/\partial\theta^n]_{\theta_o}. \quad (6)$$

In the past, most analyses have been limited to order m = 3.

Mathematical relationships between the CTE and the $\alpha_{ij}^{(n)}$ were derived, accounting also for the different reference temperature. The HTMIAC expansion of (4) can be written as

$$CTE = \sum_{n=0}^{m} a_n\theta^n. \quad (7)$$

We can then write h(0) in terms of the $\alpha_{ij}^{(n)}$ and $h(\theta_o)$ as

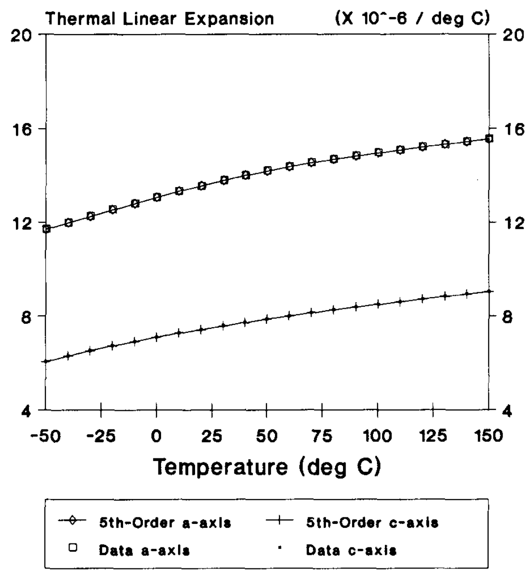

Fig. 2. A 5th-order power series least-squares fit to the recommended HTMIAC data for the coefficient of thermal linear expansion.

$$h_o = h(0) = h(\theta_o)\left[\sum_{n=o}^{m} \alpha_{ij}^{(n)} (-\theta_o)^n\right] \qquad (8)$$

where $\theta_o = 25° C$.

Table I

HTMIAC CTE POWER SERIES COEFFICIENTS

a-axis

Order of Fit

Coeff.	5	4	3	Units
a_0	13.1	13.1	13.1	$X\ 10^{-6}/K$
a_1	26.1	25.8	25.0	$X\ 10^{-9}/K^2$
a_2	-60.6	-54.3	-64.7	$X\ 10^{-12}/K^3$
a_3	-466.2	-320.3	45.1	$X\ 10^{-15}/K^4$
a_4	4645	1827		$X\ 10^{-18}/K^5$
a_5	-11270			$X\ 10^{-21}/K^6$
c-axis				
a_0	7.1	7.1	7.1	$X\ 10^{-6}/K$
a_1	17.2	17.5	17.7	$X\ 10^{-9}/K^2$
a_2	-45.5	-50.9	-48.4	$X\ 10^{-12}/K^3$
a_3	323.0	-200.0	111.7	$X\ 10^{-15}/K^4$
a_4	-2817	-441.6		$X\ 10^{-18}/K^5$
a_5	9503			$X\ 10^{-21}/K^6$

From (5) and (6) we find that

$$\partial h(\theta)/\partial\theta = h(\theta_o)\left[\sum_{n=o}^{m} n\alpha_{ij}^{(n)} (\theta-\theta_o)^{n-1}\right] \qquad (9)$$

thus

$$CTE = 1/h_o\ \partial h(\theta)/\partial\theta \approx \sum_{n=o}^{m} n\alpha_{ij}^{(n)} (\theta-\theta_o)^{n-1}, \qquad (10)$$

since the summation in (8) is ≈ 1. Therefore, we find

$$CTE = \sum_{n=o}^{m} a_n\theta^n = \sum_{n=o}^{m} n\alpha_{ij}^{(n)} (\theta-\theta_o)^{n-1}. \qquad (11)$$

Expanding both summations and equating coefficients of equal powers of θ, one obtains (after some manipulation) the

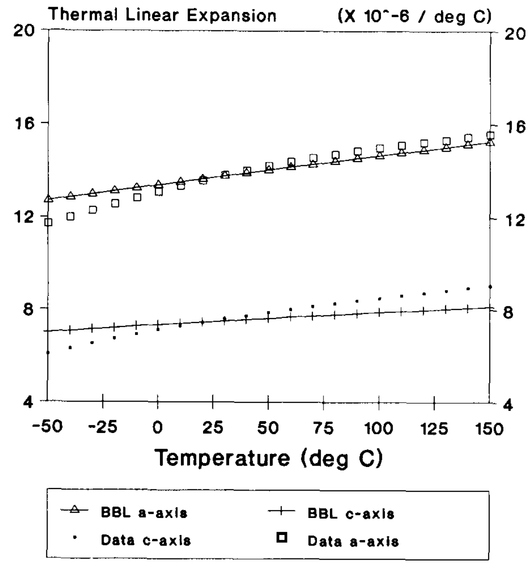

Fig. 3. Thermal expansion calculated using the thermal expansion coefficients of BBL [4] vs the HTMIAC data.

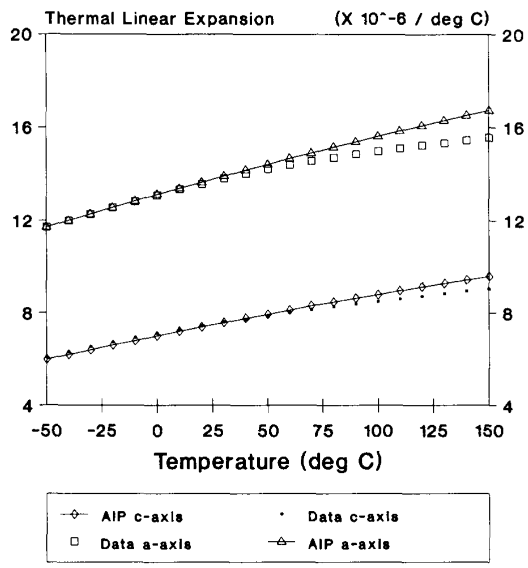

Fig. 4. Thermal expansion calculated using the thermal expansion coefficients of AIP [5] vs the HTMIAC data.

following thermal expansion coefficients up to order six in terms of the CTE power series coefficients:

$$\begin{aligned}
\alpha_{ij}^{(1)} &= a_0 + a_1\theta_0 + a_2\theta_0^2 + a_3\theta_0^3 + a_4\theta_0^4 + a_5\theta_0^5 \\
\alpha_{ij}^{(2)} &= 1/2[a_1 + 2a_2\theta_0 + 3a_3\theta_0^2 + 4a_4\theta_0^3 + 5a_5\theta_0^4] \\
\alpha_{ij}^{(3)} &= 1/3[a_2 + 3a_3\theta_0 + 6a_4\theta_0^2 + 10a_5\theta_0^3] \\
\alpha_{ij}^{(4)} &= 1/4[a_3 + 4a_4\theta_0 + 10a_5\theta_0^2] \\
\alpha_{ij}^{(5)} &= 1/5[a_4 + 5a_5\theta_0] \\
\alpha_{ij}^{(6)} &= 1/6[a_5].
\end{aligned} \qquad (12)$$

Note that an analysis of CTE $_{(n)}$ to order m corresponds to an analysis of α_{ij} to order m+1.

The HTMIAC results recast in thermal expansion form are listed in Table II. Most calculations of quartz resonator temperature behavior are carried out only to third order. If any one of the HTMIAC analyses is truncated to third order, the calculated results do not reproduce the HTMIAC data (see, e.g., Fig. 6). Because of this, a least-squares fit to the HTMIAC data to third order in α_{ij} was developed. This fit is called KGB and is plotted in Fig. 7. The values up to third order are listed together with BBL, AIP, and MB in Table III.

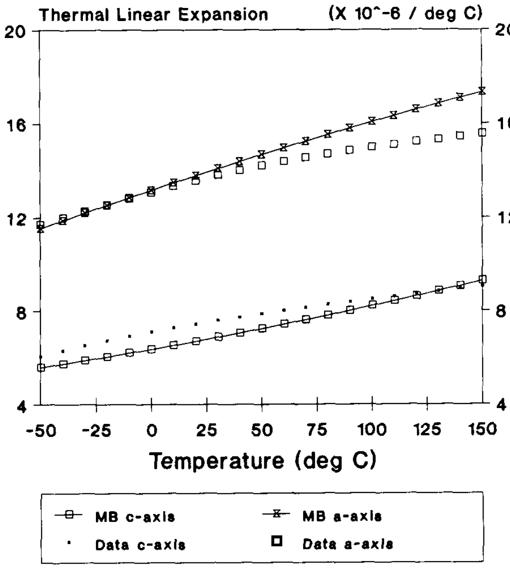

Fig. 5. Thermal expansion calculated using the thermal expansion coefficients of MB [6] vs the HTMIAC data.

Discussion

To understand the impact of using the KGB $\alpha_{ij}^{(n)}$ instead of, for example, using the BBL $\alpha_{ij}^{(n)}$, we have investigated the determination of material-constant

Table II

HTMIAC RESULTS IN THERMAL EXPANSION FORMAT

$\alpha_{11}^{(n)}$

(n)	Order of Fit: 6	5	4	Units
1	13.673	13.668	13.659	X 10^{-6}/K
2	11.231	11.299	10.926	X 10^{-9}/K^2
3	-26.646	-23.834	-20.422	X 10^{-12}/K^3
4	-18.034	-34.400	11.278	X 10^{-15}/K^4
5	647.250	365.400		X 10^{-18}/K^5
6	-1878.333			X 10^{-21}/K^6

$\alpha_{33}^{(n)}$

(n)	6	5	4	Units
1	7.508	7.513	7.515	X 10^{-6}/K
2	7.686	7.653	7.746	X 10^{-9}/K^2
3	-10.131	-12.502	-13.328	X 10^{-12}/K^3
4	25.173	38.960	27.925	X 10^{-15}/K^4
5	-325.825	-88.320		X 10^{-18}/K^5
6	1583.833			X 10^{-21}/K^6

Table III

THIRD ORDER THERMAL EXPANSION

$\alpha_{11}^{(n)}$

(n)	BBL	AIP	MB	KGB	Units
1	13.71	13.77	13.92	13.65	X 10^{-6}/K
2	6.50	13.03	15.09	11.02	X 10^{-9}/K^2
3	-1.90	-6.33	-7.86	-19.32	X 10^{-12}/K^3

$\alpha_{33}^{(n)}$

(n)	BBL	AIP	MB	KGB	Units
1	7.48	7.48	6.79	7.50	X 10^{-6}/K
2	2.90	9.41	8.69	8.00	X 10^{-9}/K^2
3	-1.50	-5.44	6.88	-10.44	X 10^{-12}/K^3

temperature coefficients (obtained by the resonator method) as well as the calculation of frequency-temperature behavior from the material constants.

The use of KGB thermal expansion coefficients instead of BBL coefficients will result in nontrivial differences when extracting temperature coefficients of the piezoelectric and dielectric constants from measured resonator frequency-temperature behavior. Values of the temperature coefficients of selected elastic constants for rotated Y-cuts extracted from measurements on rotated Y-cut resonators are given in Table IV. The previously reported [3] values using BBL thermal expansion coefficients may be compared to the values obtained using KGB thermal expansion coefficients.

The influence of the $\alpha_{ij}^{(n)}$ on the calculation of frequency-temperature behavior has been examined by using (1) in conjunction with the BBL set of material constants (with the exception of the BBL $\alpha_{ij}^{(n)}$). It is best to write (1) in the form [19]:

$$2\hat{T}_f^{(n)} = \hat{T}_{\bar{c}}^{(n)} - \hat{T}_\rho^{(n)} - 2\hat{T}_h^{(n)} \qquad (13)$$

where

$$\hat{T}^{(1)} = T^{(1)}$$
$$\hat{T}^{(2)} = T^{(2)} - 1/2(T^{(1)})^2 \qquad (14)$$
$$\hat{T}^{(3)} = T^{(3)} - T^{(2)}\,T^{(1)} + 1/3(T^{(1)})^3.$$

If we then assume $T_{\bar{c}B}^{(n)} = T_{\bar{c}K}^{(n)}$, where B=BBL and K=KGB, then

$$[\hat{T}_{fB}^{(n)} - \hat{T}_{fK}^{(n)}] = 1/2\ \{[\hat{T}_{\rho K}^{(n)} - \hat{T}_{\rho B}^{(n)}] + 2[\hat{T}_{hK}^{(n)} - \hat{T}_{hB}^{(n)}]\}. \qquad (15)$$

The $\alpha_{ij}^{(n)}$ values found in Table III together with

$$T_\rho^{(n)} = -(2\alpha_{11}^{(n)} + \alpha_{33}^{(n)}) \qquad (16)$$

allow the calculation of temperature coefficients of density. These coefficients are given in Table V.

Similarly, the temperature coefficients of thickness for rotated Y-cut resonators may be calculated using the equation

$$T_h^{(n)} = \alpha_{11}^{(n)} \cos^2\theta + \alpha_{33}^{(n)} \sin^2\theta \qquad (17)$$

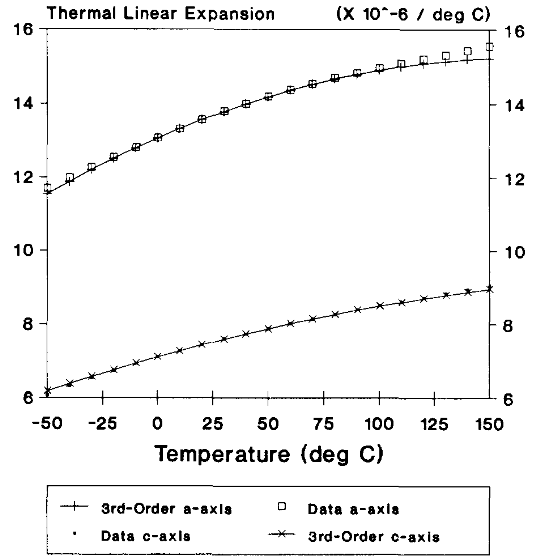

Fig. 6. Calculation of thermal expansion using 6th-order thermal expansion coefficients (truncated to 3rd-order) vs the HTMIAC data.

Table IV

TEMPERATURE COEFFICIENTS OF ELASTIC CONSTANTS ($T_{c_{ij}^{E}}^{(n)}$)

(n) \ ij ->	66	44	14	Units
		BBL		
1	176.9	-170.4	103.9	$\times 10^{-6}/K$
2	163.8	-252.5	-19.4	$\times 10^{-9}/K^2$
3	43.9	-201.1	-509.1	$\times 10^{-12}/K^3$
		KGB		
1	176.9	-170.3	103.9	$\times 10^{-6}/K$
2	158.7	-256.6	-23.9	$\times 10^{-9}/K^2$
3	51.6	-177.3	-492.8	$\times 10^{-12}/K^3$

The temperature coefficients of thickness for AT-cut and BT-cut resonators are given in Table VI:

Table V

TEMPERATURE COEFFICIENTS OF DENSITY

(n)	KGB $T_\rho^{(n)}$	BBL $T_\rho^{(n)}$	KGB $\hat{T}_\rho^{(n)}$	BBL $\hat{T}_\rho^{(n)}$	Units
1	-34.80	-34.90	-34.80	-34.90	X 10^{-6}/K
2	-30.04	-15.90	-30.65	-16.51	X $10^{-9}/K^2$
3	49.08	5.30	48.02	4.73	X $10^{-12}/K^3$

Table VI

TEMPERATURE COEFFICIENTS OF THICKNESS

AT-Cut

(n)	KGB $T_h^{(n)}$	BBL $T_h^{(n)}$	KGB $\hat{T}_h^{(n)}$	BBL $\hat{T}_h^{(n)}$	Units
1	11.60	11.63	11.60	11.63	X 10^{-6}/K
2	10.01	5.30	9.94	5.23	X $10^{-9}/K^2$
3	-16.36	-1.77	-16.48	-1.83	X $10^{-12}/K^3$

BT-Cut

(n)	KGB $T_h^{(n)}$	BBL $T_h^{(n)}$	KGB $\hat{T}_h^{(n)}$	BBL $\hat{T}_h^{(n)}$	Units
1	10.13	10.14	10.13	10.14	X 10^{-6}/K
2	9.29	4.44	9.24	4.39	X $10^{-9}/K^2$
3	-14.23	-1.67	-14.32	-1.71	X $10^{-12}/K^3$

The differences in the temperature coefficients of frequency, density, and thickness obtained from (15) are listed in Table VII. A number of observations may be made. First, the differences in predicted first order temperature coefficient of frequency are very small. For example, from Bechmann [20] we know that:

AT-cut $\partial T_{fR}^{(1)}/\partial\theta = -5.15 \times 10^{-6}/K,°\theta$

and for the

BT-cut $\partial T_{fR}^{(1)}/\partial\theta = 2.14 \times 10^{-6}/K,°\theta$

Thus, the difference between the predicted AT-cut frequency-temperature behaviors is equivalent to an angular error of -14" of arc and the BT-cut difference is equivalent to an angular error of 1'07" of arc. Second, the differences in higher order temperature coefficients of frequency are values of the differences in temperature coefficients of frequency for the AT-cut with the actual magnitudes of these coefficients as published by Ballato [21]:

AT-cut $T_{fA}^{(2)} = -0.45 \times 10^{-9}$ $T_{fA}^{(3)} = 108.6 \times 10^{-12}$

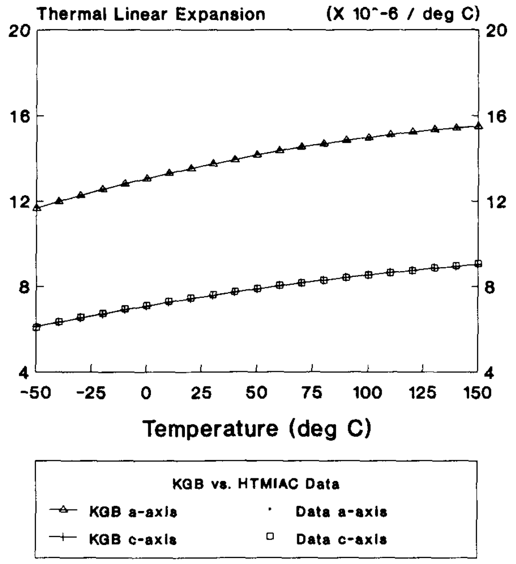

Fig. 7. Third-order thermal expansion coefficient least-squares fit to the HTMIAC data.

Table VII

DIFFERENCES IN TEMPERATURE COEFFICIENTS OF FREQUENCY, DENSITY, AND THICKNESS

(n)	$[\hat{T}_{fB}^{(n)} - \hat{T}_{fK}^{(n)}]$ AT	$[\hat{T}_{fB}^{(n)} - \hat{T}_{fK}^{(n)}]$ BT	$[\hat{T}_{\rho K}^{(n)} - \hat{T}_{\rho B}^{(n)}]$	$[\hat{T}_{hK}^{(n)} - \hat{T}_{hB}^{(n)}]$ AT	$[\hat{T}_{hK}^{(n)} - \hat{T}_{hB}^{(n)}]$ BT
1	0.02	0.04	0.10	-0.03	-0.01
2	-2.36	-2.22	-14.14	4.71	4.85
3	7.00	9.04	43.29	-14.65	12.61

Note: The usual units apply.

Conclusions

An analysis of existing experimental thermal expansion data for quartz has been performed, yielding an improved, average, set of higher order thermal expansion coefficients α_{11} and α_{33}. Comparisons with previously used sets are given. The new values are required for redeterminations of the temperature coefficients of the elastic, piezoelectric, and dielectric constants of this important crystalline material.

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