U. S. DEPARTMENT OF COMMERCE NATIONAL BUREAU OF STANDARDS

RESEARCH PAPER RP784

Part of Journal of Research of the National Bureau of Standards, Volume 14, May 1935

THERMAL EXPANSION OF MONOCRYSTALLINE AND POLYCRYSTALLINE ANTIMONY

By Peter Hidnert

ABSTRACT

Measurements were made on the linear thermal expansion of eleven samples of single crystals of antimony and three samples of polycrystalline antimony at various temperatures between 20 and 560° C, and the data were correlated with the results obtained by previous investigators to 300° C. The linear expansion depends upon the direction along which the measurements are made. For example, the linear expansion along the trigonal axis (0° orientation) of a single crystal is about twice as large as the expansion along a direction perpendicular to this axis (90° orientation). Equations were derived which show the relationships between the coefficients of expansion and the orientations of single crystals.

The linear thermal-expansion curves of polycrystalline antimony show that there is no polymorphic transition between 20 and 560° C. The differences in the linear expansion of different samples of polycrystalline antimony are attributed to variations in the average orientation of the crystals. Figure 13 shows a comparison of the linear thermal expansion of monocrystalline and polycrystalline antimony.

CONTENTS

I. INTRODUCTION

It has been pointed out by Carpenter[1] that the scientific investigation of metals in the future will have to take into account both the monocrystalline and the polycrystalline states, and the variations in any particular property which may be secured by varying the orientation of the crystal. The properties of a specimen of polycrystalline metal depend upon the extent of crystallization, the nature of the crystals, their magnitude and their orientation, as well as upon the admixture of foreign crystals and the presence of impurities. Such knowledge should form the basis of the scientific manufacture of metals and alloys possessing specified properties. Promising applications have already appeared in the field of metallurgy.

---

[1] Carpenter, Nature, **126**, 17 (1930).

Antimony[2] crystallizes in the rhombohedral form of the hexagonal system. It is one of the metals that is anisotropic and therefore some of its properties, such as thermal expansion, depend on the orientation of the crystals. Data on the thermal expansion of single crystals of antimony are fundamental, for they are necessary in order to understand the behavior of this metal in the polycrystalline state.

Fizeau in 1869, and Bridgman in 1925, reported data on the linear thermal expansion of monocrystalline antimony for temperature ranges between 0 and 100° C. Between 1754 and 1910 a number of investigations were made on the thermal expansion of polycrystalline antimony for various temperature ranges between −190 and +100° C. In 1920 Braesco reported coefficients of linear expansion of polycrystalline antimony between 100 and 300° C. No data have been found in the literature for higher temperatures.[3] Tables 1 and 2 give summaries of available data by previous observers on the thermal expansion of monocrystalline and polycrystalline antimony.

TABLE 1.—*Summary of expansion data by previous observers on monocrystalline antimony*

Observer	Date	Reference	Material	Temperature or temperature range	Coefficient of linear expansion per degree centigrade
				°C	×10⁻⁶
Fizeau	1869	Compt.rend. **68**, 1125 (1869), or Ann.Phys. **138**, 26 (1869).	Antimony crystal:		
			Parallel to axis	40	16.9
				0 to 100	16.8
			Perpendicular to axis	40	8.8
				0 to 100	9.0
Bridgman	1925	Proc.Am.Acad. Arts Sci. **60**, 305 (1924–25). See also Proc. Acad.Sci. **10**, 411 (1924).	Antimony, in single grain casting:		
			Parallel to axis [1]	15 to 25	15.6
			Perpendicular to axis [1]	15 to 25	8.0

[1] Specimen cut from a large casting.

The present investigation was undertaken in order to extend our knowledge of the linear thermal expansion of monocrystalline and polycrystalline antimony above these temperatures, and to determine the effect of orientation on thermal expansion. Eleven samples of single crystals of antimony and three samples of polycrystalline antimony were investigated at various temperatures between 20 and 560° C.

---

[2] The principal uses of antimony have been given by Chung Yu Wang in a book entitled Antimony (C. Griffin & Co., London, 1909).

[3] Roeser, Schofield, and Moser, Melting point of antimony, 630.5° C. BS J. Research **11**, 1 (1933) RP**573**.

TABLE 2.—*Summary of expansion data by previous observers on polycrystalline antimony*

Observer	Date	Reference	Material	Temperature or temperature range	Coefficient of linear expansion per degree centigrade	Remarks
				°C	$\times 10^{-6}$	
Smeaton	1754	Phil.Trans. **48,** part 2, 598 (1754)	Antimony	0 to 100	10.8	Computed from Smeaton's data.
Kopp	1852	Ann.Chem.Pharm. 81, 1 (1852)	do	12 to 42		Average coefficient of cubical expansion 33×10^{-6} per degree C, obtained by density method.
Calvert, Johnson and Lowe.	1859	Rept.Brit.Assoc. **28,** 46 (1859); Proc. Roy.Soc. (London) **10,** 315 (1859-60); Chem.News **3,** 315, 357, 371 (1861).	Antimony, cast	0 to 100	9.8	
Matthiessen	1866	Phil.Trans. **156,** 861 (1866) or Ann. Phys. **130,** 50 (1867).	Antimony, gilded [1]	0 to 100		Average coefficient of cubical expansion 31.7×10^{-6} per degree C, obtained by weighing two samples in water at different temperatures.
Le Chatelier	1899	Compt.rend. **128,** 1444 (1899)	Antimony	63	10.	
Dorsey	1907	Phys.Rev. **25,** 88 (1907)	Antimony, cast (density= 6.88).	−170	7.5	These coefficients were obtained by Dorsey from his data on cooling and warming the specimen several times. The specimen expanded more on warming than it contracted in cooling.
				−150	7.9	
				−130	8.4	
				−110	8.9	
				−90	9.1	
				−70	9.3	
				−50	9.6	
				−30	9.7	
				−10	9.9	
				+10	10.1	
				−180 to +20	8.2	
Lussana	1910	Nuovo cimento [5] **19,** 182 (1910)	Antimony [2] (from Merck) fused in cylinder.	9 to 58 9 to 72		Between 9 and 58° C, obtained following average coefficients of cubical expansion: 34.4×10^{-6}, 32.7×10^{-6} and 31.7×10^{-6} at 1, 1,000, and 1,500 atmospheres, pressure, respectively. Similarly for the range between 9 and 72° C. obtained the following average coefficients of cubical expansion: 35.3×10^{-6}, 32.9×10^{-6}, and 31.7×10^{-6}.
Grüneisen	1910	Ann.Phys. [4] **33,** 33, 65 (1910)	Antimony (from C. A. F. Kahlbaum).	−190 to +17	10.2	
				+17 to 100	10.9	
				18	10.7	
Braesco	1920	Compt. rend. **170,** 103 (1920) and Ann. Phys. **14,** 6 (1920).	Antimony, agglomerated [3] and quenched.	100 to 300	8.3	In the second publication, Braesco states the coefficients of expansion apply at 100° C.
			Antimony, agglomerated [3] and annealed.	100 to 300	8.3	
			Antimony, cast and quenched.	100 to 300	10.0	
			Antimony, cast and annealed.	100 to 300	10.0	
Jubitz	1926	Z.tech.Phys. **7,** 522 (1926)	Antimony, cast	20 to 40	10.1	Computed from data of observer.
			Antimony, drawn	20 to 40	8.8	

---

[1] The samples were gilded to prevent action of water.

[2] Specific gravity 6.6848 at 9.1° C referred to water at 4° C as unity.

[3] Powdered antimony cemented with a small amount of silicate of soda.

II. MONOCRYSTALLINE ANTIMONY

1. MATERIALS INVESTIGATED

Eleven samples of single crystals of antimony prepared by Prof. P. W. Bridgman of Harvard University, were used in this investigation. These were prepared by slow solidification from the melt. This method[4] of growing single crystals was described[5] by him about ten years ago.

These samples cover a range of orientation[6] from 32° to 90°. Most of the orientations are between 60° and 90°, for the preferred manner of growth is with the trigonal[7] axis perpendicular to the axis of the rod (90° orientation). The author made a number of attempts to obtain a few antimony crystals having orientations between 0° and 32°, but he was unsuccessful in obtaining any of these crystals from various laboratories in this country or the National Physical Laboratory in England. Mildred Allen[8] also experienced difficulty in obtaining single antimony crystals having small orientations for an investigation on the effect of tension on the electrical resistance. She obtained only two readings for orientations less than 50° and stated "This came about for two reasons: it is comparatively difficult to grow crystals with the principal cleavage plane nearly perpendicular to the cylindrical length, and, more important, crystals with such orientations are so exceedingly brittle that it is very nearly impossible to mount them in the apparatus without sufficient jarring to break them along a principal cleavage."

Goetz and Hasler[9] observed that the probable orientation of bismuth single crystals, which are similar to antimony crystals, was between 30° and 90°, the orientation of 0° never occurring at all. They state that one influence can be considered as resident in the strain applied to the crystal during the act of crystallization by the glass walls, and due to the expansion of the metal at the melting point. "Hence, the volume of crystallizing region reaches a maximum and then decreases due to the thermal contraction during the cooling process. From the observations by the authors, it is probable that the orientation of the crystal becomes rigidly determined at the instant when a decided contraction appears, preceded by the above expansion indicating that the properties of the crystal (orientation, perfection, etc.) are determined by the forces acting at the time of this contraction to a large extent. This may be observed in certain cases by watching an upright glass tube, filled with molten bismuth, as the crystal grows upward from a cooled tip. The crystallization is seen to take place in blocks which contract visibly from the glass at the instant of formation. If one considers that the stresses, induced by previous expansion, are present at the moment of contraction, then the difficulty of obtaining crystals with small orientations may be

---

[4] There are four methods of growing single crystals, (1) from the vapor, (2) from the melt, (3) from solution, and (4) from the solid.

[5] Bridgman, Proc. Am. Acad. Arts Sci. **60,** 305 (1924–25).

[6] The term orientation is defined as the angle between the trigonal axis of the single crystal and the geometric axis of the rod along which the expansion was measured.

[7] The lattice structure (determined by James and Tunstall, Phil. Mag. **40,** 233 (1920) and Ogg, Phil. Mag. **42,** 163, (1921)) of antimony is a lattice of trigonal symmetry composed of two similar face-centered rhombohedral lattices, similarly oriented, displaced relative to each other along the longest diagonal of the rhombohedron (the axis of trigonal symmetry). The unit rhomb contains 8 atoms and the length of the edge of the unit rhomb is 6.20×10^{-8} cm.

[8] Mildred Allen, Phys. Rev. **43,** 569 (1933).

[9] Goetz and Hasler, Proc. Nat. Acad. Sci. **15,** 646 (1929).

explained as follows. As it is known that the coefficient of thermal expansion is largest parallel to the trigonal axis and smallest normal to it, it could be expected that the crystal prefers an orientation in the glass tube where the maximum contraction would be parallel to the direction of maximum compression. Thus large angle-orientations are preferred."

The direction of the trigonal axis of each antimony single crystal was most conveniently obtained from the principal cleavage plane, which is perpendicular to the axis. The principal cleavage plane may be distinguished from the secondary cleavage planes, because the cleavage on the principal plane is easier, more extensive, and noticeably more nearly perfect than on the secondary planes.

The angle between the principal cleavage plane and the axis of each rod was measured,[10] and from this measurement the orientation, or the angle between the trigonal axis of the crystal and the axis of the rod, was computed. The cleavage planes and the orientations were checked by Dr. S. B. Hendricks of the Bureau of Chemistry and Soils of the U. S. Department of Agriculture. The identification of the cleavage planes of samples 1524, 1527, and 1529 (see table 3) was ascertained by him by reflection of Cu Kα radiation from the perfect cleavage. "The angle of reflection was 11°52′, corresponding to the required angle for (111), referred to rhombohedral axes." He measured the spacing of the imperfect cleavage plane for sample 1532 and found it to correspond to that required for (110). Dr. Hendricks identified the cleavage planes of the other samples by inspection.

TABLE 3.—*Length and orientation of single crystals of antimony*

Sample	Length	Orientation	Sample	Length	Orientation
	Millimeters	*Degrees*		*Millimeters*	*Degrees*
1524	52.1	72	1530	59.0	77
1525	54.8	69	1531	67.6	74
1526	69.0	63	1532	33.7	90
1527	40.5	73	1553	20.6	90
1528	36.7	32	1553A	20.9	90
1529	32.8	32			

Table 3 indicates the length and the orientation of each single crystal. All samples except 1553 and 1553A have a circular cross section with the diameter about 3.5 mm. Samples 1553 and 1553A were cut from a piece[11] of a large single crystal (about 22-mm diameter) made a number of years before the other samples were prepared. The cross sections of samples 1553 and 1553A are nearly square, with the edge of the square about 3 mm.

The single crystals of antimony were made from "Kahlbaum grade" antimony purchased by Prof. Bridgman from Akatos, Inc., in New York City. Samples 1524 to 1532, inclusive, were made from antimony obtained in 1932, and samples 1553 and 1553A were prepared from antimony purchased about 1923. No chemical analyses were made.

As the single crystals are very fragile, Prof. Bridgman sent them to the National Bureau of Standards in a tube filled with venice

---

[10] By D. R. Miller of this Bureau.

[11] It was not possible to cut a sample with an orientation of 0° from this piece, for the layers of metal separated on cutting.

turpentine. The entire tube in which they were contained was heated on a water bath until the venice turpentine was sufficiently fluid to allow the removal of the crystals. Before expansion determinations were started on each sample, it was soaked in acetone several days, washed with a soap solution, then water, and finally alcohol, in order to remove any deposit of venice turpentine.

2. APPARATUS

The fused-quartz-tube expansion apparatus described by Hidnert and Sweeney [12] was modified so that the short single crystals of different lengths (21 to 69 mm) could be used without cutting the fragile samples to the same length. A 45-cm movable fused-quartz rod was used for all samples except 1526 and 1531. For these two samples, a 41-cm movable fused-quartz rod was employed. These movable rods are hollow, but are closed at the ends. The bottom of each movable rod is concave and the top is flat.

Two collars (made from a heat-resisting alloy and a ceramic material) and a porcelain tube supported the sample centrally in a vertical fused-quartz tube closed at the lower end, which was ground concave. A movable fused-quartz rod (41 or 45 cm) rested on top of the sample and extended above the open end of the tube. An indicator gage graduated to 0.0001 inch, rested on top of the movable fused-quartz rod. A chromel-alumel thermocouple placed inside the fused-quartz tube near the center of the sample indicated the temperature. An electric furnace surrounding the tube containing the sample was used for heating.

The indicator gage fastened near the top of the tube registered the differential expansion between the sample and an equivalent length of fused quartz. A correction for the small expansion [13] of fused quartz was made.

Figures 1 and 2 of BS Research Paper RP29 show photographs of the fused-quartz-tube apparatus and auxiliary equipment. Blueprints of the apparatus are available for persons interested in constructing this equipment.

3. RESULTS

Measurements were made on the linear thermal expansion of the single crystals of antimony at various temperatures between 20° and 500° C. Two or three tests were made on each sample. The results obtained on heating and cooling are shown in figures 1 and 2. The expansion curves are plotted from different origins in order to show the individual characteristics of each curve.

The results obtained in the first heating and cooling indicate that the single crystals of antimony behave like cast metals or alloys as far as strains are concerned, for the observations on cooling do not coincide with the expansion curves on heating. This is in agreement with Davey's statement [14] that all crystals are produced in a state

---

[12] Hidnert and Sweeney, BS J. Research **1**, 771 (1928) RP29.

[13] Souder and Hidnert, BS Sci. Pap. **21**, 1 (1926–27) S524.

[14] Davey, Phys. Rev. **29**, 206 (1927).

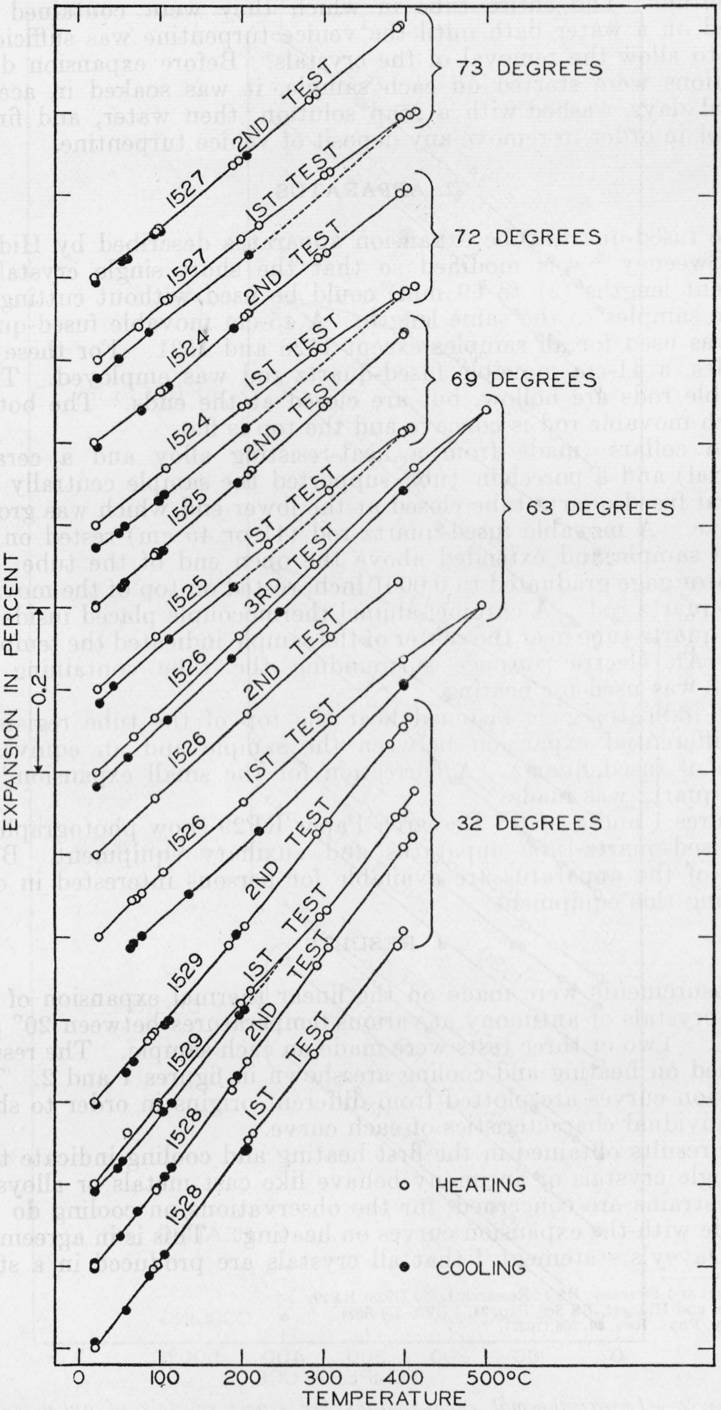

Figure 1.—*Linear thermal expansion of six single crystals of antimony with orientations from 32 to 73 degrees.*

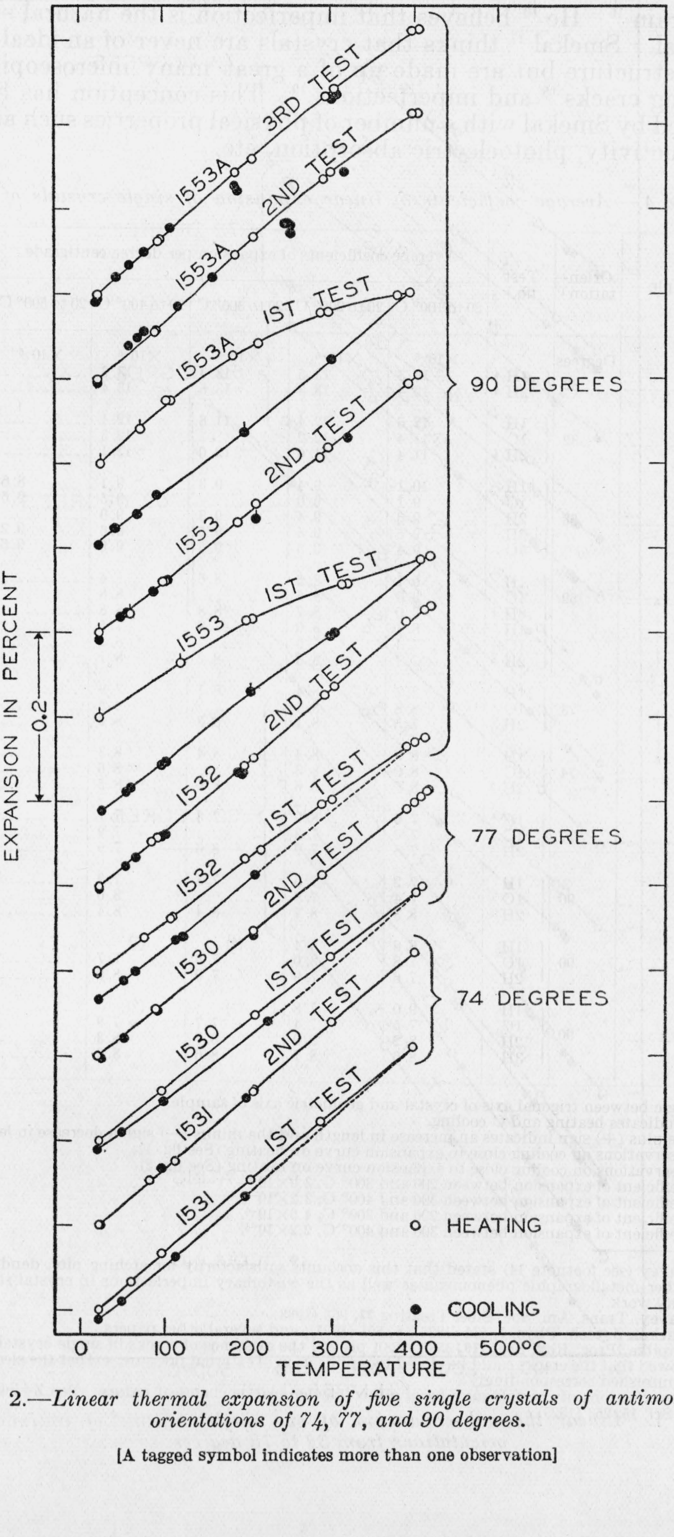

FIGURE 2.—*Linear thermal expansion of five single crystals of antimony with orientations of 74, 77, and 90 degrees.*

[A tagged symbol indicates more than one observation] of strain.[15] He[16] believes that imperfection is the natural state of a crystal. Smekal[17] thinks that crystals are never of an ideal geometrical structure but are made up of a great many microscopic blocks leaving cracks[18] and imperfections.[19] This conception has been correlated by Smekal with a number of physical properties such as electric conductivity, photoelectric absorption, etc.

TABLE 4.—*Average coefficients of linear expansion of single crystals of antimony*

Sample	Orientation[1]	Test no.[2]	Average coefficients of expansion per degree centigrade					Change in length after heating and cooling[3]
			20 to 100° C	20 to 200° C	20 to 300° C	20 to 400° C	20 to 500° C	
	Degrees		$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	$\times 10^{-6}$	*Percent*
1528	32	1H[4]	13.3	13.5	13.4	13.3	----------	+0.01
		2H[4]	12.4	13.3	13.5	13.4	----------	.00
1529	32	1H	12.6	12.4	11.8	12.1	----------	−.01
		1C	11.4	12.2	----------	12.3	----------	
		2H[4]	11.4	11.6	12.0	12.1	----------	−.00
1526	63	1H	10.1	9.4	9.3	9.1	8.6	−.05
		1C	9.1	9.0	----------	9.5	9.6	
		2H	9.6	9.4	9.3	9.0	----------	----------
		3H	9.5	9.5	9.5	9.6	9.2	−.02
		3C	9.4	9.5	9.6	9.6	9.6	
1525	69	1H	9.1	8.5	8.6	8.4	----------	−.02
		1C	9.0	8.5	----------	8.8	----------	
		2H[4]	9.0	8.7	8.8	8.8	----------	+.00
1524	72	1H	8.1	8.0	7.8	7.5	----------	−.03
		1C	7.4	7.9	----------	8.2	----------	
		2H[4]	7.7	8.3	8.2	8.1	----------	−.01
1527	73	1H	8.7	8.4	8.1	7.9	----------	−.02
		1C	8.8	8.1	----------	8.5	----------	
		2H[4]	7.5	8.1	8.3	8.2	----------	−.01
1531	74	1H	8.7	8.4	8.4	8.3	----------	−.01
		1C	8.0	8.3	----------	8.6	----------	
		2H[5]	8.5	8.6	8.6	8.5	----------	−.00
1530	77	1H	7.9	8.0	7.8	7.7	----------	−.01
		1C	7.2	7.3	----------	7.9	----------	
		2H[5]	7.8	7.9	8.0	7.9	----------	−.01
1532	90	1H	7.2	7.5	7.3	7.2	----------	−.03
		1C	7.4	7.8	----------	8.0	----------	
		2H[5]	8.3	8.2	8.4	8.4	----------	−.00
1553	90	1H	6.6	6.4	(6)	(7)	----------	−.11
		1C	7.8	8.0	7.6	7.7	----------	
		2H	7.6	7.9	7.9	8.0	----------	−.01
1553A	90	1H	9.0	7.8	(8)	(9)	----------	−.09
		1C	7.5	7.4	7.7	7.9	----------	
		2H	8.2	9.0	8.7	8.3	----------	−.00
		3H	8.6	8.7	8.6	8.2	----------	−.01

[1] Angle between trigonal axis of crystal and geometric axis of sample.
[2] H indicates heating and C cooling.
[3] The plus (+) sign indicates an increase in length and the minus (−) sign a decrease in length
[4] Observations on cooling close to expansion curve on beating (See fig. 1).
[5] Observations on cooling close to expansion curve on heating (See fig. 2).
[6] Coefficient of expansion between 200 and 300° C, 3.9×10⁻⁶.
[7] Coefficient of expansion between 300 and 400° C, 3.2×10⁻⁶.
[8] Coefficient of expansion between 200 and 300° C, 4.6×10⁻⁶.
[9] Coefficient of expansion between 300 and 400° C, 2.2×10⁻⁶.

---

[15] Davey (see footnote 14) stated that this accounts satisfactorily for etching pits, dendrite formation and other metallographic phenomena as well as the customary imperfections in crystal structure noted in X-ray work.

[16] Davey, Trans. Am. Soc. Steel Treating **21,** 965 (1933).

[17] Smekal, Z. tech. Phys. **7,** 535 (1926); **8,** 561 (1927); and several other papers.

[18] Kapitza, Proc. Roy. Soc. **119,** 358 (1928), proved the existence of cracks in single crystals of bismuth. He showed that the cracks could be closed by application of external pressure, so that the electric resistance was diminished correspondingly.

[19] Imperfections of crystals may be of either mechanical or chemical nature. See Zwicky, Proc. Nat. Acad. Sci. **15,** 253 (1929).

Most of the expansion curves on heating are irregular or concave towards the temperature axis. The expansion curves of samples 1553 and 1553A in the first heating show a "flattening" between 200 and 400° C. In order to determine if this phenomenon was caused by evolution of gas from these samples, a piece of the antimony crystal (as received) from which samples 1553 and 1553A were cut, was subjected to analysis[20] by the vacuum fusion method. The results indicate that the evolution of gas up to 700° C was negligible, of the order of magnitude of the blank correction for the apparatus. The same results were also obtained on sample 1553 after the expansion tests had been completed. It therefore appears that the "flattening" of the expansion curves cannot be accounted for by an evolution of gas.

Most of the observations on cooling are below the expansion curves on heating. The deviations between the observations on cooling and the expansion curves on heating are less in the second tests than in the first tests.

Table 4 gives coefficients of expansion which were obtained from the expansion curves on heating and cooling (figs. 1 and 2). In most cases, the coefficients of expansion are higher in the second tests on heating than in the first tests on heating.

From the coefficients of expansion obtained in the second and third tests for the range between 20 and 100° C (table 4), the following empirical equation[21] was derived by the method of least squares:

$$ {}_{20}E_{100} = (17.17 - 0.1951\theta + 0.001039\theta^2)10^{-6} \tag{1} $$

where ${}_{20}E_{100}$ represents the average coefficient of linear expansion between 20 and 100° C, and θ represents the orientation, or the angle between the trigonal axis of a single crystal and the geometric axis of the specimen. This equation[22] applies for single crystals of antimony with orientations between 32 and 90°.

---

[20] By H. C. Vacher of this Bureau.

[21] Helen D. Megaw, Proc. Roy. Soc., London [A] **142,** 198 (1933), stated that the theory of homogeneous deformation (Fizeau, 1868; Fletcher, 1880; Voigt, Lehrbuch der Kristallphysik, 1910) shows that a sphere of unit radius at a given temperature in any crystal changes at a neighboring temperature into an ellipsoid, the axes of which serve, in length and direction, to determine the expansion of the crystal over this temperature range. "Let the expansion coefficients in the direction of the axes be e_1, e_2, e_3. Then the semiaxes of the ellipsoid corresponding to a temperature change of 1° C are $1+e_1$, $1+e_2$, $1+e_3$, and the expansion coefficient e in any other direction making angles θ_1, θ_2, θ_3 with the axes of the ellipsoid is given by the equation

$$ \frac{1}{(1+e)^2} = \frac{\cos^2\theta_1}{(1+e_1)^2} + \frac{\cos^2\theta_2}{(1+e_2)^2} + \frac{\cos^2\theta_3}{(1+e_3)^2} $$

"For purposes of calculation, it is more convenient to use Voigt's 'deformation tensor.' This is a conic with its axes in the same direction as those of the ellipsoid previously described, but involving the expansion coefficients directly. It is given by the equation

$$ e = e_1 \cos^2\theta_1 + e_2 \cos^2\theta_2 + e_3 \cos^2\theta_3 $$

It can be derived from the previous ellipsoid by neglecting the squares of the expansion coefficients." The latter equation has been given by Fizeau, Compt. rend. **62,** 1101 (1866), who also indicated an equation of the form

$$ e = e_1 \cos^2\theta + e_2 \sin^2\theta $$

for the case where $e_2 = e_3$.

[22] The equation

$$ {}_{20}E_{100} = 13.53 \times 10^{-6} \cos^2\theta + 7.97 \times 10^{-6} \sin^2\theta \tag{1a} $$

was also derived and found to apply as closely as equation 1 for the range from 32 to 90°. However, the coefficient of expansion for 0° orientation computed from equation 1a does not agree as closely with the values reported by Fizeau and Bridgman, as the value obtained from equation 1 (table 6).

A comparison of the observed coefficients of expansion and the values computed from equation 1 are given in table 5. The sum of the squares of the residuals is 3.93×10^{-12} and hence the probable error of a single computed value is equal to

$$0.6745\sqrt{\frac{3.93\times10^{-12}}{14-3}} \text{ or } \pm0.4\times10^{-6}$$

TABLE 5.—*Comparison of observed and computed coefficients of expansion of single crystals*

Orientation	Average coefficients of linear expansion per degree centigrade between 20 and 100° C		Residual	Orientation	Average coefficients of linear expansion per degree centigrade between 20 and 100° C		Residual
	Observed	Computed			Observed	Computed	
Degrees	$\times10^{-6}$	$\times10^{-6}$	$\times10^{-6}$	Degrees	$\times10^{-6}$	$\times10^{-6}$	$\times10^{-6}$
32	12.4	12.0	+0.4	73	7.5	8.5	−1.0
32	11.4	12.0	−.6	74	8.5	8.4	+.1
63	9.6	9.0	+.6	77	7.8	8.3	−.5
63	9.5	9.0	+.5	90	8.3	8.0	+.3
63	9.4	9.0	+.4	90	7.6	8.0	−.4
69	9.0	8.7	+.3	90	8.2	8.0	+.2
72	7.7	8.5	−.8	90	8.6	8.0	+.6

The following equations for temperature ranges from 20 to 200°, 20 to 300° and 20 to 400° C, were also derived by the method of least squares from the coefficients of expansion obtained in the second and third tests (table 4):

$$_{20}E_{200}=(18.88-0.2440\theta+0.001411\theta^2)10^{-6} \tag{2}$$

$$_{20}E_{300}=(19.54-0.2579\theta+0.001479\theta^2)10^{-6} \tag{3}$$

$$_{20}E_{400}=(19.49-0.2548\theta+0.001430\theta^2)10^{-6} \tag{4}$$

In these equations, $_{20}E_{200}$, $_{20}E_{300}$, and $_{20}E_{400}$ represent the average coefficients of linear expansion from 20 to 200° C, 20 to 300° C and 20 to 400° C, respectively, and θ represents the orientations in degrees of the single crystals between 32° and 90°. The probable errors of $_{20}E_{200}$, $_{20}E_{300}$, and $_{20}E_{400}$ are $\pm0.4\times10^{-6}$, $\pm0.3\times10^{-6}$ and $\pm0.3\times10^{-6}$, respectively.

Figure 3 shows comparisons of the observed coefficients of expansion obtained in the second and third tests with the curves representing equations 1 to 4. The curve representing equation 1 for the range from 20 to 100° C was extrapolated to orientation 0° in order to compare this curve with the coefficients of expansion obtained by Fizeau [23] and Bridgman [24] for crystals with 0° orientation. Their values for orientation at 90°, are also indicated in figure 3.

Equations 1 to 4 and the curves in figure 3 show that the coefficients of expansion of single crystals decrease with increase in the orientation. Coefficients of expansion of single crystals of various orientations were computed from equations 1 to 4 and are shown in table 6.

---

[23] See table 1.

[24] See table 1.

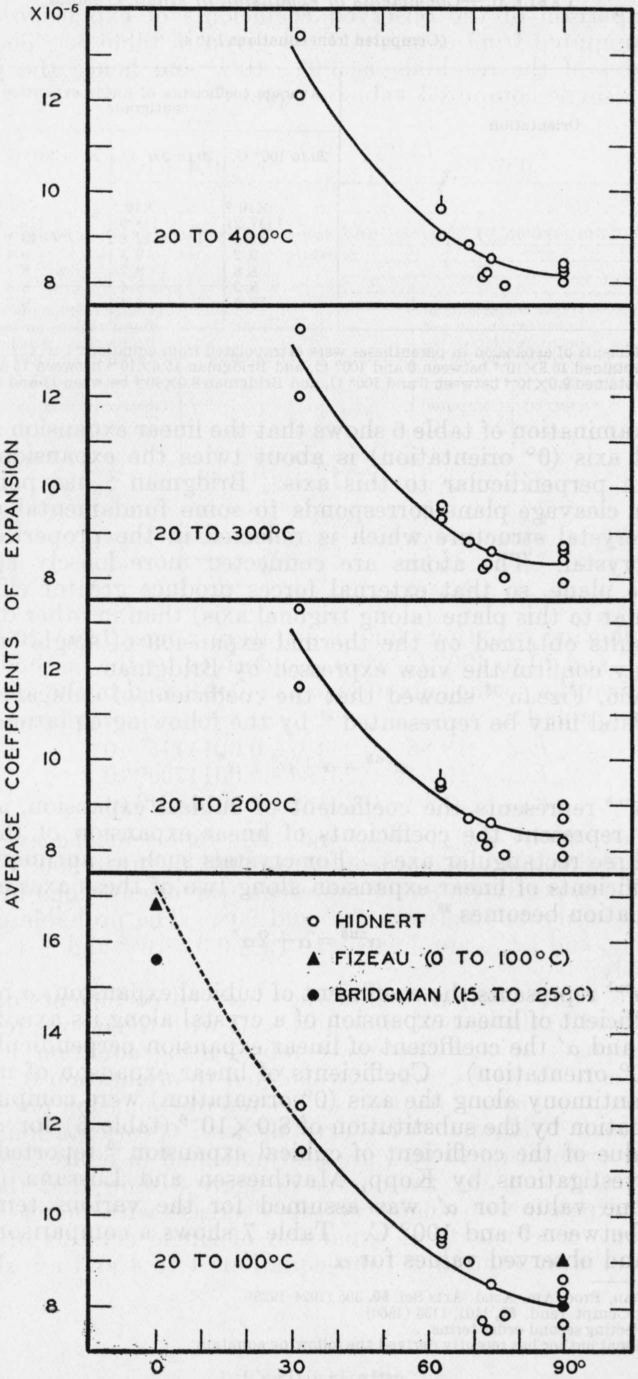

FIGURE 3.—*Relationships between coefficients of linear expansion and orientations of single crystals of antimony.*

The light circles represent observations obtained in the present investigation from which the curves (see equations 1 to 4) were derived]

TABLE 6.—*Coefficients of expansion of single crystals*

(Computed from equations 1 to 4)

Orientation	Average coefficients of linear expansion per degree centigrade			
	20 to 100° C	20 to 200° C	20 to 300° C	20 to 400° C
Degrees	×10⁻⁶	×10⁻⁶	×10⁻⁶	×10⁻⁶
0 [1]	[2] (17.2)	(18.9)	(19.5)	(19.5)
30	12.3	12.8	13.1	13.1
60	9.2	9.3	9.4	9.4
70	8.6	8.7	8.7	8.7
80	8.2	8.4	8.4	8.3
90	[3] 8.0	8.4	8.3	8.1

[1] The coefficients of expansion in parentheses were extrapolated from equations 1 to 4.
[2] Fizeau obtained 16.8×10^{-6} between 0 and 100° C, and Bridgman 15.6×10^{-6} between 15 and 25° C.
[3] Fizeau obtained 9.0×10^{-6} between 0 and 100° C, and Bridgman 8.0×10^{-6} between 15 and 25° C.

An examination of table 6 shows that the linear expansion along the trigonal axis (0° orientation) is about twice the expansion along a direction perpendicular to this axis. Bridgman [25] has pointed out that the cleavage plane corresponds to some fundamental condition in the crystal structure which is reflected in the properties of the single crystal. The atoms are connected more loosely across the cleavage plane, so that external forces produce greater effects perpendicular to this plane (along trigonal axis) than in other directions. The results obtained on the thermal expansion of single crystals of antimony confirm the view expressed by Bridgman.

In 1866, Fizeau [26] showed that the coefficient of cubical expansion of a crystal may be represented [27] by the following equation:

$$\alpha^{cub} = \alpha + \alpha' + \alpha''$$

where α^{cub} represents the coefficient of cubical expansion, and α, α', and α'' represent the coefficients of linear expansion of the crystal along three rectangular axes. For crystals such as antimony, where the coefficients of linear expansion along two of these axes are equal, this equation becomes [28]

$$\alpha^{cub} = \alpha + 2\alpha' \tag{5}$$

where α^{cub} represents the coefficient of cubical expansion, α represents the coefficient of linear expansion of a crystal along its axis (0° orientation) and α' the coefficient of linear expansion perpendicular to the axis (90° orientation). Coefficients of linear expanson of monocrystalline antimony along the axis (0° orientation) were computed from this equation by the substitution of 8.0×10^{-6} (table 6) for α' and of each value of the coefficient of cubical expansion [29] reported in separate investigations by Kopp, Matthiessen and Lussana (table 2). The same value for α' was assumed for the various temperature ranges between 0 and 100° C. Table 7 shows a comparison of computed and observed values for α.

---

[25] Bridgman, Proc. Am. Acad. Arts Sci. **60**, 305 (1924–1925).

[26] Fizeau, Compt. rend. **62**, 1101, 1133 (1866).

[27] By neglecting second-order terms.

[28] The present author has recently derived the following equation:

$$\alpha^{cub} = (1+\alpha)(1+\alpha')^2 - 1.$$

This equation may be reduced to equation 5 by neglecting second-order terms.

[29] The coefficient of cubical expansion is the same for monocrystalline and polycrystalline antimony.

TABLE 7.—*Comparison of coefficients of linear expansion of monocrystalline antimony along axis (0° orientation)*

Observer	Temperature range	Coefficients of expansion per degree centigrade	
		Computed [1]	Observed
	° C	$\times 10^{-6}$	$\times 10^{-6}$
Kopp	12 to 42	17	
Matthiessen	0 to 100	15.7	
Lussana	9 to 72	19.3	
Fizeau	0 to 100		16.8
Bridgman	15 to 25		15.6
Hidnert	20 to 100		[2] 17.2
Average		17.3	16.5

[1] Values for Kopp, Matthiessen, and Lussana were computed from equation 5 (Fizeau's equation).
[2] Extrapolated from equation 1 and taken from table 6.

The densities of four single crystals of antimony were determined [30] after the measurements on thermal expansion had been completed. Table 8 gives the densities obtained. All except that for sample 1553 A are in the neighborhood of the highest value obtained for the density of polycrystalline antimony (section III–1).

TABLE 8.—*Densities of single crystals of antimony*

Sample	Density [a] at 25° C
	g/cm³
1528	6.684
1525	6.687
1532	6.670
1553A	[b] 6.572

[a] Values are believed to be correct to within ± 0.005.
[b] The density was redetermined by W. S. Clabaugh of this Bureau, who obtained the value 6.569.

Davey [31] has pointed out that the densities of different crystals of the same metal rarely agree exactly, for crystals do not exactly duplicate each other on account of the effect of traces of impurities on the interplanar distances and the fragmentation of the crystal.

III. POLYCRYSTALLINE ANTIMONY

1. MATERIALS INVESTIGATED

Three samples of cast polycrystalline antimony were used in the investigation. Table 9 gives information about the preparation [32] of the samples:

---

[30] By S. Alpher of this Bureau.
[31] Davey, A Study of crystal structure and its applications (McGraw-Hill Book Co., Inc., New York, 1934).
[32] C. M. Saeger of this Bureau prepared these samples.

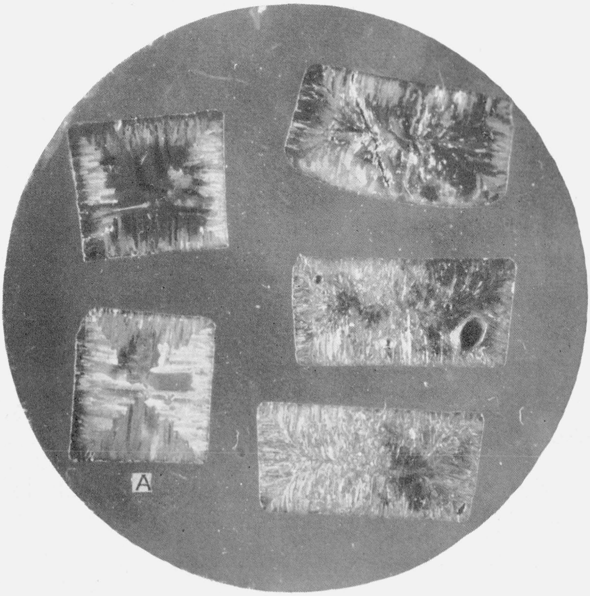

FIGURE 4.—*Two cross sections of sample 1067 (left) and three cross sections of sample 1450 (right).*

Electrolytic etch in 5 percent oxalic acid. ×2.

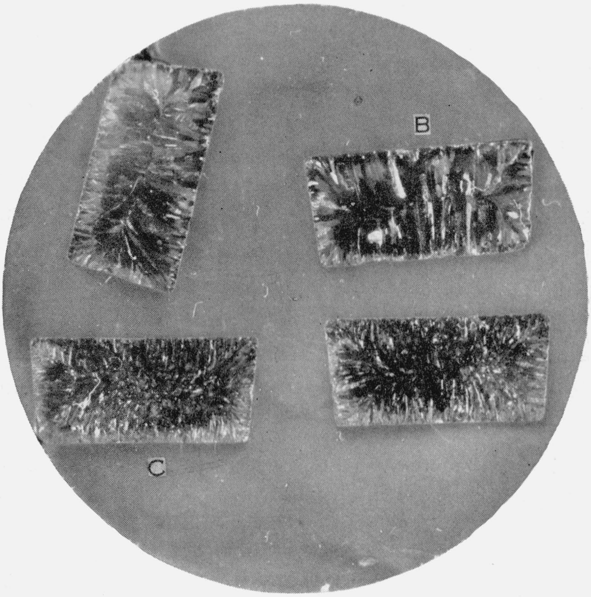

FIGURE 5.—*Four cross sections of sample 1450.*

Electrolytic etch in 5 percent oxalic acid. ×2.

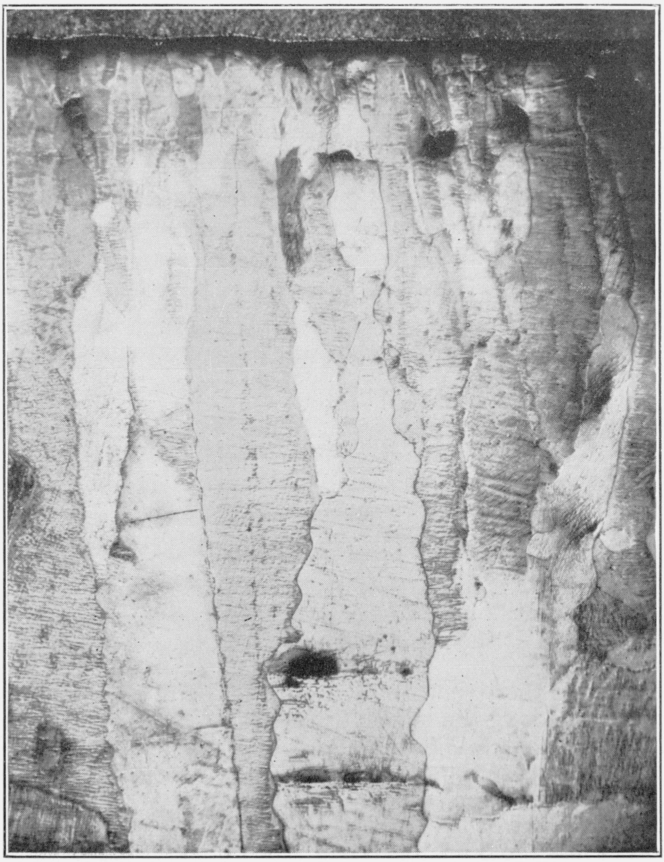

FIGURE 6.—*Area at* A, *figure 4, showing columnar structure of sample 1067.*

Edge of sample at top, center of sample near bottom of figure. Electrolytic etch in 5 percent oxalic acid. ×30.

TABLE 9.—*Preparation, chemical composition and density of polycrystalline antimony*

Sample	Cross-section of sample [a]	Origin and preparation	Purity [b]	Density[c] at 25° C
	mm			g/cm^3
1067	10×9	Antimony, Cookson's best grade (purchased from Nathan Trotter & Co., Philadelphia, Pa.), cast in a sand mold.	[d] 99.8	[e] 6.630
1450	15×7	Two bars of cast antimony, each about 15 cm long (purchased from C. A. F. Kahlbaum Chemische Fabrik, Adlershof bei Berlin, Germany) were joined together in a skin-dried sand mold to make one sample, by pouring molten antimony (from same source) at the point of joining.	[f] 99.9+	[g] 6.656
1451	19×7	Pieces of cast antimony from same source as sample 1450 were melted in a graphite crucible, open flame, and cast in a skin-dried sand mold.	[f] 99.9+	[g] 6.684

[a] The length of each sample was about 300 mm.

[b] After the thermal expansion determinations were completed, chemical analyses were made on pieces cut from the ends and the center of each sample.

[c] Density determinations were made on pieces cut from the samples after the thermal expansion measurements had been completed.

[d] Lead not detected. Chemical analysis made by H. A. Buchheit, formerly of this Bureau.

[e] Determined by Miss E. E. Hill, formerly of this Bureau.

[f] Lead, iron, copper, and zinc were not detected in a ten-gram sample. Chemical analysis made by W. D. Mogerman of this Bureau.

[g] Determined by E. L. Peffer of this Bureau.

After the determinations on the thermal expansion of the samples of polycrystalline antimony had been completed, microscopic examinations of portions from samples 1067 and 1450 were made.[33]

Sample 1067 showed only one kind of surface of fracture, which was coarsely granular and columnar in appearance. Sample 1450 displayed two different surfaces of fracture. One of these was a coarse grained fracture in which the grains were apparently columnar. The other fracture appeared to be fine grained and not markedly columnar. This suggested a variation in structure along the length of the sample, which was later confirmed by microscopic examination.

For microscopic examination three cross sections were cut from sample 1067. Two of these sections are shown at the left in figure 4. Eight cross sections were cut from specimen 1450. Three of these sections are shown at the right in figure 4 and four sections are shown in figure 5.

Figure 4 shows the structure of sample 1067 to be markedly columnar and uniform along the length of the sample. Figure 6 shows the area at A, figure 4, at a higher magnification. Near the edge (top of figure) there are a number of comparatively small grains, all apparently columnar. This was probably a region of rapid cooling due to the chilling effect of the mold in casting. Farther from the edge in a region of less rapid cooling the grains are larger and their columnar nature more pronounced. These grains, particularly the large ones in which growth was less restricted, show cleavage planes perpendicular to the axes of the columns. These are shown at a higher magnification in figure 7. It will be noted that the cleavage planes are mostly in the same direction. Many of them pass from one grain to another with little change in direction. Only in small grains, where crystalline growth was probably restricted, are there marked differences in the direction of the cleavage planes.

Antimony crystallizes in the rhombohedral form (hexagonal system) and has a perfect cleavage parallel to the basal plane of the

---

[33] By D. J. McAdam, Jr., and R. E. Pollard of this Bureau.

hexagonal system. It is probable that the cleavage planes shown in figure 7 are parallel to the basal planes. Their similarity indicates a preferred orientation in sample 1067.[34] It is probable that each of the columnar grains growing toward the center approximates a single crystal.

It will be noted from figures 4 and 5 that the structure of sample 1450 varies considerably along its length. Some of the cross sections shown in these figures are coarse grained and markedly columnar in structure. Others are fine grained and columnar only at or near the edges. It is estimated that between one-half and two-thirds of this material was fine grained and not markedly columnar. Figures 8 and 9 show the differences in structure in different sections of this material. These figures show, respectively, the areas at B and C of figure 5. In figure 8 the columnar structure is marked. In figure 9 the columnar structure is marked only at the edge (top of figure). In the interior the grains are almost entirely equiaxed.

Figure 10 shows the structure of a markedly columnar section of sample 1450 at a higher magnification. In this material the cleavage planes are diversified in direction. Even in markedly columnar sections there is much less evidence of a preferred direction of cleavage than in sample 1067.

It seems probable that in such regions relatively few crystals grew without restriction, in the direction of the radial temperature gradient, and that the rest of the material solidified between these columnar crystals so as to have a pseudomorphic columnar structure. It appears probable that when antimony crystals grow freely in the direction of a steep temperature gradient, the column elongates in the direction of the principal axis, the axis along which the coefficient of expansion is greatest.

In noncolumnar sections there is no evidence of preferred direction of cleavage. Sample 1450,[35] therefore, shows evidence of preferred orientation in only a small proportion of its length and cross section.

The results obtained on the microscopic examination of these samples may be summarized as follows:

Sample 1067 was found to be uniformly cross grained and columnar. The structure was uniform along the length of the sample. In this material the cleavage planes displayed a markedly preferred direction perpendicular to the axis of the columns.

The structure of sample 1450 was found to vary considerably along its length. Part of this material was coarse grained and columnar. In other parts the structure was fine grained and not columnar. Even in the columnar portions this material did not show as much evidence of preferred orientation as did sample 1067. In sections which were not columnar no evidence of preferred orientation was found.

The large differences of density (shown in table 8A) between the three samples of polycrystalline antimony are doubtless to be explained by the formation of microscopic cavities, on account of unequal contraction in different directions of the crystal grains in cooling from

---

[34] The thermal expansion of such material is expected to vary considerably, depending on the direction in which it is measured. Measured longitudinally (across the columns) the thermal expansion would be expected to approximate that of a single crystal perpendicular to the axis. It is significant that the coefficient of expansion of 9×10^{-6} (20 to 100° C) found for sample 1067 (table 10) is near that of 8×10^{-6} given for a single crystal perpendicular to the axis (table 6).

[35] It is significant that the coefficient of expansion (20 to 100° C) found for this sample (table 10) is practically equal to the value 11.1×10^{-6} computed for polycrystalline antimony having random orientation (page 542).

Figure 7.—*Structure of sample 1067 showing preferred direction of cleavage planes.*

Edge of sample at top of figure. Electrolytic etch in 5 percent oxalic acid. ×100.

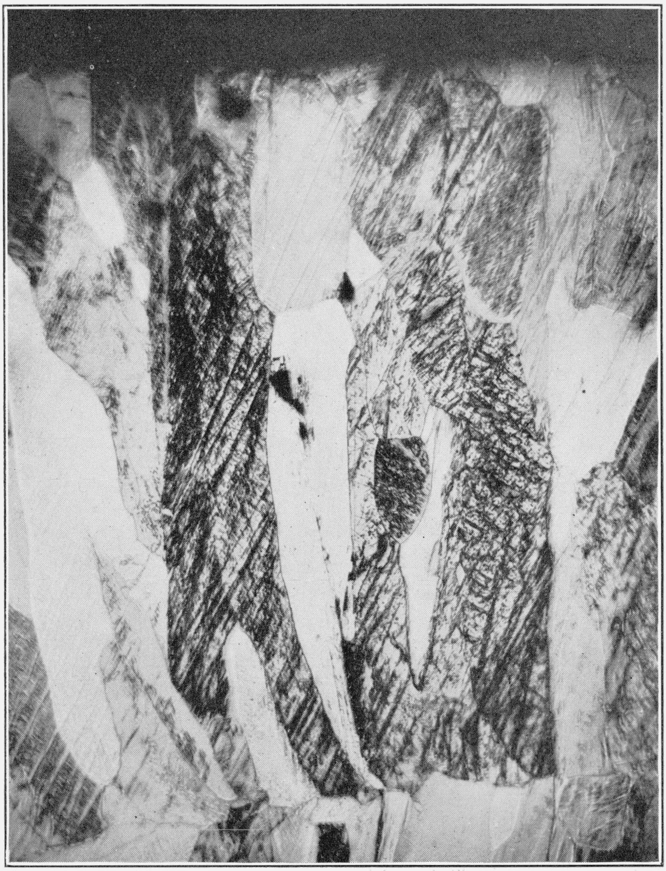

FIGURE 8.—*Area at* B, *figure 5, showing columnar structure of portion of sample 1450.*

Edge of sample at top, center of sample at bottom of figure. Electrolytic etch in 5 percent oxalic acid. ×30.

FIGURE 9.—*Area at* C, *figure 5, showing noncolumnar structure of portion of sample 1450.*

Top of sample at top, center at bottom of figure. Electrolytic etch in 5 percent oxalic acid. ×30.

FIGURE 10.—*Structure of columnar portion of sample 1450, showing more diversified direction of cleavage planes.*

Edge of sample at top of figure. Electrolytic etch in 5 percent oxalic acid. ×100.

the melting point. The cavitation is different, for the mean orientation of each sample is not the same.

2. APPARATUS

The furnace shown in figure 1 of BS Scientific Paper S488 was used for the measurements of the linear thermal expansion of sample 1067, and the white furnace shown at the extreme left of figure 1 of BS Scientific Paper S524 was used for samples 1450 and 1451. Figure 4 of the latter paper indicates the method used in mounting samples 1450 and 1451 in the furnace.

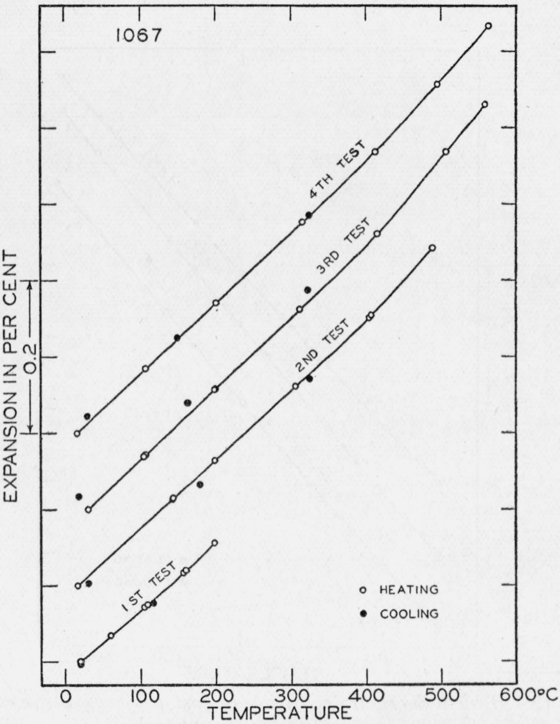

Figure 11.—*Linear thermal expansion of cast polycrystalline antimony.*
[Sample 1067.]

Expansion measurements were made by means of micrometer microscopes, which were sighted on fine wires suspended from or in contact with the ends of the specimen. For a detailed description of the apparatus and the methods used, the reader should refer to the publications mentioned. The apparatus used for the determinations of the thermal expansion of polycrystalline antimony is more accurate than the apparatus employed for the measurements of the expansion of monocrystalline antimony.

3. RESULTS

Measurements [36] were made on the linear thermal expansion of three samples of cast polycrystalline antimony at various temperatures between 20 and 560° C. The results obtained are indicated in figures 11 and 12. The smoothness of the thermal expansion curves shows that there is no polymorphic transition [37] between 20 and 560°C. From X-ray examination of the crystal structure of antimony to 210° C, and from measurements on the electrical resistance between 20 and 550° C, and on the thermal expansion up to 600° C, Schulze and Graf [38] likewise concluded that antimony has no allotropic transformation between 20 and 600° C.

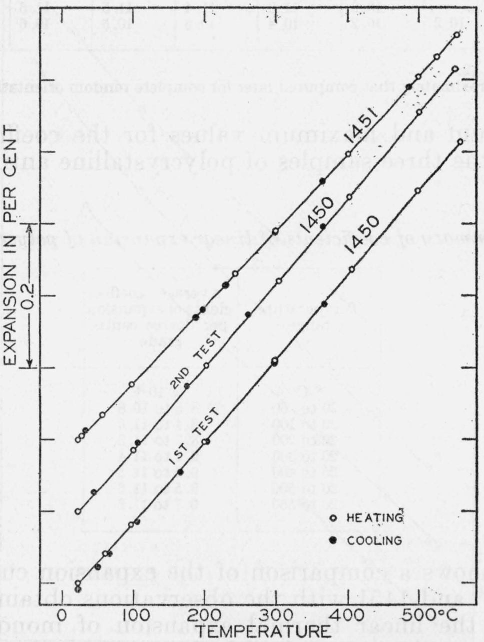

FIGURE 12.—*Linear thermal expansion of cast polycrystalline antimony.*

[Samples 1450 and 1451.]

Table 10 gives average coefficients of expansion which were derived from the expansion curves of figures 11 and 12. This table also indicates the difference in length before and after each expansion test. The plus (+) sign signifies an increase in length and the minus (−) sign a decrease in length.

---

[36] H. S. Krider assisted in the measurements on the thermal expansion of polycrystalline antimony. An abstract of these results was reported by Hidnert and Krider to the American Physical Society at the Chicago meeting in November 1932. See Phys. Rev. **42**, 911 (1932).

[37] Cohen and Van den Bosch, Z. Phys. Chem. **89**, 757 (1915); and Jaenecke, Z. physik. Chem. **90**, 337 (1915) reported transformations in antimony at 106° C and between 125 and 137° C, respectively. Kersten, Physics **2**, 276 (1932) reported that electrodeposited antimony has an amorphous structure when deposited at temperatures below 25° C, and changes from amorphous to crystalline between 25 and 30° C.

[38] Schulze and Graf, Metallwirtschaft, **12**, 19 (1933). These authors state that the irregularity found near 100° C in the electrical resistance of polycrystalline antimony before tempering or after tempering at 200° C, can apparently be explained by mechanical deformation and not by an allotropic transformation. They did not publish any quantitative data on the thermal expansion of antimony, for they were interested only in the shape of the expansion curve.

TABLE 10.—*Average coefficients of linear expansion of cast polycrystalline antimony*

Sample	Test no.	Average coefficients of expansion per degree centigrade							Change in length after heating to maximum [1] temperature and cooling to room temperature
		20 to 60° C	20 to 100° C	20 to 200° C	20 to 300° C	20 to 400° C	2 to 500° C	20 to 550° C	
		×10⁻⁶	×10⁻⁶	×10⁻⁶	×10⁻⁶	×10⁻⁶	×10⁻⁶	×10⁻⁶	*Percent*
1067	1	8.5	8.4	8.7	---------	---------	---------	---------	−0.003
	2	---------	9.8	9.3	9.2	9.2	9.5	---------	−.010
	3	---------	9.4	9.4	9.3	9.3	9.8	10.0	+.029
	4	---------	9.3	9.3	9.2	9.3	9.6	9.7	+.009
1450	1	10.8	[2] 11.0	11.1	11.3	11.4	11.5	11.5	−.008
	2	---------	10.9	11.3	11.4	11.5	11.6	11.6	.000
1451	1	10.2	10.2	10.4	10.5	10.5	10.6	10.6	+.002

[1] See figures 11 and 12.
[2] This value is approximately that computed later for complete random orientation.

The minimum and maximum values for the coefficients of linear expansion of the three samples of polycrystalline antimony are given in table 11.

TABLE 11.—*Summary of coefficients of linear expansion of polycrystalline antimony*

Temperature range	Average coefficients of expansion per degree centigrade
° C	× 10⁻⁶
20 to 60	8.5 to 10.8
20 to 100	8.4 to 11.0
20 to 200	8.7 to 11.3
20 to 300	9.2 to 11.4
20 to 400	9.2 to 11.5
20 to 500	9.5 to 11.6
20 to 550	9.7 to 11.6

Figure 13 shows a comparison of the expansion curves of samples 1067,[39] 1450,[40] and 1451 with the observations obtained by previous observers on the linear thermal expansion of monocrystalline and polycrystalline antimony, and with the expansion curves[41] of monocrystalline antimony (0° and 90° orientation only), from this paper. An examination of the results in table 10 and figure 13 shows that a different expansion was obtained for each sample of polycrystalline antimony used in the present investigation. The values obtained for the expansion of these samples of polycrystalline antimony are intermediate between those obtained for monocrystalline antimony having 0° and 90° orientation, respectively. The values for polycrystalline antimony are much closer to the values obtained for monocrystalline antimony of 90° orientation than for 0° orientation. It appears that the explanation is to be sought in the structure of the metal, for the expansion of crystalline antimony depends on its

---

[39] Curve obtained from four tests on heating.
[40] Curve obtained from two tests on heating.
[41] Obtained from the data given in table 6. The curve for 0° orientation represents extrapolated values, and was therefore drawn as a broken or dashed curve. This curve, computed from equations 1 to 4, which were derived from the expansion data for orientations between 32 and 90°, is in excellent agreement with the data reported by Fizeau and Bridgman (see fig. 13).

orientation. Assuming that there is random orientation[42] of the crystals in a bar of polycrystalline antimony, then the coefficient of linear expansion of polycrystalline antimony may be expressed by the following equation:

$$a=\frac{1}{3}(a_0+2a_{90}) \tag{6}$$

where a is the coefficient of linear expansion of polycrystalline antimony, a_0 is the coefficient of linear expansion of monocrystalline antimony having 0° orientation, and a_{90} is the coefficient of linear expansion of monocrystalline antimony having 90° orientation. If the values 17.2×10^{-6} and 8.0×10^{-6} (table 6) are substituted for a_0 and a_{90}, respectively, in equation 6, the value[43] 11.1×10^{-6} is obtained for the coefficient of linear expansion of polycrystalline antimony (random distribution) between 20 and 100° C. Values different from this may be considered as due to variations from random orientation.

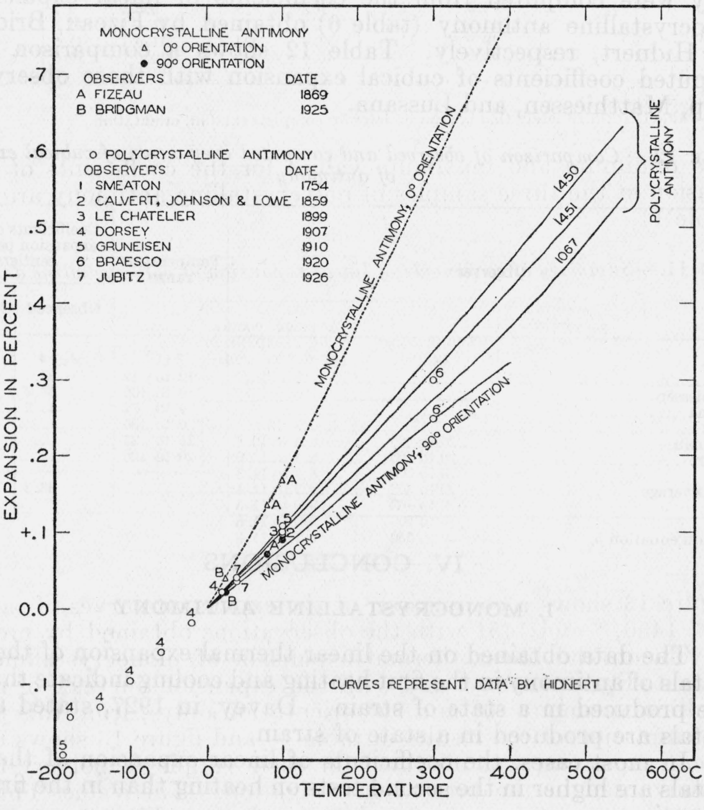

FIGURE 13.—*Comparison of linear thermal expansion of monocrystalline and polycrystalline antimony.*

---

[42] Gough, Am. Soc. Testing Materials **33**, part 2, (1933), pointed out that metals in the normal state usually consist of an aggregate of very small crystals of random orientation, this structure being due to crystallization from the liquid proceeding simultaneously from a number of different nuclei.

[43] This value can also be derived by taking one-third of the coefficient of cubical expansion of antimony (monocrystalline or polycrystalline).

The values of this paper apply only to cast antimony. Jubitz[44] found that a drawn sample of antimony[45] expands less than a sample as cast, and stated that this metal shows a distinct thermal anisotropy. He found larger differences for zinc and cadmium. He stated that working (rolling, hammering, drawing, etc.) metals[46] belonging to the regular system, has no appreciable effect on the expansivity, but that there is an effect on metals[47] which do not belong to the regular system. Braesco[48] observed that a quenched or annealed sample of agglomerated antimony expands less than a quenched or annealed sample of cast antimony.

By means of equation 5, coefficients of cubical expansion of antimony were computed from the coefficients of linear expansion of monocrystalline antimony (table 6) obtained by Fizeau, Bridgman, and Hidnert, respectively. Table 12 shows a comparison of the computed coefficients of cubical expansion with those observed by Kopp, Matthiessen, and Lussana.

TABLE 12.—*Comparison of observed and computed coefficients of cubical expansion of antimony*

Observer	Temperature range	Coefficients of cubical expansion per degree centigrade	
		Observed	Computed [1]
	° C	×10⁻⁶	×10⁻⁶
Kopp	12 to 42	33	
Matthiessen	0 to 100	31.7	
Lussana	9 to 72	35.3	
Fizeau	0 to 100		34.8
Bridgman	15 to 25		31.6
Hidnert	20 to 100		33.2
Average		33.3	33.2

[1] From equation 5.

IV. CONCLUSIONS

1. MONOCRYSTALLINE ANTIMONY

1. The data obtained on the linear thermal expansion of the single crystals of antimony in the first heating and cooling indicate that they were produced in a state of strain. Davey, in 1927, stated that all crystals are produced in a state of strain.

2. In most cases, the coefficients of linear expansion of the single crystals are higher in the second tests on heating than in the first tests on heating.

3. The following equations show the relationships between the coefficients of expansion and the orientations of the single crystals of antimony:

$$ {}_{20}E_{100}=(17.17-0.1951\theta+0.001039\theta^2)10^{-6} \qquad (1)$$
$$ {}_{20}E_{200}=(18.88-0.2440\theta+0.001411\theta^2)10^{-6} \qquad (2)$$
$$ {}_{20}E_{300}=(19.54-0.2579\theta+0.001479\theta^2)10^{-6} \qquad (3)$$
$$ {}_{20}E_{400}=(19.49-0.2548\theta+0.001430\theta^2)10^{-6} \qquad (4)$$

---

[44] See table 2.

[45] Also for bismuth.

[46] Bronze and cast iron.

[47] Antimony, zinc, cadmium, and magnesium.

[48] See table 2.

In these equations, ${}_{20}E_{100}$, ${}_{20}E_{200}$, ${}_{20}E_{300}$ and ${}_{20}E_{400}$ represent the average coefficients of linear expansion from 20 to 100° C, 20 to 200° C, 20 to 300° C, and 20 to 400° C, respectively, and θ represents the orientations of the single crystals between 32 and 90°. The probable errors of ${}_{20}E_{100}$, ${}_{20}E_{200}$, ${}_{20}E_{300}$, and ${}_{20}E_{400}$ are $\pm 0.4\times 10^{-6}$, $\pm 0.4\times 10^{-6}$, $\pm 0.3\times 10^{-6}$, and $\pm 0.3\times 10^{-6}$, respectively. The computed values for the average coefficients of expansion of monocrystalline antimony with 0° orientation and 90° orientation are 17.2×10^{-6} and 8.0×10^{-6}, respectively, between 20 and 100° C.

4. The coefficient of linear expansion depends upon the direction taken. Figure 3 and table 6 show the coefficients of expansion for various orientations. The coefficients of expansion along the trigonal axis (0° orientation) are about twice as large as the coefficients of expansion along a direction perpendicular to this axis (90° orientation). These results confirm Bridgman's statement that the atoms are connected more loosely across the cleavage plane, so that external forces produce greater effects perpendicular to this direction (along trigonal axis) than in others.

2. POLYCRYSTALLINE ANTIMONY

1. The linear thermal expansion curves of polycrystalline antimony show that there is no polymorphic transition between 20 and 560° C.

2. The average coefficients of linear expansion of three samples of polycrystalline antimony for various temperature intervals between 20 and 550° C were found to range from 8.4×10^{-6} to 11.6×10^{-6} per degree C. The cause of the differences obtained in the expansion of different samples of polycrystalline antimony is attributed to variations in the average orientation of the crystals.

3. Figure 13 shows a comparison of the linear thermal expansion of monocrystalline and polycrystalline antimony.

The author expresses his appreciation to Prof. P. W. Bridgman of Harvard University for his cooperation in the work on single crystals of antimony. Acknowledgment is also due to Dr. S. B. Hendricks of the Bureau of Chemistry and Soils of the U. S. Department of Agriculture, and to the following members of the staff of the National Bureau of Standards: L. J. Briggs, H. W. Bearce, W. Souder, M. R. Thompson, and L. V. Judson, for valuable suggestions.

WASHINGTON, December 20, 1934.