

BOULANGERITE

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ABSTRACT

Boulangerite, a lead sulfantimonide, has been studied on new material, the first which has proved suitable for detailed crystallographic examination. It is monoclinic, prismatic, $2/m$, pseudo-orthorhombic. Elements $a:b:c=0.9158:1:0.3456$; $\beta=100^{\circ}39\frac{1}{2}'$. Some 60 crystal forms were observed. Lattice constants (Berry): $a_0=21.14 \text{ \AA}$, $b_0=23.46 \text{ \AA}$, $c_0=8.07 \text{ \AA}$, $\beta=100^{\circ}48'$. New analyses by Gonyer on Washington boulangerite confirmed Shannon's earlier analysis and Berry's cell content of $\text{Pb}_{40}\text{Sb}_{32}\text{S}_{58}$.

CRYSTALLOGRAPHY

Boulangerite is one of the many fibrous sulfantimonides of lead concerning the crystal form of which little of a definite nature has hitherto been known. Its slender striated prisms generally show no terminal faces. Sjögren (1897) measured crystals from Sala, Sweden, which he regarded as orthorhombic, and on the basis of a single domal face he was able to establish an axial ratio. Shannon (1921) found a single pyramid face on a crystal from Stevens County, Washington, and also inferred orthorhombic symmetry but obtained an axial ratio in very poor agreement with that of Sjögren.

New studies on boulangerite from three localities have been successful in revealing its true crystallographic nature. The mineral proves to be monoclinic with an astonishing wealth of forms. The results of these studies are presented in the following pages.

The first measurable crystals of boulangerite were found in small vugs of a massive, coarsely fibrous material from Rocker Gulch Placer Claim near Deerlodge, Montana (Specimen No. 92671). This material was supplied by Ward's Natural Science Establishment and bore the name geocronite. In addition to the dominant boulangerite, there is coarsely granular sphalerite present, crystals of pyrite, and, in a single cavity, crystals of bournonite, identified by crystal measurement, and showing the forms:—

<i>c</i> 001	<i>e</i> 210	Σ 031	<i>z</i> 201	<i>p</i> 223	<i>O</i> 213
<i>a</i> 100	<i>k</i> 013	<i>x</i> 102	ϕ 113	<i>y</i> 111	<i>v</i> 211
<i>m</i> 110	<i>n</i> 011	<i>o</i> 101	<i>u</i> 112	<i>g</i> 221	

The boulangerite crystals are needles of dark iron-gray color, less than a half millimeter in diameter and from a few to ten or more millimeters

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in length. Since they project from the walls of vugs, they are all singly terminated. The needles are very brittle, the slightest pressure causing them to break into thin leaf-like flakes along a single pinacoidal cleavage, which proved to be parallel to $a\{100\}$. The prisms are deeply striated parallel to their length so that they gave generally a continuous chain of weak or colored signals on the goniometer, but the best crystals gave a consistent series of prism forms. The numerous terminal faces are so minute and so irregular as to give under the binoculars no definite clue of symmetrical distribution; but their weak signals gave position angles which when projected yielded a definite pattern, repeated, although with great variation in the forms present, on successive crystals. By plotting the measurements of each crystal on transparent paper and superimposing these projections, with rotation about the projection center, parallelism was obtained and the form series was developed.

The projection showed apparent orthorhombic symmetry as regards spacing of poles. However, the radial zones to prism faces did not intersect in the projection center but rather at a point about ten degrees off center—a point not represented by a crystal face on any of the crystals at first measured. This fixed the symmetry as probably monoclinic, a conclusion strengthened by each succeeding measurement and subsequently proved by *x*-ray study as reported on a later page.

Identical crystallographic characters including the wealth of forms were later found on boulangérite from the Gold Hunter Mine, Mullan, Idaho, described by Shannon (1918) as "mullanite"; and on crystals from Stevens County, Washington, also analyzed by Shannon (1925).

Boulangérite is, then, monoclinic but with pronounced pseudosymmetry both orthorhombic and tetragonal, as shown in the gnomonic projection, Fig. 1, which presents several unusual features. Although there is an unusually rich form series, one primary zone $[010]$, parallel to the symmetry plane, is almost missing since both $\{100\}$ and $\{001\}$ are seldom found and only one orthodome, $\{101\}$, is at all common. The axial zone $[001]$ is of course strong, the crystals being strongly prismatic in that direction and showing four well-established prism forms. The clinodome zone $[100]$ is also well marked. A strange feature of this projection is the absence of poles in the central area. $\{001\}$, $\{011\}$ and $\{201\}$ are among the rarest forms and the unit pyramid $\{111\}$ has been found on but two crystals. If these be omitted, there are but three forms which have a slope angle (ρ) less than 40° .

The lattice of boulangérite is pseudo-orthorhombic because x_0' is almost exactly half of p_0' , another case like that of brochantite recently described by Palache (1939). It is pseudotetragonal because p_0' and q_0' are so nearly equal and the ϕ of $\{110\}$ is about 48° .

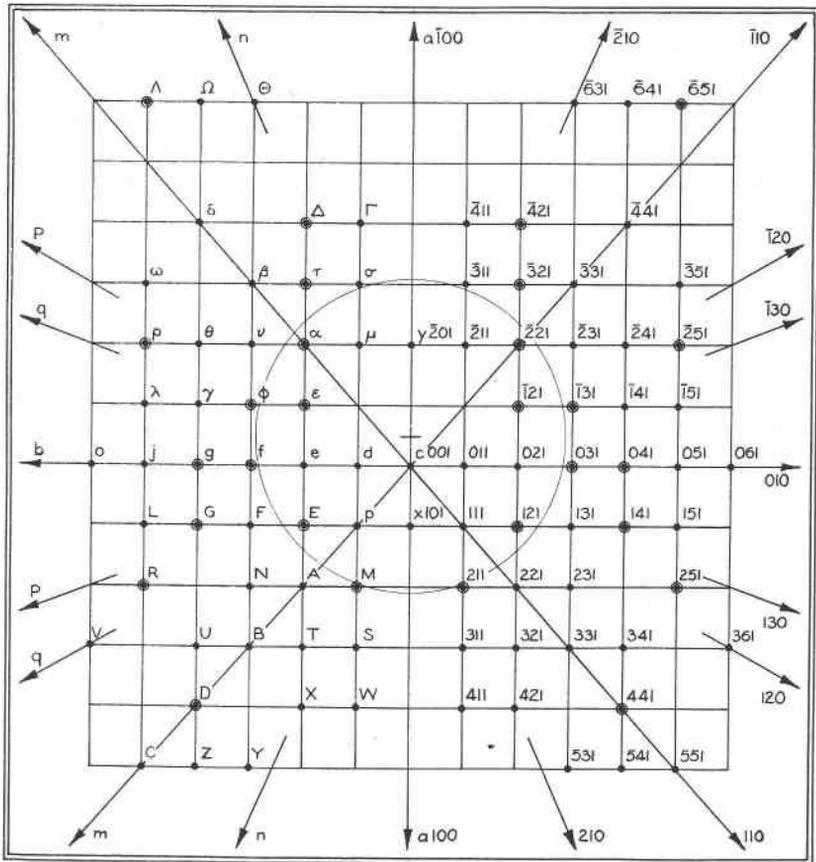


FIG. 1. Gnomonic projection of boulangierite. \odot indicates most common forms.

The combinations of forms found on eight crystals from Montana, five from Idaho, and two from Washington are shown in Table 1. From this table it appears that the most frequently occurring forms on Montana crystals are: a , q , m , n , f , g , D , δ , G , ϕ , R , ρ , X , and Λ ; on Idaho crystals c , m , f , x , α , E , and ϕ ; on Washington crystals f , x , A , α , G , ϕ , and θ . Of these forms only two, $f\{031\}$ and $\phi\{131\}$ are among the commonest for all three localities. Several, however, are common on two localities. It is clear that many forms occur at all the localities and that a larger number are confined to one. This last fact may be due to the better and more numerous crystals measured from Montana. At least ten forms not listed were found once only on the Montana crystals.

TABLE 1—Continued

Locality	Montana								Idaho					Washington	
Crystal	1	2	3	4	5	6	7	8	1	2	3	4	5	1	2
U 341	x	x			x	x									
V 361				x	x			x							
σ 311				x				x	x				x		
τ 321								x		x					
ω 351		x			x						x				
W 411	x							x	x						
X 421	x		x	x	x			x							
Γ 411				x				x							
Δ 421				x				x	x						
Y 531	x	x				x									
X 541		x													x
Θ 631				x				x							
Ω 641				x	x			x							x
Λ 651		x		x	x			x	x						

TABLE 2a. MEASURED ANGLES ON BOULANGERITE FROM MONTANA

		No.	Meas. Aver.		Range			Calculated	
			ϕ	ρ	ϕ	ρ	ϕ	ρ	
<i>b</i>	010	4	0°27'	90°00'	—	2°38'—+ 2°35'	—	0°00' 90°00'	
<i>a</i>	100	5	88 44	90 00	89 04—	94 48	—	90 00 90 00	
<i>g</i>	130	6	20 20	90 00	19 39—	21 09	—	20 19½ 90 00	
<i>m</i>	110	22	47 55½	90 00	47 15—	48 42	—	48 01 90 00	
<i>n</i>	210	18	65 39	90 00	65 00—	66 15	—	65 46½ 90 00	
<i>e</i>	021	1	15 14	35 53	—	—	—	15 14 35 37	
<i>f</i>	031	5	10 25	46 34½	9 21—	10 55	46°25'—46°43'	10 17½ 46 30	
<i>g</i>	041	6	7 58	54 09½	7 31—	8 27	54 14—55 00	7 45 54 22	
<i>j</i>	051	5	6 13	60 06	5 59—	6 31	59 49—60 30	6 13 60 05½	
<i>o</i>	061	2	5 07½	64 21	4 46—	5 29	64 18—64 24	5 11 64 20½	
<i>x</i>	101	2	90 04½	29 52	89 28—	90 41	29 50—29 54	90 00 29 46½	
<i>A</i>	221	2	53 39½	50 25	52 58—	54 21	49 45—57 05	54 08½ 49 43	
<i>B</i>	331	5	52 18	59 25	52 02—	52 26	59 17—59 34	52 16½ 59 27	
<i>D</i>	441	6	51 18	65 39	51 05—	51 27	65 32—65 55	51 16½ 65 39	
<i>C</i>	551	2	50 08	70 01½	49 50—	50 26	70 00—70 03	50 39½ 69 51½	
α	221	1	—39 37	41 56	—	—	—	—39 59½ 42 03½	
β	331	5	—42 37½	55 00	42 12—	42 52	54 40—55 22	—42 54½ 54 45½	
δ	441	5	—44 21	62 59	43 34—	45 52	62 40—63 30	—44 16½ 62 37	

TABLE 2a—Continued

	No.		Meas. Aver.		Range			Calculated	
			ϕ	ρ	ϕ		ρ	ϕ	ρ
E	121	1	39 43	42 10	—		—	39 37	41 54
F	131	3	29 09	49 51	29 05 -	29 11	49 22 -50 20	28 53½	49 49½
G	141	10	22 29	56 16	21 16 -	23 16	56 00 -57 04	22 29	56 14½
L	151	1	18 14	61 16	—		—	18 19½	61 13
ϵ	121	1	-15 19	35 45	—		—	-15 49	35 41½
ϕ	131	5	-10 52	46 08	10 09 -	11 23	45 34 -46 33	-10 41½	46 32
γ	141	2	- 8 20	54 37½	8 11 -	8 28	54 30 -54 45	- 8 03½	54 23½
λ	151	3	- 6 02	59 57	5 39 -	6 29	59 47 -60 05	- 6 28	60 06
M	211	1	69 41	45 37	—		—	70 07½	45 28½
N	231	4	42 30	54 40	41 43 -	42 51	54 25 -55 00	42 41	54 40
R	251	8	29 06	63 14½	28 42 -	29 30	63 00 -63 37	28 57½	63 08½
μ	211	1	-59 53	33 15	—		—	-59 12	34 01
ν	231	3	-27 50	50 03	27 01 -	28 46	49 38 -50 30	-29 13	49 54½
ϕ	241	5	-22 35½	56 26	22 19 -	23 04	56 09 -56 45	-22 45	56 17½
ρ	251	6	-18 32½	61 14	18 10 -	19 05	60 55 -61 33	-18 33	61 15
S	311	3	75 30	54 25	75 12 -	75 44	54 20 -54 32	75 32½	54 09
T	321	3	62 45	56 30	62 41 -	62 50	56 16 -56 54	62 43	56 27
U	341	5	44 05	62 28	43 45 -	44 20	62 00 -62 52	44 06½	62 33
V	361	3	32 54	67 58	32 49 -	33 00	67 12 -68 35	32 52½	67 57
σ	311	4	-69 55½	45 28	-69 31 -	70 10	45 19 -45 33	-70 16½	45 40½
τ	321	2	-54 10	49 46	-54 09 -	54 11	49 43 -49 49	-54 21	49 52
ω	351	3	-28 56	63 29	-28 43 -	29 08	63 12 -64 00	-29 09	63 11½
W	411	3	79 09	60 19	78 25 -	80 01	60 00 -60 42	78 40	60 22½
X	421	8	68 17	61 33	67 49 -	68 39	61 16 -61 50	68 09½	61 42½
Γ	411	2	-75 35	54 31	-75 32 -	75 38	54 22 -54 40	-75 37	54 17½
Δ	421	4	-62 35	56 25	-62 13 -	63 09	56 12 -56 32	-62 51	56 34
Y	531	5	64 00	66 50	-63 26 -	64 42	66 36 -67 12	63 48½	66 56½
Z	541	1	56 25	68 13	—		—	56 45	68 22
Θ	531	3	-63 46	67 04	-63 40 -	63 53	66 53 -67 20	-63 53½	67 00
Ω	641	3	-56 36	68 21	-56 19 -	56 47	68 15 -68 27	-56 50½	68 25
Λ	651	4	-50 28	70 00½	-50 10 -	50 41	69 45 -70 18	-50 45½	69 53½

TABLE 2b. MEASUREMENTS ON BOULANGERITE FROM IDAHO.

		No.	Meas. aver.		Range		Calculated	
			ϕ	ρ	ϕ	ρ	ϕ	ρ
<i>c</i>	001	3	88°58'	12°01'	88°58'–90°00'	10°42'–13°20'	90°00'	10°39½'
<i>a</i>	100	1	88 40	90 00	—	—	90 00	90 00
<i>P</i>	120	2	29 05	90 00	28 24–29 46	—	29 03½	90 00
<i>m</i>	110	8	48 23	90 00	47 41–49 03	—	48 01	90 00
<i>n</i>	210	3	66 11	90 00	65 03–67 00	—	65 46½	90 00
<i>d</i>	011	3	29 37	21 25	28 20–30 48	21 13–21 40	28 34½	21 29
<i>e</i>	021	1	16 27	41 42	—	—	15 14	35 47
<i>f</i>	031	6	10 33	46 32	9 44–11 29	46 06–46 54	10 17½	46 30
<i>x</i>	101	3	90 24	29 46	90 00–91 00	29 37–29 53	90 00	29 46½
<i>y</i>	201	2	–90 23	30 05	90 23–90 23	—	–90 00	30 06½
<i>p</i>	111	1	60 44	33 37	—	—	58 52	33 45½
<i>A</i>	221	1	54 14	49 14	—	—	54 08½	49 43
α	221	5	–39 57	41 32	–38 36–41 23	41 23–42 10	–39 59½	42 03½
<i>E</i>	121	5	39 56	41 39	39 42–40 14	41 06–42 00	39 37	41 54
ϵ	121	4	–15 22	35 38	–15 03–16 00	35 34–35 55	–15 49	35 41½
ϕ	131	4	–10 38	46 02	–10 15–11 19	45 08–46 00	–10 41½	46 32
<i>M</i>	211	2	69 43	45 48	69 21–70 05	45 30–46 06	70 07½	45 28½
μ	211	1	–56 14	34 48	—	—	–59 12	34 01
σ	311	1	–69 12	45 10	—	—	–70 16½	45 40½
τ	321	3	–54 19	49 34	53 31–55 06	49 10–49 52	–54 21	49 52
Λ	651	1	–51 17	69 50	—	—	–50 45½	69 53½

TABLE 3. ANGLE TABLE. BOULANGERITE

Monoclinic, prismatic $\frac{2}{m}$

$$a:b:c=0.9158:1:0.3456; \beta = 100^\circ 39\frac{1}{2}'$$

$$p_0:q_0:r_0=0.3774:0.3396:1; \mu = 79^\circ 20\frac{1}{2}'$$

$$r_2:p_2:q_2=2.9443:1.1111:1; p_0'=0.3840 \quad q_0'=0.3456 \quad x_0'=0.1882$$

Forms	ϕ	ρ	ϕ_2	$\rho_2=B$	C	A
<i>c</i> 001	90°00'	10°39½'	79°20½'	90 00'	—	79°20½'
<i>b</i> 010	0 00	90 00	—	0 00	90°00'	90°00'
<i>a</i> 100	90 00	90 00	0 00	90 00	79 20½	—
** <i>r</i> 140	15 31½	90 00	0 00	15 31½	87 52	74 28½
* <i>q</i> 130	20 19½	90 00	0 00	20 19½	86 19	69 40½
* <i>P</i> 120	29 03½	90 00	0 00	29 03½	84 51	60 56½

TABLE 3—Continued

Forms	ϕ	ρ	ϕ_2	$\rho_2=B$	C	A
* <i>m</i> 110	48 01	90 00	0 00	48 01	82 06	41 59
** <i>n</i> 210	65 46½	90 00	0 00	65 46½	80 17½	24 13½
** <i>l</i> 310	73 18	90 00	0 00	73 18	79 48	16 42
** <i>k</i> 410	77 19	90 00	0 00	77 19	79 36½	12 41
** <i>i</i> 510	79 48	90 00	0 00	79 48	79 30½	10 12
** <i>h</i> 710	82 40½	90 00	0 00	82 40½	79 26	7 19½
<i>d</i> 011	28 34½	21 29	79 20½	71 14½	18 45½	79 55
<i>e</i> 021	15 14	35 37	79 20½	55 48½	34 11½	81 12
<i>f</i> 031	10 17½	46 30	79 20½	44 28	45 32	82 33½
<i>g</i> 041	7 45	54 22	79 20½	36 21½	53 38½	83 42½
<i>j</i> 051	6 13	60 05½	79 20½	30 29½	59 30½	84 37
<i>o</i> 061	5 11	64 20½	79 20½	26 08½	63 51½	85 19½
** <i>u</i> 102	90 00	20 49	69 11	90 00	10 09½	69 11
<i>x</i> 101	90 00	29 46½	60 13½	90 00	19 07	60 13½
<i>y</i> 201	-90 00	30 06½	120 06½	90 00	40 46	120 06½
<i>p</i> 111	58 52	33 45½	60 13½	73 18	25 10½	61 35½
<i>A</i> 221	54 08½	49 43	46 17	63 27½	41 26	51 48½
<i>B</i> 331	52 16½	59 27	36 43½	58 12	51 17	47 04
<i>D</i> 441	51 16½	65 39	30 07	55 15½	57 33	44 42
<i>C</i> 551	50 39½	69 51½	25 22½	53 28½	61 47	43 26½
α 221	-39 59½	42 03½	120 06½	59 07½	49 27	115 30
β 331	-42 54½	54 45½	133 56½	53 15½	62 20½	123 47
δ 441	-44 16½	62 37	143 25½	50 31½	70 17	128 18½
<i>E</i> 121	39 37	41 54	60 13½	59 02½	35 53	64 47½
<i>F</i> 131	28 53½	49 49½	60 13½	48 01	45 23	68 20
<i>G</i> 141	22 29	56 14½	60 13½	39 48½	52 46½	71 27½
<i>L</i> 151	18 19½	61 13	60 13½	33 41½	58 23½	74 00½
ϵ 121	-15 49	35 41½	101 04½	55 51	39 45½	99 09
ϕ 131	-10 41½	46 32	101 04½	44 30	49 22½	97 44½
γ 141	- 8 03½	54 23½	101 04½	36 23½	56 53	96 33
λ 151	- 6 28	60 06	101 04½	30 31½	61 51	95 36
<i>M</i> 211	70 07½	45 28½	46 17	75 58½	36 03	47 53½
<i>N</i> 231	42 41	54 40	46 17	53 09	47 52½	56 25½
<i>R</i> 251	28 57½	63 08½	46 17	38 41	58 24½	64 24½
μ 211	-59 12	34 01	120 06½	73 21½	43 28½	118 43½
ν 231	-29 13	49 54½	120 06½	48 06½	55 40½	111 55½

TABLE 3—Continued

Forms	ϕ	ρ	ϕ_2	$\rho_2 = B$	C	A
θ 241	-22 45	56 17½	120 06½	39 54	60 56	108 46
ρ 251	-18 33	61 15	120 06½	33 47	65 05½	106 11½
S 311	75 32½	54 09	36 43½	78 49½	43 53	38 17½
T 321	62 43	56 27	36 43½	67 32½	47 09	42 12½
U 341	44 06½	62 33	36 43½	50 25	55 26½	51 51
V 361	32 52½	67 57	36 43½	38 53	62 29	59 47½
σ 311	-70 16½	45 40½	133 56½	76 01½	55 48	132 20
τ 321	-54 21	49 52	133 56½	63 32½	58 46	128 24½
ω 351	-29 09	63 11½	133 56½	38 47½	68 43½	115 46
W 411	78 40	60 22½	30 07	80 10	49 57	31 32
X 421	68 09½	61 42½	30 07	70 52½	51 54	35 11
Γ 411	-75 37	54 17½	143 25½	68 22	64 39½	141 52
Δ 421	-62 51	56 34	143 25½	67 37	66 10	137 57
Y 531	63 48½	66 56½	25 22½	66 02½	57 29	34 20½
Z 541	56 45	68 22	25 22½	59 21½	59 35½	38 59
Θ 631	-63 53½	67 00	154 42	66 06½	76 38½	145 45
Ω 641	-56 50½	68 25	154 42	59 25½	77 26	141 07
Λ 651	-50 45½	69 53½	154 42	53 33½	78 16	136 39½

* Forms also found by Sjögren.

** Forms found only by Sjögren.

Since the crystals are all so minute that the faces are practically invisible even under the binoculars, it was not deemed advisable to attempt to figure them, as drawings would only be conventional representations at best. Crystal eight from Montana with faces of 33 forms is by far the most complex one seen.

For the reason given in the last paragraph no attempt has been made here to characterize the individual forms and their relative importance must be judged solely by frequency of occurrence.

Table 2a contains a summary of the angular measurements obtained from the eight Montana crystals and Table 2b similar figures for those from Idaho. The Washington crystals were of far inferior quality and are not recorded. If there appears to be a somewhat wide range in the observed angles of individual forms, it must be remembered that all of these measurements were obtained from crystal facets so minute that often the signal was but the faintest spot of light. It was impossible to distinguish the faces on most of the crystals sufficiently clearly to recognize their symmetry relations. Only after projection could the orientation of the crystal be determined, and it was a matter of astonishment to find

how closely these weak reflections repeated on each new projection the pattern of the common gnomonic lattice. The Montana crystals gave, on the whole, the better reflections, but both sets of measurements were used to calculate elements with the following concordant results:

Boulangerite, Montana	$a:b:c=0.9158:1:0.3456; \beta=100^{\circ}39\frac{1}{2}'$
Boulangerite, Idaho	$a:b:c=0.9252:1:0.3437; \beta=100^{\circ}52'$

The first set of elements, based on measurements of 90 faces of 17 forms on 8 crystals, was accepted as best established and was used in the calculation of an angle table, Table 3.

Reference has been made earlier to the paper in which Sjögren first described crystals of boulangerite. His results have been correlated with our established position by the following transformation formula and they show excellent correspondence.

Transformation, Sjögren to Palache 041/800/002
 Elements of Sjögren, calculated to new position: $a:b:c=0.9315:1:0.3383; \beta=100^{\circ}27\frac{1}{2}'$

Four of his prisms were found on our crystals. Five others, probably very weak forms, were not found by us but are included in the angle table as well as his single terminal form, which becomes the dome {102}.

The measurements of Shannon (1921) could not be satisfactorily correlated with our position.

X-RAY CRYSTALLOGRAPHY

Dr. Hurlbut reports as follows on his study of *x*-ray photographs made in 1939 on crystals of boulangerite from Montana. From measurements of a rotation photograph: $d_{001}=8.00 \text{ \AA}$. From Weissenberg zero and second-layer photographs: $d_{010}=23.16 \text{ \AA}$ and $d_{100}=21.10 \text{ \AA}$. $\mu=79^{\circ}19'$ calculated from *x*-ray data. Hence, $a_0=21.47$, $b_0=23.16$, $c_0=8.00$.

$$a_0:b_0:c_0=0.9166:1:0.3475; \beta=100^{\circ}41' \text{ compared with}$$

$$a:b:c=0.9158:1:0.3456; \beta=100^{\circ}39\frac{1}{2}'$$

Space group $P2_1/a$ fixed by the conditions

- hkl all present
- $0k0$ present only with k even
- $h00$ present only with h even
- $h0l$ present only with h even

This cell taken with the measured specific gravity, 5.98, gives $M_0=14468$.

These results agree very closely with the measurements published by Berry (1940), which were made on boulangerite from Mullan Co., Idaho, the so-called mullanite for which the crystal measurements are given above.

CHEMICAL COMPOSITION

Boulangerite from Rocky Gulch, Montana, was analyzed by Gonyer on a carefully selected sample prepared by Dr. Berman. The density was determined on the microbalance as 5.98 ± 0.02 , the mean of many measurements by several observers. Gonyer also analyzed the Stevens Co., Washington, boulangerite and confirmed Shannon's analysis. This second analysis is interpreted in the following table prepared by Dr. Berman.

TABLE 4. ANALYSIS OF BOULANGERITE FROM WASHINGTON

	1	2	3	4	5
Pb	55.42	55.28	55.91	39.03	40
Sb	25.69	25.40	25.69	30.53	32
S	18.89	18.19	18.40	83.03	88
Fe		.39			
Insol.		.62			
Total	100.00	99.88	100.00		

1. $Pb_8Sb_4S_{11}$.
2. Boulangerite from Cleveland Mine, Stevens Co., Washington, U.S.N.M. 95414. *Pb* and *S* average of two determinations. F. A. Gonyer, analyst.
3. Column 2 recalculated to 100%.
4. Atoms per unit cell with $M_0 = 14468$ and $d = 5.98$.
5. $8(Pb_8Sb_4S_{11})$.

The experience of Mr. Gonyer in making this analysis proved that the complete separation of lead and antimony was not attained by some well-tried methods, leading to low results for lead. This observation suggests that possibly the numerous earlier analyses of boulangerite with low lead content may have been at fault through this determinative error. For instance, the so-called plumosite of English writers with a formula given as $Pb_2Sb_2S_6$ has physical characteristics differing in no way from boulangerite

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