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LAWSONITE AND PUMPELLYITE IN GLAUCOPHANE SCHIST, NORTH BERKELEY HILLS, CALIFORNIA

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WITH NOTES ON THE X-RAY CRYSTALLOGRAPHY OF LAWSONITE

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ABSTRACT. A previously undescribed lawsonite-pumpellyite locality in the North Berkeley Hills is characterized by exceptional development of lawsonite as veins and as lithologic layers in glaucophane schist. The glaucophane schist outcrop, a tectonic block in serpentinite, is believed to have been derived at least in part by retrogressive metamorphism of eclogite. Lawsonite-rich layers in the schists and the vein minerals lawsonite and pumpellyite are regarded as products of metamorphic differentiation under conditions of glaucophanitic metamorphism.

Optical and morphological data for lawsonite and pumpellyite are presented together with a new lawsonite chemical analysis and a semi-quantitative spectroscopic analysis of pumpellyite. A description is given of lawsonite-pumpellyite intergrowths and of pumpellyite twinning.

The cell dimensions of lawsonite from three California localities and from Cuba have been determined by means of calibrated Weissenberg patterns. All values obtained lie within the limits 8.788 ± 0.008 Å for a_o, 13.129 ± 0.010 for b_o and 5.846 ± 0.006 for c_o. A powder diffraction pattern giving calculated and observed *d* values to 1.46 Å has been tabulated. The thermal transformation of lawsonite to metastable hexagonal $CaAl_2Si_2O_8$ has been studied by means of single crystal diffraction. The change can be detected after prolonged heating at temperatures as low as 410°C. In the neighborhood of 700°C it is complete in an hour or less. At intermediate temperatures partly changed lawsonite may persist for long periods. Single crystal patterns are more sensitive than powder patterns for detecting the beginning and end of the transformation. Lawsonite that had been heated under a bonfire was found to be partly transformed.

INTRODUCTION

Glaucophane schists from the North Berkeley Hills and associated occurrences of lawsonite and pumpellyite have been described by Brothers (1954). One lawsonite-pumpellyite-glaucophane schist locality not observed by Brothers has been found on the private estate of Mrs. Anson Blake and the late Mr. Blake about one-eighth mile southeast of Brothers' locality number 4. This occurrence, although similar to others in the area, is characterized by an unusually fine development of lawsonite as veins and as lithologic layers parallel to the foliation of enclosing glaucophane schists. The locality is near the western edge of the estate, now the Blake Gardens, a research facility for the Department of Landscape Architecture of the University of California, and it and other outcrops of glaucophane schist on the grounds will be preserved.

Lawsonite and pumpellyite are found on the estate in a large block of glaucophane schist, 20 feet high and 25 feet across, apparently resting on serpentinite. The locality is probably within the Hayward fault zone, the main trace of which lies several hundred feet to the east. This schist block and others of similar or larger size in the area are regarded, in agreement with Brothers (1954, p. 616), as tectonic inclusions within serpentinites of the fault zone, although it is impossible to ascertain whether the Blake Gardens block is now *in situ.* Several other outcrops of serpentinite and glaucophane schist on the estate will not be described in this paper.

Occurrences in North Berkeley of lawsonite in tectonic inclusions of glaucophane schist are somewhat similar to the occurrence of this mineral in chlorite schist inclusions in serpentinite of Santa Clara province, Cuba (Schürmann, 1936). Exogenic blocks of eclogite and actinolite schist are found in the serpentinites of both areas. Glaucophane is not found in the lawsonite-bearing rocks of the Cuban association, but it does occur in some of the eclogites (Schürmann, 1936, p. 246).

GLAUCOPHANE SCHISTS

The glaucophane schists of the lawsonite-bearing block are fine-grained and have a well-defined foliation. They comprise three mineral associations:

1) glaucophane-sphene-(chlorite)-(colorless mica)
2) glaucophane-lawsonite-(sphene)
3) glaucophane-epidote-sphene

Anhedral apatite and spheroids of pyrite up to 2 cm in diameter are common accessory minerals of all associations. Small, ragged, often turbid grains of a pale green pyroxene believed to be relict are found in some specimens of the glaucophane-sphene and glaucophane-lawsonite assemblages.

The relict pyroxene occurs as individual grains and as aggregates parallel to the schistosity. It is consistently associated with a colorless mica, probably muscovite ($2V_x = 37°$-$44°$), and a pale green chlorite. An X-ray powder pattern of this mineral prepared by Dr. A. Pabst is identical with that of an acmitic diopside-jadeite from Healdsburg, California. The optics of this pyroxene are within the ranges described by Borg (1956, p. 1571) for the Healdsburg mineral: $2V_z = 76°$-$82°$ (Borg, 70°-82°) and $c \wedge Z = 49° \pm 1°$ (Borg, 44°-50°). Both Brothers (1954) and Borg (1956) consider glaucophane schists containing relict pyroxenes of this type to have been derived retrogressively from eclogite. Garnets were not found in schists of the pyroxene-bearing associations.

Glaucophane crystals commonly show color zoning, the intensity of color increasing outward in distinct zones from the crystal core. Typical pleochroic values are X = colorless to pale yellow, Y = violet to dark violet, and Z = sky blue to indigo blue. Universal stage measurements in sodium light give $2V_x$ variable between 35° and 45°. Dispersion is strong, $r<v$.

The epidote mineral pistacite occurs in amounts up to 25 percent by volume in some of the glaucophane schists. It is found as individual subhedral crystals and less commonly in aggregates of grains around which the foliation is deflected. The cores of most crystals are pleochroic in shades of green conforming to the pleochroic scheme X = colorless, Z = colorless to very pale apple green, and Y = pale apple green. Optic angle is negative and varies between 68° and 74°. Sections cut at high angles to an optic axis exhibit anomalous blue and yellow interference colors; dispersion $r > v$ is moderate. The pistacite crystals, elongated parallel to Y, are often simply twinned on the composition plane {100}.

Anhedral to subhedral sphene grains are present in quantities up to 10 percent by volume in all of the glaucophane schists examined. These grains have a maximum size of 1 mm and are dichroic from tan to light brown; a small percentage exhibit elbow-shaped contact twins.

Lawsonite-bearing glaucophane schists (association 2) appear to be a variant of the first association resulting from crystallization of lawsonite aggregates along surfaces of schistosity. Alternation of light and dark-colored layers in the resulting rocks gives them a distinctly gneissic appearance. Lawsonite was not observed as discrete rock-forming grains within the glaucophane schists of this locality. The lawsonite-rich folia, discontinuous in lateral extent and 0.1 to 3.0 cm thick, are regarded as products of either metamorphic segregation or hydrothermal deposition, rather than a reflection of initial compositional layering. A total of 1450 point counts across a lawsonite-rich layer 1.3 cm wide gives the following mode computed in weight percent:

lawsonite	71.3	pyroxene	1.3
glaucophane	19.7	muscovite	0.1
pyrite	6.2	apatite	0.1
sphene	1.3		100.0

The above mode is considered representative of such layers, which are coarse-grained enough to permit megascopic comparison of the proportions of white lawsonite and blue-black glaucophane grains within them. From this mode an approximate chemical composition (table 1) for the lawsonite-rich band can be calculated by using chemical analyses of Berkeley Hills glaucophane and lawsonite (this paper). Pyrite is not included within this computation as it appears to be a late-stage post-lawsonite mineral.

TABLE 1
Calculated chemical composition of lawsonite-glaucophane layer in glaucophane schist, Blake Gardens

SiO_2	41.9
TiO_2	0.9
Al_2O_3	25.5
Fe_2O_3	2.8
FeO	2.6
MnO	0.1
MgO	1.6
CaO	14.3
Na_2O	1.5
K_2O	0.2
H_2O	8.6
	100.0

It is unlikely that the chemical composition of table 1 with its low silica, high lime-alumina content represents that of an original layer in the premetamorphic rock. As these glaucophane-lawsonite layers are cut by pumpellyite and lawsonite-pumpellyite veins of the variety to be described, their origin is probably best explained by a process of metamorphic differentiation occurring prior to the formation of the veins. It seems likely that the veins themselves are products of such a differentiation, perhaps a more mobile, aqueous culmination of the process giving rise to the lawsonite-glaucophane "gneisses".

VEIN MINERALS

The glaucophane schists just described are host to several generations of the vein minerals lawsonite, pumpellyite, and calcite; albite veinlets are present, some containing vugs lined with small euhedral crystals, but they are not common. With one notable exception veins are 0.2 to 1.5 cm wide and can be grouped into two sets governed by cross-cutting joints. An early set contains pumpellyite only or pumpellyite as the predominant vein mineral. These veins are consistently crosscut by a second set perpendicular to them in which lawsonite is generally the predominant mineral. Both sets contain calcite as the last mineral to be deposited. Textural evidence indicates deposition of the minerals in open spaces and by limited replacement of earlier vein minerals.

The exception noted above is a large lens-shaped vein, predominantly lawsonite, not related to or found cross-cutting either of the two joint sets described. This vein is unusual in its dimensions (maximum width 9 inches) and the large size of certain lawsonite crystals within it (2 to 3 inches in length). Lawsonite makes up most of the central portion of the vein and is bordered by a discontinuous pumpellyite zone up to 4 cm wide; pumpellyite also occurs as irregular patches within the lawsonite core. Several fragments of glaucophane schist completely enclosed by lawsonite and pumpellyite were found within the vein.

Lawsonite.—Lawsonite from the large vein is seen in thin section as anhedral crystals generally within the size range 0.2 to 2.0 cm. These crystals, tabular parallel to $\{010\}$, have maximum elongation parallel to c $(=X)$. They possess perfect $\{010\}$, well-developed but discontinuous $\{100\}$, and imperfect $\{101\}$ cleavages. The larger crystals are invariably strained and exhibit marked undulatory extinction and polygonization of the crystal into smaller, discretely bounded areas of variable extinction (1 to 20° variance within single crystals). Polysynthetic $\{101\}$ and $\{\bar{1}01\}$ twins are well developed only in those crystals lacking pronounced strain effects $(X\Lambda\{101\} = 34°)$. The Blake estate lawsonite is slightly altered to a dusty unidentified phase spatially related to lawsonite cleavages. Calcite is associated with lawsonite as cavity fillings between anhedral-subhedral crystals, as veinlets cross-cutting lawsonite, and as minor localized patches replacing that mineral. Pumpellyite and lawsonite are frequently intergrown so that X of lawsonite is parallel to Y $(=b)$ of pumpellyite (plate 1, fig. 1). This relationship has been previously described by Coombs (1953, p. 120) in terms of close correspondence between the a and b cell dimensions of lawsonite and pumpellyite, and consequent parallelism in intergrowth of their a and b axes ($a_L \,||\, a_P$ and $b_L \,||\, b_P$, or in the settings

PLATE 1

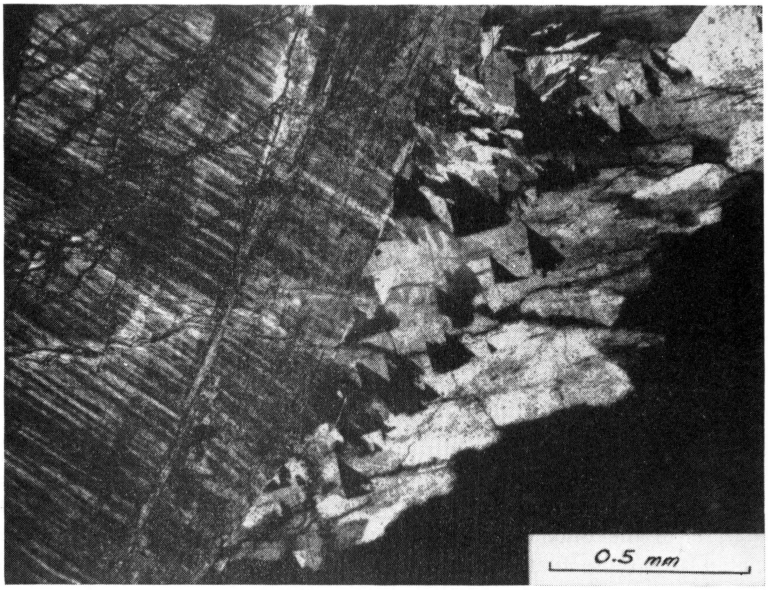

Fig. 1. Scalene-shaped intergrowths of pumpellyite (black) in lawsonite. Lawsonite with polysynthetic twinning on left. Crossed nicols.

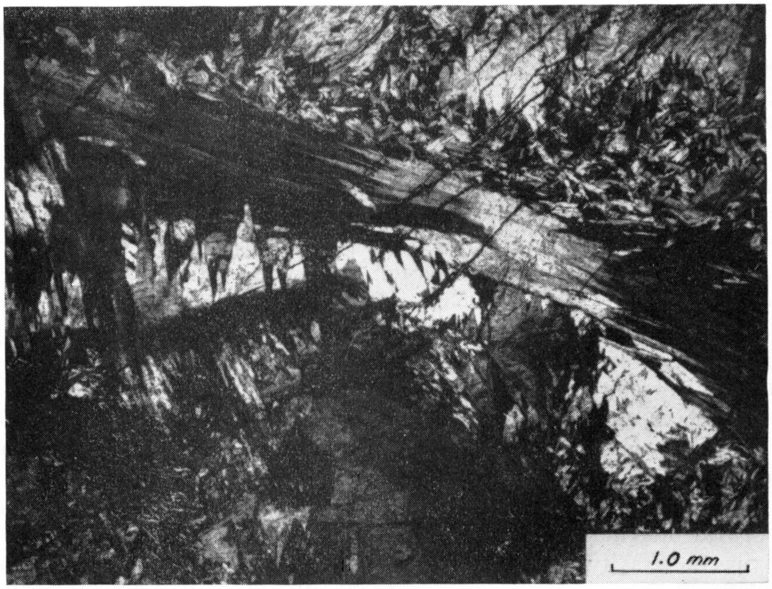

Fig. 2. Sheath of pumpellyite crystals in upper center of picture with "oak-leaf twins" below. Crossed nicols.

adopted in this paper $a_L \parallel a_P$ and $c_L \parallel b_P$). The sequential relationships between lawsonite and pumpellyite are not always clear; in some cases, however, the intergrowth cited above is due to replacement of lawsonite by pumpellyite.

Optical properties of vein lawsonite from the Blake Gardens are summarized in table 2. These values agree quite closely with optical data on lawsonite from other localities.

TABLE 2

Optical properties, lawsonite, Blake Gardens

Orientation:	X=*c*, Y=*a*, Z=*b*
n_x	1.665 ± .002
n_y	1.673 ± .002
n_z	1.686 ± .002
n_z-n_x	0.021 ± .004
$2V_{zNa}$	79-85°
Disp.	r>v, weak
Sp.G.	3.07 ± 0.01

A comparison of four superior chemical analyses of lawsonite, including an analysis of the Blake Gardens mineral, is presented in table 3. Formulae calculated from these analyses to conform to the simplified formula $CaAl_2Si_2O_8 \cdot 2H_2O$[1] illustrate extremely limited ionic substitution within the lawsonite structure.

Intergrown pumpellyite crystals are usually in the shape of a scalene triangle with interfacial angles, as measured by universal stage on a true {010} section of 97°, 128° and 135°. These angles correspond closely with calculated interfacial angles given by Coombs (1953, p. 117) for (001) Λ (100), (001) Λ ($\bar{1}02$), and ($\bar{1}02$) Λ (100) respectively and support his contention that the bounding faces for such scalene-shaped intergrowths are {001}, {100}, and {$\bar{1}02$} forms (1953, p. 127). However, variability of interfacial angle between the forms {100}? and {$\bar{1}02$}? noted by Coombs was also found in pumpellyite from the Blake Gardens, and over the same range (135-140°). Furthermore the interfacial angle between (001) and the face believed to be (100) is more consistently on the order of 92-93° than the 97° measured in one true {010} section. No reasons for such interfacial variability other than those suggested by Coombs (1953, p. 128) are offered here.

The scalene-shaped intergrowths may occur as isolated individuals (plate 1, fig. 1) or as series of back-to-back crystals usually separated by a narrow septum of lawsonite intergrown as previously described. The term "oak-leaf twin" has been applied by Coombs to back-to-back individuals in which {001} is a plane of symmetry between opposite crystals (plate 2, fig. 2). In some instances, however, back-to-back crystals in specimens from the Blake Gardens are not in twin relationship to each other, i.e. the crystallographic orientation in opposed individuals on either side of a lawsonite septum is the same.

Specific gravity of the Blake Gardens pumpellyite is 3.16 ± 0.01. Optical properties, in summary, are: $n_{yNa} = 1.689$; $2V_{zNa} = 51°\text{-}52°$; $Z\Lambda c = 8°$; X

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[1] Structural analysis by Wickman (1947) gives the formula $CaAl_2Si_2O_7(OH)_2 \cdot H_2O$.

and Z = colorless, Y = bright apple green; dispersion r<v is strong. Coombs (1953, p. 130-131) has demonstrated that the optical properties are correlative with iron content. Plottings of n_y, $2V_z$, and $Z\Lambda c$ against total iron content of 5.4 percent as Fe_2O_3 (see table 4) give points which fall on total iron-optics curves presented by Coombs.

Pumpellyite.—In the Blake Gardens, pumpellyite in the glaucophane schists examined is restricted to veins, although it is found as a rock-forming mineral elsewhere in the North Berkeley Hills (Brothers, 1954, p. 619). It is found as either bundle-like or matted aggregates of bladed crystals up to 2 cm long (plate 1, fig. 2), often with calcite or chlorite filling crystal interstices (plate 2, fig. 1), or as intergrowths with lawsonite as described above.

TABLE 3

Chemical analyses of lawsonite with corresponding atomic ratios

	A.	B.	C.	D.
SiO_2	38.14	38.81	37.80	38.45
Al_2O_3	30.91	32.02	32.56	31.35
Fe_2O_3	1.56	0.21	0.05	0.86
TiO_2	0.29	0.12	0.69	0.38
FeO	0.13	0.28	0.34	0.10
MnO	0.01	tr.	tr.	tr.
MgO	0.03	0.16	0.32	0.17
CaO	17.54	17.81	16.10	17.52
Na_2O	0.06	0.18	0.51	0.06
K_2O	0.14	0.21	0.24	0.23
H_2O	10.72	10.67	11.26	11.21
H_2O	0.00	0.36	0.04	
F_2	—	—	0.02	—
	99.53*	100.83	99.93	100.33

* Total loss on ign. = 11.40; theoretical H_2O_{total} = 11.42.

Atomic ratios calculated on basis of O = 8.

	A.		B.		C.		D.	
Si	2.014		2.009		1.978		2.008	
Al	1.914	1.987	1.954	1.966	2.007	2.036	1.929	1.978
Fe'''	0.062		0.008		0.002		0.034	
Ti	0.011		0.004		0.027		0.015	
Fe''	0.006	0.008	0.011	0.023	0.015	0.040	0.004	0.017
Mg	0.002		0.012		0.025		0.013	
Ca	0.988	1.003	0.988	1.019	0.902	0.970	0.980	0.997
Na	0.006		0.018		0.052		0.006	
K	0.009		0.013		0.016		0.011	
H_2O	1.887		1.844		1.964		1.946	

A. $(Ca_{0.988}Na_{0.006}K_{0.009})(Fe''_{0.006}Mg_{0.002})(Al_{1.914}Fe'''_{0.062}Ti_{0.011})Si_{2.014}O_8 \cdot 1.887\ H_2O$

B. $(Ca_{0.988}Na_{0.018}K_{0.013})(Fe''_{0.011}Mg_{0.012})(Al_{1.954}Fe'''_{0.008}Ti_{0.004})Si_{2.009}O_8 \cdot 1.884\ H_2O$

C. $(Ca_{0.902}Na_{0.052}K_{0.016})(Fe''_{0.015}Mg_{0.025})(Al_{2.007}Fe'''_{0.002}Ti_{0.027})Si_{1.978}O_8 \cdot 1.964\ H_2O$

D. $(Ca_{0.980}Na_{0.006}K_{0.011})(Fe''_{0.004}Mg_{0.013})(Al_{1.929}Fe'''_{0.034}Ti_{0.015})Si_{2.008}O_8 \cdot 1.946\ H_2O$

A. Blake Gardens, North Berkeley Hills, California. Doris Thaemlitz, anal.

B. Kanto Mountainland, Japan. Seki, 1957, p. 372. Y. Seki, anal.

C. Santa Clara province, Cuba. Schürmann, 1936, p. 250. van Tongeren, anal.

D. Tiburon Peninsula, Marin Co., California. Schaller and Hillebrand, 1904, p. 197. W. T. Schaller, anal.

PLATE 2

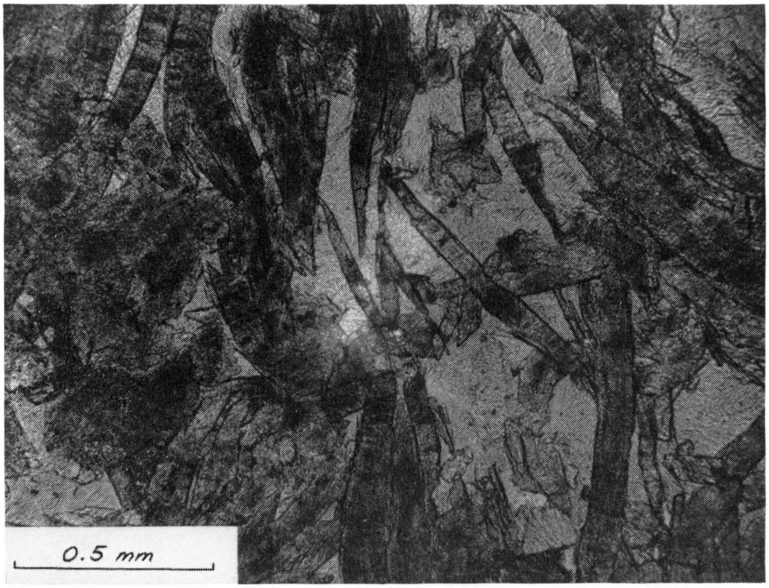

Fig. 1. Bladed pumpellyite crystals with interstitial chlorite.

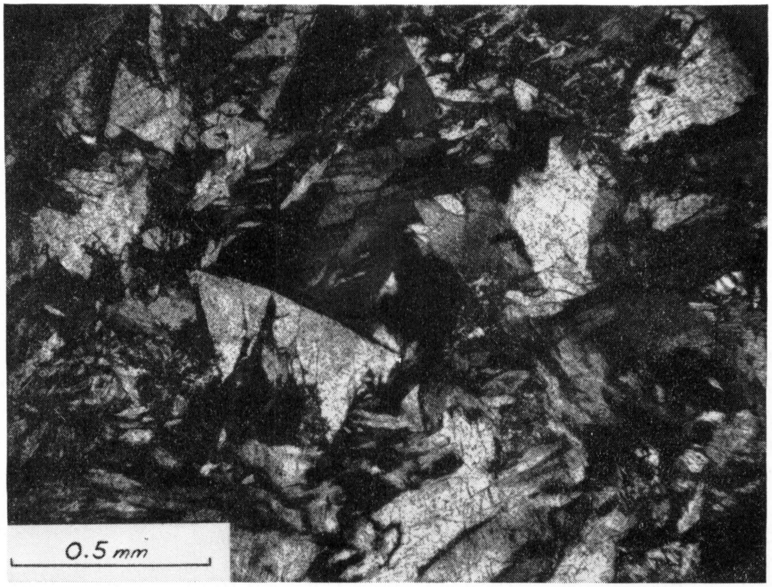

Fig. 2. "Oak-leaf twin" in pumpellyite. Crossed nicols.

TABLE 4

Semi-quantitative spectroscopic analysis, pumpellyite, Blake Gardens

Principal constituents: Ca, Al, Si

Fe_2O_3	5.4%*	Na_2O	0.1-0.2
TiO_2	0.5	V_2O_5	0.1
Cr_2O_3	0.01	Ni	0.002
MgO	5.0	Ga	0.005
MnO_2	0.1-0.5		

K, Zr, Cu, Ge, and Sr below level of sensitivity

* Quantitative determination, total iron expressed as Fe_2O_3. Average of two (5.1 and 5.7%).

Analyst, George M. Gordon

Pumpellyite crystals in specimens from the Blake Gardens are elongated parallel to *b* (=Y) and are sometimes tabular parallel to {100}. Some crystals, diamond-shaped in {010} section, appear to be bounded by poorly developed {100} and {$\bar{1}$02} or {102} forms. Strong dispersion of the optic axes ($2V_{z_r} =$ 51°-52°, $2V_{z_v} =$ 57°-59°) results in anomalous blue and brown interference colors in sections cut at high angles to the optic plane.

DISCUSSION

Field relationships of glaucophane schist in the Blake Gardens and elsewhere in the North Berkeley Hills impose restrictions on petrogenetic interpretations pertaining to the schists themselves and the lawsonite and pumpellyite within them. The reader is referred to the paper by Brothers (1954) and a discussion of it by de Roever (1955) for opposing views on the origin of these schists. The present study, confined to the tectonic block on the western border of the Blake Gardens, suggests: 1) derivation of at least part of the glaucophane schists from eclogite; 2) formation of layered lawsonite-glaucophane rock by metamorphic differentiation; and 3) an alternating sequence of lawsonite and pumpellyite deposition in dilation fractures, these minerals being deposited from solutions derived by metamorphic differentiation accompanying glaucophanitic metamorphism.

ACKNOWLEDGMENTS

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NOTES ON THE X-RAY CRYSTALLOGRAPHY OF LAWSONITE

MATERIAL

Lawsonites from four localities, showing a variety of habits and associations, have been examined by single crystal and powder X-ray diffraction.

X-ray observations were also carried out on single crystals and powder after heat treatment. The following lawsonites were used:

1. from the type locality, Tiburon Peninsula, Marin County, California. (Ransome, 1895 and Schaller and Hillebrand, 1904)
2. from the Blake Gardens, Contra Costa County, California. (Occurrence described above by Mr. Davis)
3. from veins and cracks in low-grade metamorphic rock about $2\frac{1}{2}$ miles northeasterly from Valley Ford, Sonoma County, California. (Some of the Franciscan rocks in this area have been described by Bloxam (1956) and the geology has been mapped by Travis (1952).)
4. from the Province of Santa Clara, Cuba. (Described by Schürmann (1936). Specimen kindly furnished by Dr. Schürmann.)

SETTINGS

Various settings have been used for lawsonite. The original setting chosen by Ransome (1895) was followed by Schaller and Hillebrand (1904) and by Rogers (1915). Gossner and Mussgnug (1931), who were the first to report X-ray observations on lawsonite, retained Ransome's orientation but found it necessary to double the *c* axis. Strunz (1937) chose a new orientation but adopted yet another setting in his *Mineralogische Tabellen* (1941 and later editions). This later Strunz setting was followed by Wickman (1947) who summarized the relations of the several settings in connection with his determination of the structure of lawsonite and by Seki (1957). Table 5 gives the transformations between all of the settings. In this report the setting chosen by Strunz in 1937 will be used. It conforms to the convention $c<a<b$ and has been used by Donnay (1951) and by Switzer (1951) who presented an angle table for lawsonite with indices referred to this setting.

TABLE 5

Lawsonite transformations

to	Ransome 1894	Gossner & Mussgnug 1931	Strunz 1937	Strunz 1941
from Ransome 1894		100/010/002	010/002/100	010/$\bar{1}$00/002
Gossner & Mussgnug 1931	100/010/00½		010/001/100	010/100/001
Strunz 1937	001/100/0½0	001/100/010		100/001/0$\bar{1}$0
Strunz 1941	010/$\bar{1}$00/00½	010/100/001	100/001/0$\bar{1}$0	

HABIT

The habit of lawsonite from the type locality has been pictured by Ransome (1895) and by Schaller and Hillebrand (1904). All crystals from this source examined during the current study were found to be short prismatic or nearly equidimensional, {101} usually being the dominant form. This is in contrast to the habit of lawsonite in the other specimens examined. In all of these the crystals, though sometimes in compact masses and sheaflike aggregates, are invariably elongated in the direction of the *c* axis (*b* axis of Wickman) as would be expected from the cell dimensions and structures. About a dozen crystals were measured. All faces observed are in the zone of the *c* axis. Generally a near continuum of reflections is observed over much of the *hk*0 zone but no indices other than those of forms listed by Switzer, {100}, {010} and {210}, could be surely established. No crystals with *b* axis elongation such as shown by Switzer (1951, fig. 2) were encountered.

CELL DIMENSIONS

The cell dimensions of lawsonite from the four localities were determined by means of quartz-calibrated Weissenberg patterns. All results are shown in table 6. It is clear that any variations in the dimensions are very slight. The average values with the stated limits can be considered to apply to any of these lawsonites. In the lower part of table 6 are given the cell dimensions previously reported for lawsonite. Wickman (1947) examined lawsonite from the type locality and it may be assumed that Gossner and Mussgnug (1931), who did not state the provenance of their material, obtained it from the same source. The marked differences in the dimensions for lawsonite reported by these observers from the newly determined dimensions are puzzling. Seki (1957) examined lawsonite from the Kanto Mountainland, Japan. The small differences between the cell dimensions he reported and those newly determined may be due to real differences in the materials.

TABLE 6

Cell dimensions of lawsonite

	a_0	b_0	c_0
Tiburon	8.786		5.848
xl. #2		13.134	5.851
Blake Gardens	8.787	13.127	
xl. #5		13.127	5.842
Valley Ford	8.793		5.848
xl. #3	8.786	13.134	
Cuba		13.127	5.842
xl. #3	8.785	13.126	
average	8.788 ± 0.008	13.129 ± 0.010	5.846 ± 0.006 Å
Gossner & Mussgnug (1931)	8.87	13.25	5.88 Å *
Wickman (1947)	8.90	13.33	5.76 Å *
Seki (1957)	8.75	13.09	5.84 Å

* Converted to Å units from the values originally reported in kX units.

POWDER PATTERN

The only published records of X-ray powder diffraction patterns of lawsonite known to the writer are that of Seki (1957) with observations in terms of 2θ and ASTM card 8-137. In table 7 is given the record of concurrent measurements, over the same angular range as given by Seki, on powder patterns of the three California lawsonites examined, presented in the conventional manner in terms of d-spacings and indexed to conform to the setting here chosen. To convert to the indexing of Seki, which is also given, k and l must be interchanged. Intensities were estimated by superposition of a scale of spots of graded intensities. Except for minor differences in resolution and the recording of some very weak reflections the agreement with Seki's report is excellent. However, Seki indexed lines at 2θ 31.36° and 37.04°, equivalent to spacings 2.852 and 2.429, 021 and 023 respectively. This is permissible in the space group $C222_1$ chosen by Wickman, but lawsonite shows false symmetry due to the special values of parameters of all atoms in general positions (Pabst, 1959) which results in zero intensity for reflections of this type. The space group symbol in the new orientation is $B22_12$ and the symbol of the space group simulated due to the false symmetry is $B\ 2/b\ 2_1/m\ 2/m$. The indexing has been checked for every recorded line by comparison with precession or Weissenberg patterns. (See postscript.)

HEATED LAWSONITE

A few yards from the lawsonite outcrops in the Blake Gardens some rock fragments resembling lawsonite vein material but brownish in color were found in a spot where there was evidence of old bonfires. A small crystal selected from such a specimen was examined by means of an oscillation pattern. This showed the normal diffraction spots for lawsonite plus a great number of strong powder arcs which were readily identified as due to the metastable hexagonal modification of $CaAl_2Si_2O_8$ (Davis and Tuttle, 1952). Upon heating this partly changed crystal to 760°C it was completely converted to hexagonal $CaAl_2Si_2O_8$. Goldsmith and Ehlers (1952) have reported that this phase is produced when lawsonite is heated in air for 24 hours at 650°C, that anorthite begins to form after 24 hours at 800°C and that the conversion can be completed at 1,050°C in 4 hours or less. Similar results were obtained by Douglass (1954). Goldsmith and Ehlers stated that "the lowest temperature at which lawsonite decomposes in air in a reasonable time has not yet been established." Switzer (1951, fig. 3) published a differential thermal curve for lawsonite and stated "A large endothermic peak at 641°C and a smaller one at 744° indicate loss of water at two different temperatures." A differential thermal curve of lawsonite from the Blake Gardens, kindly run by Mr. Larry Godwin, shows similar features.

TABLE 7

X-ray powder diffraction pattern of lawsonite

$c<a<b$ *hkl*	$b<a<c$ *hkl*	$d_{calc.}$	$d_{obs.}$*	I	$c<a<b$ *hkl*	$b<a<c$ *hkl*	$d_{calc.}$	$d_{obs.}$*	I
020	002	6.56	6.58	3½	113	131	1.883	1.884	1½
101	110	4.87	4.84	6	351	315	1.854	1.852	½
111	111	4.56	4.57	3	123	132	1.827		
200	200	4.39		3	440	404	1.826		
210	201	4.17	4.18	4	252	225	1.785	1.786	½–
121	112	3.91	3.91	½	402	420	1.756		
220	202	3.65	3.66	6	062	026	1.752	1.752	3½
040	004	3.28			171	117	1.750		
			3.26	3½	133	133	1.745		
131	113	3.25			412	421	1.741		
230	203	3.10			270	207	1.725	1.726	2
002	020	2.923	2.925	4½	422	422	1.696	1.697	1½
141	114	2.721	2.726	7	450	405	1.685	1.685	½
022	022	2.670	2.680	5	501	510	1.683		
240	204	2.629			361	316	1.679		
			2.624	10	511	511	1.669		
301	310	2.619		10				1.657	½
311	311	2.579	2.573	½	143	134	1.646		
202	220	2.434			080	008	1.641		
			2.433	6	521	512	1.630	1.631	3
321	312	2.432			432	423	1.630		
212	221	2.393	2.395	3	262	226	1.627		
151	115	2.311	2.311	1½				1.626	4
222	222	2.282	2.284	2	303	330	1.622		
250	205	2.254	2.254	3½	313	331	1.610		
331	313	2.247			323	332	1.575	1.575	1–
400	400	2.197			531	513	1.571		
			2.190	1½B	181	118	1.555		
060	006	2.188			460	406	1.550	1.550	8
042	024	2.182			442	424	1.548		
410	401	2.167			153	135	1.541		
232	223	2.127	2.129	6	280	208	1.537		
420	402	2.083	2.089	½	371	317	1.525		
341	314	2.047	2.053	½	333	333	1.521		
161	116	1.996	1.990	½	541	514	1.499	1.501	1
430	403	1.963	1.965	3	272	227	1.486		
260	206	1.959			600	600	1.483		
242	224	1.955			004	040	1.461	1.464	4
103	130	1.902			452	425	1.460		

* Cu radiation, Ni filter; Camera diameter 114.59 mm, Straumanis setting, Measurements corrected for film shrinkage.

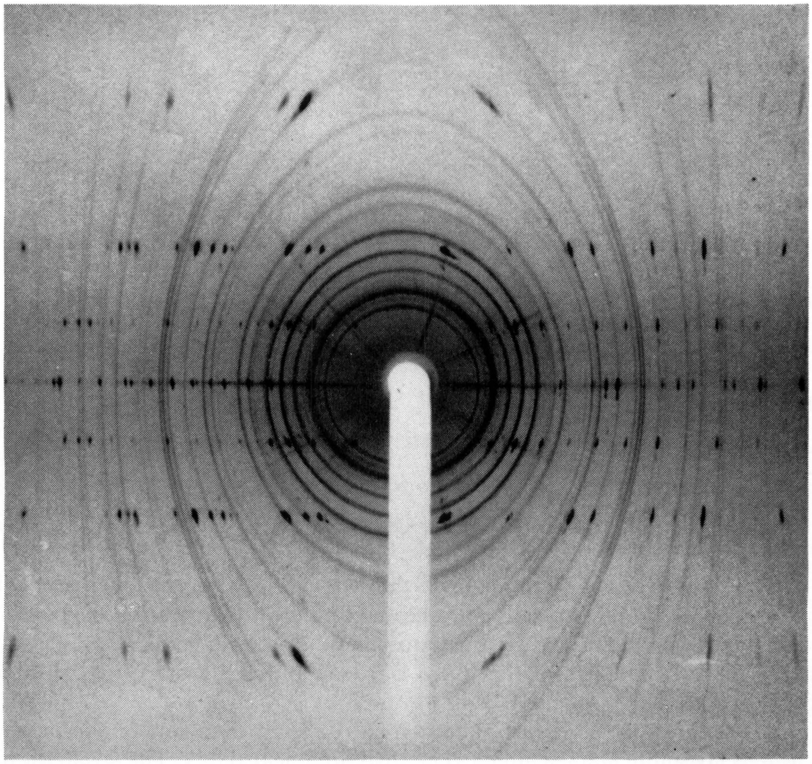
Some observations were made on the sequence of changes when lawsonite is heated in air. Single crystals were heated to known temperatures for various periods and then examined by *c* axis oscillation patterns after each period of heating. Five crystals from the Blake Gardens and one from Cuba, all about 0.5 x 0.1 x 0.1 mm, were used.

Besides the powder lines of hexagonal $CaAl_2Si_2O_8$ a few of the patterns of heated lawsonite show a single very weak line near 3.2 Å. The strongest line of orthorhombic $CaAl_2Si_2O_8$ is at 3.21 (Davis and Tuttle, 1952) and the strongest anorthite line at 3.20 Å. Since anorthite is formed only at higher temperatures (Goldsmith and Ehlers, 1952, p. 395) it seems reasonable to attribute this line to a minute amount of orthorhombic $CaAl_2Si_2O_8$ in those cases where it appears in patterns obtained from crystals heated to 500°C or less. In the one case in which it appeared in a pattern of material heated to 700°C. it might be attributed to anorthite. Some of the patterns of material heated for very long periods or at rather high temperatures (670 to 760°C) show a weak, diffuse line near 3.35, close to the position of the strongest quartz line, but one would hardly expect the appearance of quartz and so this line remains unexplained.

Heatings were carried out for periods from half an hour to five days and at temperatures from 410° to 760°C. The results may be summarized as follows:

A. At 410°C. After 16 hours no change can be observed in a single crystal by oscillation pattern. After 40 hours the strongest powder arcs due to hexagonal $CaAl_2Si_2O_8$ can be seen.

PLATE 3



c axis oscillation pattern of lawsonite crystal from Blake Gardens after heating for 154 hours at 450°C. Cu radiation, Ni filter, 6-hour exposure.

B. At 450°C. After 3 hours no change is discernible in a single crystal. After 16 hours a few powder arcs are superposed on a sharp single crystal pattern. Lawsonite ground to less than 300 mesh and heated for 16 hours at this temperature is not sufficiently transformed to allow detection by powder pattern. After heating the powder for a total of 34 hours at this temperature the three strongest lines of the dehydration product are discernible in the pattern among the full complement of lawsonite lines. A single crystal heated at this temperature in several increments for a total of 154 hours yields the complete hexagonal $CaAl_2Si_2O_8$ powder pattern superposed on a lawsonite oscillation pattern of sharp spots with intensity scarcely diminished (pl. 3).

C. At 555°C. A crystal heated at 3 hours to this temperature after previous heatings of 3 hours at 455° and 15 plus 21 hours at 515°C shows only the last traces of the lawsonite single crystal pattern with the hexagonal $CaAl_2Si_2O_8$ pattern but the few remaining spots are sharp, though weak.

D. At 580°C. The same crystal was completely converted by further heating for 16 hours.

E. At 690°C. A single crystal after 35 minutes yields a complete, very sharp, hexagonal $CaAl_2Si_2O_8$ powder pattern and only a few weaks spots of a lawsonite oscillation pattern. Powdered lawsonite after the same period of heating yields no trace of unchanged material by X-ray pattern.

F. At 700°C. A single crystal is completely converted in an hour and a half or less. The initially clear crystal is porcelaneous in appearance after conversion.

These observations show that the thermal decomposition of lawsonite to hexagonal $CaAl_2Si_2O_8$ begins very slowly at 410°C or less, at least 230° below the most prominent endothermic DTA peak at 641°C, and proceeds rapidly, being complete in approximately an hour, in the neighborhood of 700°C well below the second endothermic DTA peak at 744°C. The first stages of conversion can be recognized by oscillation patterns on single crystals when they are not detectable by powder pattern. Initially clear crystals show only the faintest milkiness in these stages. In the range from about 410 to about 550°C lawsonite crystals are converted very slowly or maybe only partly, the change being incomplete even after heating for several days.

Postscript (*February 5, 1960*).—After this paper had been accepted for publication our attention was drawn to a paper on "The crystalline structure of lawsonite" by I. M. Rumanova and T. I. Skipetrova (1959 Doklady, Akad. Nauk, S. S. S. R., v. 124, p. 324-327, [GeoScience Abstracts, abstract 1-3099, Dec. 1959 and Chem. Abst. 53, 9914*e*, June 10, 1959]). These authors report the cell dimensions $a = 8.83$ Å, $b = 5.80$, $c = 13.20$, using the setting of Strunz (1941) and Wickman. They arrive at a structure in the space group *Ccmm*, which requires the systematic absences that Pabst (1959) attributed to the false symmetry associated with Wickman's structure in the space group $C222_1$. See also "Considerations on false symmetry in the structure of lawsonite" by I. M. Rumanova and N. V. Belov, 1960, Kristallografiya, v. 5, p. 215-217.

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