Augite from Bail Hill, Dumfriesshire.

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THE crystals to which this paper refers are found at Bail Hill, about three miles north of Sanquhar, in Dumfriesshire. At this locality volcanic rocks of Arenig age outcrop in the midst of the greywacke of the Southern Uplands. The rocks are mainly lavas, and include some coarse-grained tuffs which are composed chiefly of augite- and hornblende-andesites. In the andesitic fragments, there are numerous porphyritic crystals of a black augite, which can be easily separated from the rest of the rock. Isolated crystals of augite and hornblende occur also in the matrix of the tuffs. No mention of this occurrence is made in Heddle's 'Mineralogy of Scotland', and apparently the only reference to it in previous literature is the short petrographic description in the Geological Survey Memoir.[1] There, Teall mentions the occurrence of sharp crystals of augite showing the common forms (100), (010), (110), (Ī11). The mineral is described as yellow to yellowish-green in colour, slightly pleochroic, and containing some inclusions of magnetite.

The crystals are black in colour, fairly opaque, and have the usual well-developed cleavage parallel to the prism-faces. The forms observed are few in number, comprising only the four mentioned above, namely, a (100), b (010), m (110), and s (Ī11), while there is nothing unusual in the developments of these forms in the two types which are commonly found. The first type consists of untwinned crystals with an approximately equal development in the three axial directions, and varying in size up to 1·5 centimetres in diameter. The faces in the prism-zone have a bright lustre, but the pyramid-faces are usually dull. With the exception of some very small crystals, most of the specimens of this kind show a curious deformation. The b- and m-faces are perfectly plane,

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[1] 'Silurian Rocks of Scotland,' Mem. Geol. Survey, 1899, pp. 87, 306.

but the a-faces are convex in the plane parallel to b (010), the degree of convexity being a maximum near the ' acute angle '[1] of the crystal, and a minimum at the ' obtuse angle '. The face, of course, tends to broaden out as it curves inwards, and on the whole somewhat resembles a portion of a parabolic cylinder, having as its axis the b-axis of the crystal. The s-faces are correspondingly concave, as are also the s-s edges, which are often replaced by a curved surface at the ' acute ' contact with the a-face, especially in those crystals which are highly distorted.

The second type is composed of crystals elongated parallel to the prism-faces, and invariably twinned on a (100). As before, the faces in the prism-zone are bright and the s-faces ' matte ', but in this case the distortion is either very slight or absent. In both types intergrowths both of twinned and untwinned crystals are very common, and occasionally as many as twelve crystals are found thus intergrown. The individuals of these groups are arranged sometimes in a roughly stellate fashion (as figured by Rosenbusch [2]), and sometimes with the respective prisms more or less parallel. In one example, the upper half is a twin of the type described above, while the lower half is untwinned and composed of a single crystal, but the habit is different from the crystals of this kind described by Williams from Orange County, New York.[3]

It was found impossible to measure the larger crystals accurately, on account of the curvature of the faces, but a number of small crystals, up to 1·5 mm. in diameter, gave consistent readings, though the faces were so dull that a film of silver had to be chemically deposited on them in order to obtain good reflections. Five crystals were measured, and the results are given below.

		Calculated.	Observed.	No.	Limits.
am	$= (100) : (110)$	46° 30′	46° 28′	12	46° 21′–46° 38′
bm	$= (010) : (110)$	*	43 30	12	43 17 –43 42
ab	$= (100) : (010)$	90 0	89 58	1	— —
mm	$= (110) : (\bar{1}10)$	87 0	—	—	— —
sm	$= (\bar{1}11) : (\bar{1}10)$	*	59 7	4	58 48 –59 26
ms	$= (110) : (\bar{1}11)$	78 41	78 52	2	78 38 –79 6
bs	$= (010) : (\bar{1}11)$	*	60 36	5	60 25 –60 46
ss	$= (\bar{1}11) : (1\bar{1}1)$	58 48	58 41	2	58 34 –58 48

The axial ratios are :—

$$a : b : c = 1\cdot0932 : 1 : 0\cdot5844; \; \beta = 74° 12'.$$

[1] i. e. the acute angle between a and the s-s edge.

[2] H. Rosenbusch, Mikroskopische Physiographie, 1905, vol. i (2), p. 207, Pl. VI, fig. 3.

[3] G. H. Williams, Amer. Journ. Sci., 1887, ser. 3, vol. xxxiv, p. 275.

In thin section, the augite is yellowish-green in colour (in sections 0·04 mm. thick) with slight pleochroism, the absorption for $\mathfrak{a}$ and $\mathfrak{c}$ being the same, while that for $\mathfrak{b}$ is slightly greater. In a section of a thickness of 0·5 mm., the difference in absorption is very marked, the colours being $\mathfrak{a}$ and $\mathfrak{c}$ yellow-green, $\mathfrak{b}$ deep red-brown.

A section in the symmetry-plane shows the hour-glass structure so characteristic of augites containing titanium. In general there are four sectors, each triangular in shape and having a common point in the centre of the crystal. Two of these have the trace of the a-face as their external boundary, and the other two the s-s edge or the trace of an s-face. Sometimes the sectors do not meet in the centre, which is then occupied by a non-sectored core approximately rhomb-shaped, as shown in fig. 1. A section through the centre of the crystal and perpendicular to the prism-faces shows eight sectors, two bounded by the a-faces and lines joining the a-m angles to the centre, two by the b-faces and corresponding lines, and the remaining four by the m-faces and lines to the centre. The divisions separating the a-sectors from the others are generally well-defined, but those separating the b- and m-sectors are often indefinite, and can only be recognized in very thick sections. Fig. 2 shows the arrangement in such a section. Sections parallel to a (100) show different arrangements according as they are through the centre of the crystal, or between there and the a-face. In the former, four sectors are seen, two bounded externally by the b-faces, and the other two each by two contiguous s-faces. In the latter the trace of an a-sector appears in the middle, the size varying with the distance from the centre. It is six-sided, the sides being parallel to, and in the same ratio (in size) as, the boundaries of the a-face. The remainder of the section is occupied by the s- and m-sectors (as in fig. 3), unless the section is near the centre of the crystal, when part of the b-sectors appear in addition.

Considering now the distribution of the various sectors in three dimensions, it is obvious that the a-sectors each consist of a pyramid with a hexagonal base, the a-face; the b-sectors each of a pyramid with a rectangular base, the b-face; the m-sectors each a pyramid with a trapezohedral base, the m-face; while the two s-sectors occupy the remainder of the crystal. In none of the sections could any indication be obtained that each of the s-sectors was divided into two, one for each face. If they were so divided the optical properties of both parts were apparently the same. The optical properties of the a- and s-sectors were determined, but not those of the others. It is most probable that

the b- and m-sectors resemble closely the s-sectors, as in some of the sections cut, these three sectors could not be distinguished optically from each other.

The various sectors are not homogeneous in themselves, but show a number of parallel bands resembling multiple-twinning. This is only occasionally visible in ordinary light, but it can generally be seen when the section is between crossed nicols, through a few degrees on each side of the extinction position. The bands in each case are parallel to the base of the pyramid which composes the particular sector, but are not
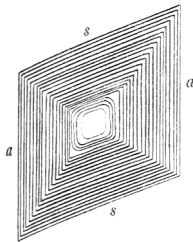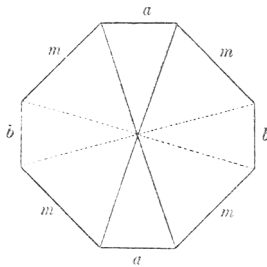

Sections of Augite crystals.

Fɪɢ. 1.—Parallel to b (010), through centre of ' cored ' crystal and showing 'periodic' banding.

Fɪɢ. 2.—Perpendicular to prism-zone, through centre of non-cored crystal.

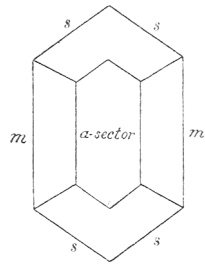Fɪɢ. 3.—Parallel to a (100), through m-faces.

equally distinct in all the sectors, being more pronounced in the a- and s-sectors than in the b-sectors, while they are often difficult to detect in the m-divisions. The extinction varies continuously from one band to the next (like the 'shadowy' extinction of strained plagioclase) and the maximum variation in any one sector is less than $3°$. Where a central core exists it is not, in general, homogeneous, but shows a pronounced zonal structure, the extinction varying as in the sectors.

This structure bears a close resemblance to those obtained by 'periodic crystallization', such as Liesegang's phenomenon.[1] There seems to be the same alternate deposition of two layers differing in composition, though in the one case the layers are spherical, in the other, plane. It is difficult to see any reason for this periodicity, as there is no evidence to show that titanaugites crystallize from more viscous solutions (magmas) than any other pyroxene, nor, so far as is known, is there any greater tendency for supersaturation to occur.[2]

[1] R. E. Liesegang, ' Ueber die Schichtungen bei Diffusionen,' Leipzig, 1907.
[2] The most generally accepted explanation of Liesegang's phenomenon is that

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This hour-glass structure is common to most of the augites which contain titanium oxide, and it was first described by Werveke.[1] The explanation first given by Rosenbusch[2] was that the crystals developed in such a manner that a section in the *b*-plane showed a kind of bifurcated structure and that the pyramids at the *a*-faces were filled up later. On the other hand, many mineralogists maintain that the various sectors all grow more or less at the same rate, just as any ordinary crystal. In the present case the latter is the most plausible, as some of the bands seem to pass from one sector to the next. The crystals probably developed by the addition of successive series of layers, the latter varying somewhat in composition, and the deposition of the layers taking place more or less uniformly on all the faces of the crystals.

With regard to the different sectors, the difference in optical properties (see below) would argue a difference in chemical composition. Since the structure is apparently confined to pyroxenes containing a fair amount of titanium oxide,[3] it is reasonable to assume that the titanium content varies in the different sectors. But whether it is the amount of titanium alone that varies or that of some complex molecule containing titanium, must remain a matter of conjecture, as nothing is known, at present, about the condition of the titanium in these pyroxenes. In connexion with this structure, some results of Gaubert's work may be significant. He found[4] that the habit of crystals of hydrated phthalic acid was greatly modified by the presence of substances, such as dyes, in the solution, and further, that such crystals, growing from a solution containing methylene blue, were coloured blue and showed an hour-glass structure, the pyramids with (011) as their base being more intensely coloured than those with (210) or (212) as base. Crystals of meconic acid[5] show a more pronounced difference between the colour of the various sectors, the prism (110) being the base of the most intensely coloured sector. (Meconic acid crystallizes in the orthorhombic system.) A curious coincidence is that these crystals showed a much greater

due to Ostwald (Zeits. Physik. Chem., 1897, vol. xxiii, p. 365), who attributes it to supersaturation.

[1] L. van Werveke, Neues Jahrb. Min., 1879, pp. 482, 882.

[2] H. Rosenbusch, Mikroskopische Physiographie, 1905, vol. i (2), p. 210.

[3] J. Blumrich (Min. Petr. Mitt., 1892, vol. xiii, p. 239) was the first to notice that this hour-glass structure is practically confined to augites in alkali-rich rocks. A summary of the different occurrences is given by Pelikan (Min. Petr. Mitt., 1896, vol. xvi, pp. 1-64).

[4] P. Gaubert, Bull. Soc. franç. Min., 1905, vol. xxviii, p. 286.

[5] P. Gaubert, Compt. Rend. Acad. Sci. Paris, 1910, vol. cli, p. 1134.

tendency to develop curved faces than those prepared from pure solution. It seems feasible, as indeed Gaubert has suggested,[1] that the structure of titanaugites arises in some similar way and, as has been indicated above, the development of the hour-glass structure seems to be connected with the presence of titanium. Complete analyses of the various sectors would probably assist much in the explanation of this phenomenon, but so far this has not been found possible.

The *extinction-angles* given below are the means for the respective sectors and were measured from the trace of the twinning-plane in a section of a twin-crystal cut parallel to b. The plane of the optic axes is the symmetry-plane, and $\mathfrak{b} = b$. The values of $c : \dot{c}$ were determined for thallium-, sodium-, and lithium-light, a Hilger monochromatic illuminator being used for the production of the light. (This was used for all the optical properties.)

λ.	a-sector.	s-sector.
Li-light 6710	$c : \dot{c} = 47° 48'$	45° 2'
Na-light 5890	„ = 48 32	45 42
Tl-light 5350	„ = 49 19	46 19

Refractive Indices.—For the determination of the refractive indices, six prisms were prepared, all with a refracting angle of about 40°. With any higher angle the minimum deviation could not be found exactly, owing to the strong absorption. Two of the prisms were cut so that each gave a and γ for the a-sector, while other two, from the same crystal, gave a and γ for the s-sector. One of the remaining two gave β and an index near γ for the a-sector, and the other the corresponding values for the s-sector. The values obtained for thallium- and sodium-light are given below :—

	a.	β.	γ.	
a-sector	1·708	1·713	1·732	Na-light.
	1·708		1·732	
	1·713	1·718	1·737	Tl-light.
s-sector	1·697	1·702	1·722	Na-light.
	1·696		1·721	
	1·703	1·708	1·727	Tl-light.
	1·702		1·727	

Consistent values were obtained, the variations from the mean being always less than 0·001 for any one prism.

[1] P. Gaubert, 'Recherches récentes sur le Facies des Cristaux,' 1911, p. 26.

The mean values of the double-refraction are:—

		$\gamma-a.$	$\gamma-\beta.$	$\beta-a.$	
a-sector	{	0·024	0·019	0·005	Na-light.
	{	0·024	0·019	0·005	Tl-light.
s-sector	{	0·025	0·0195	0·0055	Na-light.
	{	0·0245	0·019	0·0055	Tl-light.

In order to determine the *optic axial angles,* four sections, two perpendicular to the acute bisectrix and two perpendicular to the obtuse, were prepared, and the apparent acute and obtuse angles measured by immersing the section-plates in a-monobromonaphthalene of refractive index 1·665. The values of β obtained from the prisms were used in calculating the true angles in the results tabulated below:—

	Section.	Sector.	2H(Na).	2H(Tl).	2V$_a$(Na).	2V$_a$(Tl).
Acute angle	1.	a	64° 32′	64° 5′	62° 30′	61° 52′
	2.	a	64 18	63 55	62 18	61 44
	2.	s	63 28	62 53	61 59	61 6
Obtuse angle	3.	a	123 58	123 47	61 52	62 34
	3.	s	123 22	123 20	61 8	61 42
	4.	a	122 28	123 6	63 6	62 54

The average values are:—

a-sector $2V_a(Na) = 62° 34′$ $\qquad 2V_a(Tl) = 62° 16′$

s-sector $\qquad\quad = 61\ 30$ $\qquad\qquad\quad = 61\ 24$

The values calculated from the mean values of the acute and obtuse angles for the a-sector are:—

$$2V_a(Na) = 62° 26′$$
$$2V_a(Tl) = 62\ \ 2$$

and the values of β are:—

$$\beta(Na) = 1·712 \qquad \beta(Tl) = 1·713,$$

which agree fairly well with the values obtained from the prisms. In the case of the s-sectors the agreement is not so good, the values being:—

$$\beta(Na) = 1·708 \qquad \beta(Tl) = 1·705\ [1]\ \text{as compared with}$$

the prism values $\beta(Na) = 1·702 \qquad \beta(Tl) = 1·708.$

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[1] The differences between the values of β for the s-sector as calculated from the axial angle and as observed by the prism method are probably to be explained by the fact that section 3 contained b- and m-sectors in addition to a and s, and it is very difficult to distinguish those sectors from the s. The prism values of β are certainly the more reliable.

The dispersion of the bisectrices is fairly strong, so that a section parallel to b never shows complete extinction between crossed nicols in white light. The axial dispersion is slight and is $\rho > v$.

Chemical Composition.—As it was found impossible to separate the sectors and analyse them individually, a bulk analysis was made of a sample composed of several crystals. Column I in the following table gives the percentage by weight, and column II the molecular percentage, water being neglected in the calculation.

	I.		II.
SiO_2	45·66	... ...	47·71
TiO_2	2·16	... ...	1·76
Al_2O_3...	3·53	... ...	2·13
Fe_2O_3...	8·78	... ...	3·45
FeO	4·16	... ...	3·64
MnO	0·19	... ...	0·12
MgO	9·34	... ...	14·79
CaO	22·17	... ...	24·75
Na_2O	1·50	... ...	1·50
K_2O	0·26	... ...	0·15
H_2O above 110° ...	1·37	... ...	—
H_2O below 110° ...	0·61	... ...	—
P_2O_5	trace [1]	... ...	—
	99·73		100·00

If the mineral is assumed to consist of the molecules aegirine-acmite —Na (Fe, Al) Si_2O_6,—diopside-hedenbergite—Ca (Mg, Fe, Mn) Si_2O_6,— and Tschermak's hypothetical silicate—Mg (Al, Fe)$_2$$SiO_6$—the composition by weight is 11·8 per cent. of the first of these, 73·0 per cent. of the second, and 15·2 per cent. of the third.

The determination of the specific gravity of the powdered mineral by a pycnometer method gave a mean value of 3·291.

Numerous attempts have been made to correlate the optical properties of the pyroxenes with their chemical composition, and various rules have been formulated. Tschermak [2] stated that the optical properties varied with the composition, the amount of iron oxides being a potent factor in

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[1] Less than 0·006 per cent.

[2] G. Tschermak, Min. Mitt., 1871, p. 21.

the non-aluminous pyroxenes. Wiik[1] found that in the Finnish diopsides the extinction-angle increased in a somewhat irregular fashion with the amount of ferrous oxide, while Doelter[2] formulated the law that the extinction-angle increased with the sum of the ferrous and ferric oxides and alumina, the increase being more regular if the amounts of the different metasilicates were considered in place of the oxides. Thus when the ratio of the iron-bearing metasilicates is less than 1 : 4 the extinction is less than 41°; when the ratio is between 1 : 4 and 1 : 3 the extinction varies from 41° to 45°, and when it is greater than 1 : 3 the extinction is greater than 45°. The extinction also seems to increase with the amount of alumosilicate and ferrisilicate molecule. In the present case the extinction varies from 45° to 49°, and the ratio of calcium-ferrous metasilicate to the other metasilicates is approximately 1 : 4, the amount of Tschermak's silicate being about 15 per cent. While comparison is difficult owing to the non-homogeneity of the Bail Hill crystals, it will be seen that they agree approximately with Doelter's rule.

Most of the values for the refractive indices given by Hintze[3] are lower than those obtained in this instance. The indices would seem to increase with the amount of the hedenbergite molecule. Michel Lévy and Lacroix[4] found refractive indices for two augites from Auvergne which resemble our values fairly closely :—

		α.	β.	γ.
Auvergne ...	...	{1·712	1·717	1·733
		{1·706	1·712	1·728
Bail Hill ...	...	1·708	1·713	1·732

Tschermak[5] described a mineral from the Bohemian Mittelgebirge with the optical properties :—

$$c : \dot{c} \; 45° \; 30', \; \beta = 1·70, \; 2V = 61°.$$

(Cf. analysis V in table below).

These minerals, however, were not analysed. Wülfing[6] investigated crystals with a composition similar to those of Bail Hill (Analysis VI in

[1] F. J. Wiik, Zeits. Kryst. Min., 1884, vol. viii, pp. 208-210.

[2] C. Doelter, Neues Jahrb. Min., 1885, vol. i, p. 56.

[3] C. Hintze, 'Handbuch der Mineralogie,' 1897, vol. ii, pp. 1025-1029.

[4] A. M. Lévy and A. Lacroix, 'Les Minéraux des Roches,' 1888, p. 265.

[5] G. Tschermak, Min. Mitt., 1871, p. 29.

[6] E. A. Wülfing, Min. Petr. Mitt., 1896, vol. xv, pp. 29-48.

table given below), and found the following values for the optical constants :—

$a(D) = 1.6975$　　$\beta(D) = 1.7039$　　$\gamma(D) = 1.7227$　　$2V(D) = 61° 22'$
$a(E) = 1.7029$　　$\beta(E) = 1.7093$　　$\gamma(E) = 1.7284$　　$2V(E) = 60\ 45$

which are close to the values found above for the s-sector :—

$a(Na) = 1.696$　　$\beta(Na) = 1.702$　　$\gamma(Na) = 1.722$　　$2V(Na) = 61° 30'$
$a(Tl)\ = 1.703$　　$\beta(Tl)\ = 1.708$　　$\gamma(Tl)\ = 1.727$　　$2V(Tl) = 61\ 24$

The following table gives the analyses of some augites which have approximately the same composition as the Bail Hill mineral :—

		I.	II.	III.	IV.	V.	VI.
SiO_2	...	45.66	45.52	47.76	45.83	44.18	49.72
TiO_2	...	2.16	2.05	1.24	3.57	—	—
Al_2O_3	...	3.53	5.48	4.21	7.47	8.00	4.23
Fe_2O_3	...	8.78	7.42	7.28	4.90	11.00	4.74
FeO	...	4.16	2.96	2.65	4.11	3.50	5.13
MnO	...	0.19	—	—	—	—	0.50
MgO	...	9.34	12.09	12.12	10.92	10.57	23.19
CaO	...	22.17	22.48	22.18	22.83	23.22	12.49
Na_2O	...	1.50	0.91	1.60	—	—	—
K_2O	...	0.26	0.35	0.36	—	—	—
H_2O	...	1.98	0.37	0.13	—	—	—
P_2O_5	...	trace	0.71	0.71	—	—	—
		99.73	99.63	100.22	99.63	100.47	100.00

I. Bail Hill.
II. Liebhards, Rhön Mtns.
III. Gehülfensberg, Rhön Mtns.} Analysed by Galkin.[1]
IV. Brown-violet augite from a tephrite ; Kaiserstuhl, Baden.[2]
V. Augite from tuff : Boreslau, Mittelgebirge, Bohemia.　Analysed by Schmidt.[3]
VI. Augite from Renfrew.　Described by Wülfing.[4]

Analyses II and III represent crystals from a basalt-tuff, which bears some petrological resemblance to the Arenig tuffs of Bail Hill.

[1] X. Galkin, Neues Jahrb. Min., 1910, Beil.-Bd. xxix, pp. 681–718.
[2] Quoted in H. Rosenbusch, 'Elemente der Gesteinslehre,' 1910, 3rd edit., p. 445.
[3] W. B. Schmidt, Min. Petr. Mitt., 1882, vol. iv, p. 14.
[4] E. A. Wülfing, loc. cit.

The first three analyses show several striking similarities, particularly in the silica and titanium oxide content, the amount of sesquioxides and the lime-magnesia ratio.   They differ from analysis IV, which represents a typical titanaugite from a strongly alkaline rock (tephrite), in two significant ways; namely, they contain less titanium oxide, and ferric oxide preponderates over alumina.   In practically all the analyses of titanaugites from basic rocks of pronounced alkaline affinities, such as teschenites, theralites, tephrites, &c., the amount of titanium oxide is over 8 per cent., and alumina preponderates over ferric oxide.   The Bail Hill rocks are also alkalic, as the dominant felspars are albite and soda-orthoclase, but they are not so strongly alkalic as the analcite, nepheline, and leucite rocks mentioned above.   This lesser degree of alkalinity seems to be reflected in the composition of the augites, which are practically intermediate between the non-titaniferous varieties of the calcic rocks and the so-called titanaugite of the alkalic rocks.

In conclusion, I wish to express my indebtedness to Mr. B. K. N. Wyllie for the loan of many crystals, and especially to Professor H. L. Bowman for his stimulating interest and advice throughout the course of this work.

Since the above work was done, it has been found possible to determine the refractive indices in the b- and m-sectors.   The values obtained were :—

	$a.$	$\beta.$	$\gamma.$	
b-sector	1·712	1·717	1·735	Na-light.
	1·715	1·720	1·738	Tl-light.
m-sector	1·710	1·715	1·734	Na-light.
	1·712	1·717	1·736	Tl-light.

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