

*Augite from Nishigatake, Japan.*

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THE following study was carried out in the Mineralogical Laboratory of the University of Cambridge upon specimens which were brought by myself from Japan. My hearty thanks are offered to Professor W. V. Lewis and Dr. A. Hutchinson for their kind permission to use the laboratory and the instruments, and for their constant help throughout my work.

The crystals of augite from Nishigatake,<sup>1</sup> province of Hizen, Kiushū, are found loose in decomposed basalt. They are small, up to 7 or rarely 10 millimetres in length, by not less than one half of this in thickness. In colour they are olive-green to almost black. The cleavage is rather indistinct. In section, the colour is pale-green. Pleochroism is scarcely discernible. No indications of zonal structure are observed. Inclusions are very abundant. Most of them are brown glass which looks sometimes almost black owing to the presence of minute opaque particles. These inclusions are not, however, objectionable for the chemical analysis, because of their solubility in hot hydrochloric acid. Magnetite is rarely present as inclusions in the crystals.

The specific gravity is 3.344 at 15.7° C., as determined, on the selected fragments used for the analysis, by hydrostatic weighing in a small glass-tube as devised by Professor Penfield,<sup>2</sup> a method which is capable of very accurate results. Correcting this value for the temperature and for the buoyancy of air, the value 3.338 is obtained for 4° C.

*Crystallography.*

The forms observed are  $a(100)$ ,  $b(010)$ ,  $c(001)$ ,  $m(110)$ ,  $o(221)$ ,  $p(101)$ ,  $s(111)$ ,  $z(021)$ . The faces  $a$ ,  $b$ , and  $m$  are smooth and shining,

<sup>1</sup> A brief description and figures of the crystals from this locality are given in T. Wada, *Minerals of Japan* (English translation), 1904, pp. 125-126 and fig. 53; second edition (in Japanese), 1916, p. 204, and figs. 4 and 5 on p. 201.

<sup>2</sup> S. L. Penfield, *Zeits. Kryst. Min.*, 1896, vol. 26, p. 136.

*o* and *z* are rough, *s* is very rough and sometimes curved, *c* and *p* are very rough and curved. Among these faces *a*, *b*, *m*, *s* are always present and well developed, *o* is very common and quite narrow, *z* is often present and small, while *c* and *p* are rare and indistinct. The crystals show fairly sharp edges except that between *s* and *s'* which is always rounded. They assume three habits, (1) stout prismatic, (2) prismatic along the axis *c*, and (3) prismatic along the zone-axis *b s*. The first type is the commonest. Only the faces of the prism-zone give single bright reflections of the goniometer-signal. Nine selected crystals were measured with the theodolite-goniometer and the following values were obtained.

Face	Refl.	Number of measurements.	$\phi$	$\rho$
<i>a</i> (100)	f	12	89° 58'	90° 0'
<i>b</i> (110)	f	18	0 0	"
<i>m</i> (110)	g	20	43 35	"
<i>o</i> ( $\bar{2}21$ )	b	14	35 16½	55 34
<i>s</i> ( $\bar{1}11$ )	b	20	24 37	33 18½
<i>z</i> (021)	p	16	14 5½	50 41½

The qualities of the reflections are distinguished as:—g good, f fair, p poor, and b bad.

The interfacial angle *m*:*m'''* observed is 92° 49½' (mean of 20 measurements), which agrees closely with that of diopside given in Dana's 'System' (92° 50'); while the angle *s*:*s'* is 59° 54' (mean of 20 measurements), which is greater than that of diopside (59° 11', Dana).

Twinning about *a* (100) is very frequent, and the twinned crystals amount to more than one-half of the whole number of specimens. Most of the twinned crystals assume an arrow-head form, but a few of them do not show any re-entrant angles and are apparently single. Very rarely the crystals appear to be single when viewed from one end, while they are seen to be twinned when viewed from the other end. In the last case we have a crystal which shows only two faces *s* ( $\bar{1}11$ ) and *s'* ( $\bar{1}\bar{1}1$ ) at one end of the vertical axis, while the other is terminated by two pairs of these faces in the twinned position. Such a peculiar development of faces is, in the present case, the result of twinning of two individuals about the plane *a*. Faces apparently single are really composite, one half of each face belongs to the other individual and has a different index. Thus the apparent face *s* really consists of two faces, namely *s* ( $\bar{1}11$ ) of one crystal and *s* (011) of the partner. On cutting

a section from the crystal and examining it under the microscope, one sees that the contact-plane runs through the middle of the crystal from one end to the other, and that the crystal consists of two halves of nearly equal size in twinned position. The angular relations of pairs of faces such as  $s(\bar{1}11) e(011)$ ,  $c(001) p(\bar{1}01)$ ,  $o(\bar{2}21) \mu(121)$ ,  $z(021)$ ,  $\epsilon(\bar{1}21)$ , &c., are so close that they cannot be distinguished even by very accurate measurements, unless the optical orientation is taken into account. That this is so can be seen from the following values of the angles for diopside taken from the standard works of Dana and of Hintze.

$e(011): e'(0\bar{1}1) = 59^\circ 6'$	$s(\bar{1}11): s'(\bar{1}\bar{1}1) = 59^\circ 11'$
$e(011): a(100) = 76 16$	$s(\bar{1}11): a'(\bar{1}00) = 76 34$
$c(001): a(100) = 74 10$	$p(\bar{1}01): a'(\bar{1}00) = 74 30$
$c(001): m(110) = 79 9\frac{1}{2}$	$p(\bar{1}01): m'(\bar{1}10) = 79 23$
$z(021): a(100) = 79 36$	$\epsilon(\bar{1}21): a'(\bar{1}00) = 79 49$
$z(021): b(010) = 41 24$	$\epsilon(\bar{1}21): b(010) = 41 22$

If we adopt another orientation<sup>1</sup> in which the axis  $a$  is chosen so as to make the angle  $\beta$  nearly  $90^\circ$ , the pairs of faces  $se$ ,  $cp$ ,  $z\epsilon$ ,  $qF$ , &c., can be expressed by similar indices; and if the axial ratios selected be  $a:b:c = 1.0507:1:0.5893$ ,  $\beta = 89^\circ 50'$ , then  $s = (\bar{1}22) e = (\bar{1}22)$ ,  $c = (102) p = (\bar{1}02)$ ,  $z = (142) \epsilon = (\bar{1}42)$ ,  $q = (502) F = (502)$ . Such is one of the advantages of this orientation, which is rather preferable to the usual one.

G. H. Williams<sup>2</sup> suggested that monoclinic pyroxene crystallizes in the domatic class, by reason of the hemimorphic development of the faces of certain crystals from Orange County, N.Y., and some other localities. However, the crystals discussed by him can be explained in four ways (fig. 1), viz. (i) as a polysynthetic twin in which alternate lamellae are of imperceptible thickness towards the opposite ends of the crystal; (ii) as a contact-twin, composed of two individuals in juxtaposition, as proved in the case of the Nishigatake augite; (iii) as a single crystal which shows unequal development of faces at both ends,

<sup>1</sup> This method of orientation was first proposed by Koksharov and afterwards suggested by vom Rath and by Tschermak, and adopted by P. Groth in his 'Tabellarische Übersicht der Mineralien' (3rd edit., 1889). See N. I. Koksharov, *Mém. Acad. Sci. St. Pétersbourg*, 1865, ser. 7, vol. 8, no. 12, pp. 53-69; G. vom Rath, *Ann. Phys. Chem. (Poggendorff)*, 1874, suppl. vol. 6, pp. 338-342; G. Tschermak, *Miner. Mitth.*, 1871, vol. 1, p. 18; J. Götz, *Zeits. Kryst. Min.*, 1885, vol. 11, pp. 241-245.

<sup>2</sup> G. H. Williams, *Amer. Journ. Sci.*, 1887, vol. 34, p. 275; and 1889, vol. 38, p. 115.

but possessing the highest degree of symmetry of the monoclinic system ; and (iv) as a single crystal which belongs to the domatic class of that system.

The etch-figures produced by hydrofluoric acid on the faces of the prism-zone show that the crystals of the Nishigatake augite possess the symmetry of the monoclinic holosymmetric class, as shown in the

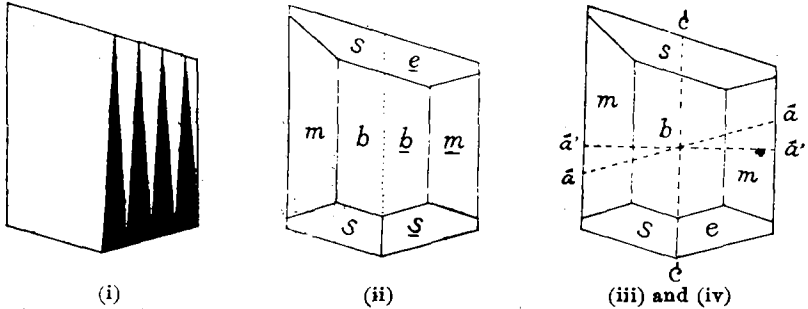


FIG. 1.—Alternative explanations of the apparent hemimorphic development of crystals of augite.

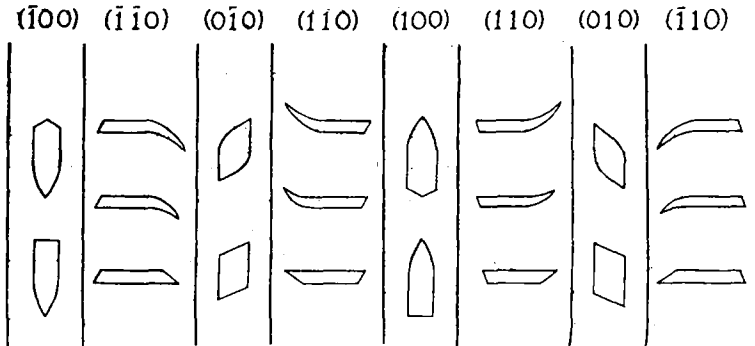


FIG. 2.—Etch-figures on faces in the prism-zone of augite from Japan.

accompanying drawing. G. Greim <sup>1</sup> observed the etch-figures produced by hydrofluoric acid on the diopside crystals from Ala, Piedmont, which show the same degree of symmetry. An early observation made by H. Baumhauer <sup>2</sup> of etch-figures on the *b* face of diopside shows also no

<sup>1</sup> G. Greim, Neues Jahrb. Min., 1889, vol. 1, p. 252 and pl. 4.

<sup>2</sup> H. Baumhauer, Ann. Phys. Chem. (Poggendorff), 1874, vol. 153, p. 75 and pl. 1, fig. 11.

indication of a lower grade of symmetry. W. H. McNairn<sup>1</sup> has studied recently the growth of etch-figures. Although his experiments were not specially made with the object of determining the symmetry, his results obtained on diopside crystals agree with those obtained by previous observers. The hemimorphism of pyroxene suggested by Williams, therefore, appears not to be well founded.

*Optical properties.*

As the interior of the crystals is crowded with inclusions, the determination of the three principal indices of refraction was made on a natural crystal-face *b*, which was 3 × 4 mm. in size, by means of total reflection using the Hutchinson universal goniometer<sup>2</sup> arranged as a Kohlrausch total-reflectometer.

The crystal was mounted, under the microscope, on a pin placed parallel to the vibration-direction  $\gamma$  of the crystal, adjusted so as to be capable of rotation about this direction, and was employed for the determination of two of the principal indices  $\beta$  and  $\gamma$ . After this, the crystal was remounted parallel to the vibration-direction  $\alpha$ , and then  $\beta$  and  $\alpha$  were determined.

The observed critical angles and the computed indices are given in the following table.

Critical angles observed.	Temperature.	$\mu_D$ of Methylene iodide.	Principal indices of the mineral for sodium-light.
$\theta_\alpha = 71^\circ 26'$	17.5° C.	1.7784 (at 17.6° C.)	$\left\{ \begin{array}{l} \alpha = 1.6859 \\ \beta = 1.6917 \end{array} \right.$
$\theta_\beta = 72^\circ 1\frac{4}{5}'$			
$\theta_\beta = 71^\circ 51\frac{3}{5}'$	14.5° C.	1.7802 (at 14.5° C.)	$\left\{ \begin{array}{l} \beta = 1.6917 \\ \gamma = 1.7105 \end{array} \right.$
$\theta_\gamma = 73^\circ 55\frac{2}{5}'$			

The edge of the shadows produced by total reflection was not very sharp, owing probably to the abundance of inclusions and the roughness of the face. The index of refraction of methylene iodide saturated with sulphur was determined before and after each of the main determinations by using a highly polished glass strip of which the index had been

<sup>1</sup> W. H. McNairn, Trans. R. Canadian Institute, 1917, vol. 11 (for 1916), pp. 243-251, and pls. 26, 27 [Min. Abstr., p. 170].

<sup>2</sup> A. Hutchinson, Mineralogical Magazine, 1911, vol. 16, p. 100.

accurately determined by S. Kōzu<sup>1</sup> and checked by myself before the experiment.

The strengths of birefringence of the mineral are  $\gamma - a = 0.0246$ ,  $\gamma - \beta = 0.0188$ ,  $\beta - a = 0.0058$ .

The measurement of the axial angle was made upon the same crystal cut perpendicular to the acute bisectrix. The same instrument arranged as a conoscope was employed for the determination. Five pairs of readings were taken from one side of the crystal plate and then the same number of readings were taken from the other side. The means obtained were  $2H_x = 59^\circ 33.6'$  at  $18.6^\circ \text{C.}$ , and  $59^\circ 50.6'$  at  $18.7^\circ \text{C.}$ , respectively, for sodium-light. The mean of these two values is  $59^\circ 42.1'$ . The refractive index of  $\alpha$ -monobromonaphthalene in which the crystal-plate was immersed was determined before and after the experiment and found  $\mu_D = 1.6587$  at  $18.4^\circ \text{C.}$

The true axial angle, as calculated from  $\mu$  of the liquid,  $\beta$  and  $H$  of the mineral, is  $2V = 58^\circ 25.3'$ , which agrees very closely with that computed from the principal indices, namely  $58^\circ 20.7'$ .

The determination of the position of the bisectrices was made on four sections cut parallel to the face  $b$ , by using a Bertrand ocular, and the results obtained are :

No. of Crystal.	$\acute{c} : \epsilon$ in the obtuse axial angle.	$\acute{c} : a$ in the acute axial angle.
1	41.4°	48.6°
2	42.0	48.0
3	41.7	48.3
4	41.3	48.7
mean	41.6	48.4

The extinction-angles measured on  $m$  (110) were  $31.0^\circ$  for  $\acute{c} : \epsilon$ , and  $59.0^\circ$  for  $\acute{c} : a$ .

Comparing these optical constants with those of augites from Auvergne, 'Renfrew', and Stromboli, the refractive indices are found to be lower than those of any augite previously investigated; and the optic axial angle is smaller than that of common augite and even than that of diopside, except the Stromboli augite which shows a particularly small axial angle.

These characters together with the small angles of extinction suggest that the mineral contains larger quantities of the diopside molecule than any common augite previously described.

<sup>1</sup> S. Kōzu, Mineralogical Magazine, 1916, vol. 17, p. 254.

	a.	$\beta$ .	$\gamma$ .	$\gamma$ -a.	2 V.
Auvergne <sup>1</sup> ...	1.706	1.712	1.728	0.022	—
„ ...	1.712	1.717	1.733	0.021	—
'Renfrew' <sup>2</sup> ...	1.6975	1.7039	1.7227	0.0252	61° 12'
Stromboli <sup>3</sup> ...	1.693	1.699	1.719	0.026	58 20.5
Nishigatake ...	1.6859	1.6917	1.7105	0.0246	58 25.3

*Chemical composition.*

To remove the glass inclusions, which the mineral contains in remarkable quantity, the coarsely powdered crystals were digested for several hours in hot, dilute hydrochloric acid. The powder was then washed in running water. The mineral fragments after this treatment, when examined under the microscope, are entirely clear and free from inclusions. If the washing was not complete, silica separated out as a white powder when the fragments were dried. However thoroughly the mineral was washed there would probably be some silica contained in the cavities which were formerly occupied by inclusions. This explains the excess of silica found by the analyses. Any magnetite which was left unattacked by the acid was removed with a bar-magnet.

The mineral powder thus prepared was olive-green in colour. The specific gravity was 3.388 at 4° C. The mineral fragments were then finely powdered in an agate mortar. The analyses were carried out by the usual methods as given by Washington<sup>4</sup> and the mean results obtained from two determinations which agreed closely are as follows:

	Percentage.	Molecular ratios.
SiO <sub>2</sub> ...	51.37	0.852
Al <sub>2</sub> O <sub>3</sub> ...	5.24	0.051
Fe <sub>2</sub> O <sub>3</sub> ...	2.02	0.013
FeO ...	2.96	0.041
CaO ...	21.58	0.385
MgO ...	16.94	0.420
TiO <sub>2</sub> ...	0.58	0.007
	100.69	

<sup>1</sup> A. Michel Lévy and A. Lacroix, *Les minéraux des roches*, 1888, p. 265.

<sup>2</sup> E. A. Wülfing, *Tschermaks Min. Petr. Mitt.*, 1896, vol. 15, pp. 43 and 47. The locality given as 'Renfrew, New Jersey, N.A.' is doubtful.

<sup>3</sup> S. Kōzu and H. S. Washington, *Amer. Journ. Sci.*, 1918, vol. 45, p. 463.

<sup>4</sup> H. S. Washington, *Manual of the chemical analysis of rocks*, 1904.

From these numbers the following percentage constitution is computed:

			Molecular percentage.		Weight percentage.
Diopside,	$\text{CaMgSi}_2\text{O}_6$	...	78.2	...	74.6
Hedenbergite,	$\text{CaFeSi}_2\text{O}_6$	...	9.4	...	10.3
Tschermak's salt,	$\text{MgAl}_2(\text{Si,Ti})\text{O}_6$	...	8.8	...	10.5
—	$\text{MgFe}_2(\text{Si,Ti})\text{O}_6$	...	2.1	...	3.3
Enstatite,	$\text{MgSiO}_3$	...	1.5	...	1.3
			<hr/>		<hr/>
			100.0		100.0
	$\text{SiO}_2$ (free)	...	0.7	...	0.8

The mineral consists therefore very largely of the diopside molecules accompanied by fair amounts of hedenbergite and Tschermak's salt, and a little  $\text{MgFe}_2(\text{Si,Ti})\text{O}_6$  and enstatite.

If we compare the composition of the Nishigatake augite with that of the Stromboli augite,<sup>1</sup> the essential differences are that the former shows slightly higher alumina, lower ferrous oxide, and higher magnesia, the ratio  $\text{MgO}:\text{FeO}$  being 10.2:1 in the former, while it is 3.5:1 in the latter. H. S. Washington<sup>2</sup> has recently published analyses of augites from Vesuvius and Etna. The Etna augite differs from that of Nishigatake in possessing more lime and titanium dioxide and less magnesia, as shown in the following table:

		$\text{SiO}_2$	$\text{Al}_2\text{O}_3$	$\text{Fe}_2\text{O}_3$	$\text{FeO}$	$\text{MnO}$	$\text{CaO}$
•Stromboli ...		50.94	3.37	2.05	7.41	0.10	20.34
Etna ...		50.09	3.71	1.47	4.96	0.21	22.48
$\text{MgO}$	$\text{SrO}$	$\text{Na}_2\text{O}$	$\text{K}_2\text{O}$	$\text{H}_2\text{O}+$	$\text{TiO}_2$	Total.	
14.59	0.08	0.61	0.18	0.08	0.96	100.71	} (Washington).
14.01	n.d.	0.78	0.01	0.22	2.11	100.00	

In conclusion, the essential character of the Nishigatake augite is its high content in magnesia, whence we conclude that it contains a large number of diopside molecules.

<sup>1</sup> S. Kōzu and H. S. Washington, Amer. Journ. Sci., 1918, vol. 45, p. 463.

<sup>2</sup> H. S. Washington and H. E. Merwin, Amer. Journ. Sci., 1921, vol. 1, pp. 20-30 [Min. Abstr., p. 161].